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Conceptual Designs for a Long Term $^{238}\text{PuO}_2$ Storage Vessel

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Conceptual Designs For A Long Term $^{238}\text{PuO}_2$ Storage Vessel*

by

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Abstract

This is a report on conceptual designs for a long term, 250 years, storage container for plutonium oxide ($^{238}\text{PuO}_2$). These conceptual designs are based on the use of a quartz filter to release the helium generated during the plutonium decay. In this report a review of filter material selection, design concepts, thermal modeling, and filter performance are discussed.

* Prepared for
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Executive Summary

Sandia National Laboratories investigated design options for a long term plutonium oxide ($^{238}\text{PuO}_2$) storage container at the request of Los Alamos National Laboratory. The investigation was conducted over a two month period. The primary design goals for the plutonium oxide container were to have a useful life of 250 years and to contain 300 grams of $^{238}\text{PuO}_2$. In the course of identifying conceptual designs Sandia investigated numerous materials for helium filtration, down-selected design options, developed thermal models, calculated internal pressures of the design concepts as a function of time, and determined the pressure limits of the designs.

After considering ten types of glasses, vitreous fused silica (v-SiO₂) was selected as the filter material. One of the most important characteristics of the optimum filtering material is its permeability to helium. Fused silica was selected because it had the highest helium permeability, the lowest activation energy, and superior fabrication characteristics when compared to other glass candidates.

The design concepts were down-selected based on cost, manufacturability, working pressure, and impact on transportability. Several design concepts that have a large surface area in a small volume were discounted early in the project because they relied on difficult and expensive manufacturing processes like ultrasonic machining. The design options selected were tubular designs and a convex disk design. All the designs are inexpensive and manufacturable, have high safety factors, and fit within the design envelope of the existing shipping container.

The design concepts were developed iteratively with thermal analysis. The temperature of the storage vessel has a dramatic impact on the effectiveness of the quartz filter and on the handling of the vessel. As concepts were developed thermal analysis was performed and the effect on helium permeation as well as the outer wall temperature was calculated. The selected designs have a filter wall temperature of approximately 280 °C and outer wall temperatures of approximately 100 °C.

The partial pressure of helium was calculated based on the difference between the generation rate from the plutonium decay and the permeation rate through the filters. The design parameters affecting permeation were: the quantity of plutonium in the vessel, the temperature of the vessel, the surface area of the quartz, and the thickness of the quartz. Since the designs used the same type of quartz and contained the same quantity of plutonium, the variables affecting the internal pressure of the design concepts were the surface area and the thickness of the quartz.

The internal pressure of the vessels was limited by the strength of the quartz. The entire storage vessel is made of stainless steel except for the filter. Since quartz is a brittle, relatively low strength material compared to stainless steel it is the limiting material relative to the working pressure of the vessel. In developing conceptual designs peak stresses in the quartz were calculated using thin walled cylinder and disk formulas. Based on these calculations filter stresses from the partial pressure of helium are many orders of magnitude below the theoretical failure level for the designs presented.

1.0 Introduction

Long term storage of plutonium-238 is a necessity because of the potential loss of production capability. The potential decrease in NASA and defense funding for $^{238}\text{PuO}_2$ heat source production, as well as reconfiguration of the DOE production complex, present a need for a long term $^{238}\text{PuO}_2$ storage capability¹.

The key to long term storage of plutonium-238 is relieving the gas pressure created by the decay products. The helium generated during the decay of plutonium-238 must be vented from the storage vessel to avoid over-pressurization. This report describes design concepts developed at Sandia National Laboratories to support Los Alamos National Laboratory in designing long term storage vessels that incorporate helium filters.

This report summarizes design specifications, filter material selection, conceptual designs, thermal modeling, permeation calculations, and preliminary stress calculations.

2.0 Design Specifications

The following design specifications were established at the beginning of the project.

- Design a vessel that will extend the $^{238}\text{PuO}_2$ storage life up to 250 years
- Investigate incorporation of a quartz helium filter to relieve container pressure build-up for long term storage of $^{238}\text{PuO}_2$
- Accommodate 300 grams of $^{238}\text{PuO}_2$ powder at 2.5 grams/cc, or two 1 inch diameter, 150 gram pellets
- Minimize outer wall temperature, desired temperature is 40° C
- Minimize cost
- Maximize manufacturability
- Design for use in a glove box
- Design as a storage vessel, compatibility with existing shipping container is desirable but not required
- Venting of helium to the room is acceptable
- Plutonium powder release to the room is not acceptable
- Identify cooling methods, water bath is acceptable but not desirable
- Minimize diffusion of other gasses
- Must address radiation hazard and shielding issues

The scope of the project included:

- Conceptual design
- Conceptual drawings
- Supporting calculations

The scope of the project did not include:

- Usable hardware
- Extensive theoretical models
- A fabrication drawing set

2.1 Baseline Concept

The existing container design is a dual containment system. The inner container that holds the plutonium-238 is called EP-60, and the outer container is called EP-61. The assembly is referred to as the "Flashlight" because its geometry resembles a large flashlight.

The Specifications for the Flashlight are summarized below.

Volume = 300 cm³

Plutonium Mass = 300 grams

Pressure = 400 - 500 psi

Temperature = 200 °C

Cost ≈ \$500-600/ container (EP61)

Pu 238, 1/2 life = 86.4 yr.

Cooling methods - in water

Pu 238 oxide powder - currently stored at SR & at LANL

300 cc He4 @ 3 atm in 10 yr. (300 g Pu238 O2, 600 cm3, 50% free)

LANL receives 10 yr. Old SR standards

Tamb ≈ 68 - 70 °F

3.0 Filter Material Selection

The theory of absolute reaction rates formulated by Glasstone, Laidler, and Eyring¹¹ predicts the temperature dependence of diffusion of monatomic gas such as helium in glass by the equation (3.1):

$$D = D_0 T \exp\left(\frac{-Q_D}{RT}\right)$$

where D = diffusivity, cm²/sec
 D_0 = pre-exponential factor, cm²/sec K
 T = absolute temperature, K
 Q_D = activation energy, kcal/mole
 R = gas constant. (Eq. 3.1)

Doremus² confirmed equation (3.1) by plotting the experimental data of helium permeation through vitreous silica as $\log(D/T)$ versus reciprocal temperature and yielded a straight line.

The temperature dependence of permeation of monatomic gas (helium) in glass (vitreous silica) also obeys a similar modified Arrhenius Equation (3.2):

$$K = K_0 T \exp\left(\frac{-Q_K}{RT}\right)$$

where K = permeability, atom/s cm atm
 K_0 = pre-exponential factor, atom/s cm atm K
 T = absolute temperature, K
 Q_K = activation energy, kcal/mole
 R = gas constant. (Eq. 3.2)

This relationship (Eq. 3.2) was confirmed by Swets, et al.,³ Shelby⁴, and Perkins and Begeal⁵ experimentally. Figure 3.1 shows permeability of helium through the silica glass, plotted according to equation (3.2). Open circles represent data obtained by Swets, et al., for helium permeation through cylindrical membranes of wall thickness 1 and 1.5-mm, triangles represent data obtained by Shelby for 1.6-mm thick disk of CGW 7940 vitreous silica, and solid circles represent data obtained by Perkins and Begeal for 3 to 5- μ -thick silicon oxide films deposited on porous Vycor substrates.

Figure 3.1: Permeability of Helium Through Silica Glass
Data taken from: o - Swets, Lee, and Frank; Δ - Shelby, • - Perkins and Begeal

Shelby⁶ states that the permeability and diffusivity of gases in glass are strong functions of the atomic structure of the glass and of the morphology of the sample. As a result, the mobility of helium in a given sample is a function of both the glass composition and its thermal history. In single phase samples, the diffusivity of helium can decrease by as much as a factor of two upon annealing of a highly quenched sample. However, if a change in thermal history affects the morphology of the sample, the mobility of helium has been found to vary by as much as two orders of magnitude in the same sample.

Shelby also states that studies of a wide variety of systems have shown that the diffusing gas will tend to migrate along paths through the higher diffusivity, lower activation energy phase.

Figure 3.2 shows the permeation rates for helium in various glasses as a function of temperature, measured by Norton.⁷ This figure shows the effect of composition on molecular transport in glass. As more network modifying ions are added, the permeation rate decreases and the activation energy increases. Therefore, pure vitreous fused silica (v- SiO_2) which contains the least impurities has the highest permeability (See Figure 3.2). The addition of impurities decrease the permeability to helium since helium moves through the interstitial gaps in the lattice structure. Impurities act to fill these spaces, thereby decreasing the mobility of helium.

Figure 3.2: Permeation rates for helium in various glasses as a function of temperature. Permeation units are cm^3 of gas at STP per sec per cm^2 area per mm thickness per cm Hg pressure.

The following tables, Table 3.1 and Table 3.2, show the diffusivity and the permeability values of helium in different glasses. The glass that gives the best diffusivity and permeability characteristics is again the pure vitreous fused silica (99.9% SiO₂). It has the lowest activation energy.

Table 3.1: Diffusivity Values of Helium in Glass

D ₀ (10 ⁻⁷ cm ² /(s K))	E _D (kcal/mol)	Temp (°C)	Glass (Reference)
2.97	4.83	25-500	Optical quality porous GCW 7930 substrate - same properties as bulk fused silica (Perkins)
3.38 or 3.31	4.95	286-602	Spectrasil A; Na 3 ppm, OH 1280 ppm (Shelby, 1971)
3.241 3.013	4.90 4.84	286-602	Infrasil; Al 64 ppm, Fe 11 ppm, Na 21 ppm, OH 11 ppm (Shelby, 1971)
4.4	4.8	103-165	SiO ₂ (Shelby, 1972)
3.11 12.8	4.91	102-701	Vitreosil-IR, v-SiO ₂ , OH 10 ppm (Shelby, 1973)
3.5	4.71	103-165	99.9 % SiO ₂ (Shelby, 1973)
3.4	5.7	25-544	CGW-7740 (Shelby, 1974)
16.2 18.2	5.000 5.111	66-140 59-154	99.9 % B ₂ O ₃ (Shelby, 1973)
16.7 10.9	7.5 6.9	107-174 110-176	GeO ₂ (Shelby, 1973)

Table 3.2: Permeability Values of Helium in Glass

K ₀ (10 ¹⁰ atoms/s cm atm K)	E _K (kcal/mol)	Temp (°C)	Glass (Reference)
20.6	4.302	25-500	Optical quality porous GCW 7930 substrate - same properties as bulk fused silica (Perkins)
6.3 7.0	4.1 1.43	90-171 103-165	99.9 % SiO ₂ (Shelby, 1973)
6.4	5.2	25-544	CGW-7710 (Shelby, 1974)
0.0017	6.5		75% SiO ₂ , 12.7% B ₂ O ₃ , 4.6% Na, 3.4% PbO, 2.4% A ₂ O ₃ , 1.8% U ₂ O ₃ (Walters)
14.4 12.8	3.97 4.03	66-140 59-154	99.9 % B ₂ O ₃ * (Shelby, 1973)
3.8 4.7	5.7 5.82	107-174 110-176	GeO ₂ (Shelby, 1972)

* Values reported are annealed values. Annealing (192 °C) decreased diffusion and permeability values up to 30% from the quenched values.

After conducting an extensive literature search and reviewing many research reports on helium permeation on different glasses, vitreous fused silica (v- SiO_2) was chosen as the filter material for the long term storage container design. The following list shows the rationale for choosing v- SiO_2 :

- Fused silica has the lowest activation energy (See Tables 3.1 & 3.2).
- Fused silica has a very high helium permeability (See Figure 3.2).
- Viscosity and coefficient of expansion characteristics, important to filter fabrication, were superior for fused silica over other glass candidates.

4.0 Design Concepts

Three design concepts were selected for conceptual development. Specialized filters were eliminated as potential designs based on manufacturability and cost. The three design concepts suggested here are inexpensive, manufacturable, complimentary to the existing storage and shipping containers, and low in stress. All of the designs discussed are in their simplest form. Combinations of the concepts as well as innovations can accommodate different design criteria. Only simple baseline concepts are presented here as governed by the planned scope of the project.

The first concept is a modified EP-61 end-cap. The intent was to utilize the large diameter of the EP-61 to accommodate a disk shaped filter. By incorporating the filter into the EP-61 endcap the existing EP-60 and EP-61 components become the storage vessels. This minimizes vessel development, qualification, and fabrication costs. Since the EP-61 is utilized currently, new transportation containers are not required. The radiation shielding methods are not addressed by this design.

Figure 4.1. EP-61 filtered endcap concept.

The EP-61 filtered endcap concept shown in Figure 4.1 has a convex quartz filter. This geometry keeps the quartz in compression. This minimizes potential failure from imperfections in the quartz and utilizes the high compressive strength of the quartz, assuming the supporting features keep the filter in compression. (The compressive strength for quartz is 160,000 psi compared to a tensile strength of only 7000 psi¹².)

Utilizing the EP-61 Concept requires modifying the EP-60 endcap. By drilling a hole in the endcap of the EP-60 the helium can vent to the new EP-61 filtered endcap. Inserting a stainless steel filter in the hole will ensure plutonium containment in the event of a filter failure.

The second concept is a modified EP-60 endcap. A simple disk shaped filter will not work with the EP-60 because of its small diameter. Surface area is critical to helium permeation and the area of a disk in the EP-60 endcap would result in prohibitively high pressures. A simple tubular geometry was chosen because it has a large surface area, performs well in compression, and costs less to fabricate than a complex geometry.

Figure 4.2 and Figure 4.3 show the EP-60 filtered endcap concept. This design has the same advantages as the EP-61 filtered endcap but has a greater surface area, lowering the internal pressure. The internal pressure histories are discussed in Section 6 of this report.

The geometry also lends itself to a graded glass to metal seal. Glass to metal seals may use several types of glasses and metals to transition from the quartz to the steel. Manufacturing the seal is easier with a long cylindrical geometry than a disk or more complex geometry.

This design requires separation of the quartz from the plutonium. Housing the quartz in a sintered metal sleeve, or coating the quartz with a metal will avoid any potential degrading effects the plutonium might have on the quartz.

Figure 4.2. The EP-60 filtered endcap concept.

Figure 4.3. The EP-60 filtered endcap in the EP-60.

The third concept is a quartz containment vessel. The plutonium is stored inside the filter in this design. The purpose for storing the plutonium inside the filter is to maximize the filter area and minimizing the internal pressure. A model of this concept is shown in figure 4.4.

Figure 4.4. The EP-3 plutonium storage concept.

As mentioned for the EP-60 filtered endcap a separation material between the quartz and plutonium is required. The separation material will also keep the quartz out of the plutonium in the event of a filter failure.

The steel cylinder surrounding the filter acts as a container in the event that the filter fractures. A stainless steel sintered metal filter is staked in the cylinder to allow helium to vent to the atmosphere and will contain glass and plutonium in a filter failure scenario.

This concept remains within the EP-61 geometry envelope allowing the use of existing transportation containers. For transportation an EP-61 sleeve is welded to the EP-3 body. Figure 4.5 illustrates the EP-3 and EP-61 sub-assembly. The welded assembly results in a hermetically sealed vessel with the same external dimensions as the existing EP-61.

Figure 4.5. EP-3 assembly to an EP-61 sub-assembly.

A bolt together version of the EP-3 was also designed. The driver for this design was cost. The design, shown in Figure 4.6, uses standard vacuum flanges and a metal gasket for sealing. The advantages of this design is easy access to the plutonium and low manufacturing costs. A disadvantage of this design is that it is not hermetically sealed, making shipping difficult.

Figure 4.6. EP-3 bolt-together hardware.

This design accommodates powder or pellets and has a neutron shield. The 1.3 inch filter diameter of the EP-3 concept will house powder or 1 inch diameter pellets. The outermost wall is 3 inches in diameter and is made of NORYL 5104. This is a polymer that will act as a neutron shield and an insulator. This material is resistant to radiation degradation. The insulating properties will increase the helium permeation and minimize the outer wall temperature for handling.

The EP-3 design can be used as a welded assembly or as a bolt-together assembly. The bolt-together assembly relies on a metal gasket for sealing the helium in the EP-3. Since helium is difficult to contain the integrity of the seal design needed testing. A set of blank flanges were used to investigate the sealing integrity of the metal gasket. The hardware shown in Figure 4.7 had no detectable leak when pressurized with 250 psi of helium. The test was performed in a bell jar calibrated to detect leaks as small as $1 \times 10^{-6} \text{ cm}^3/\text{sec}$ at STP (standard temperature and pressure).

Figure 4.7. Leak test fixture.

5.0 Thermal Modeling

Thermal modeling and analysis described in this section is for the EP-3 Plutonium Storage Design Concept shown in Figure 4.4. Analysis for the other design concepts described in section 4.0 was not required since the other designs incorporated filters directly into the existing EP endcaps and Los Alamos has already analyzed that design.

The $^{238}\text{PuO}_2$ container design utilizes a passive heat transfer system. Heat from the radioactive decay (0.569 W/gram for $^{238}\text{PuO}_2$) is conducted from the oxide powder to the side wall of the quartz glass container. The heat is then transported radially by radiation and conduction to the stainless steel inner container. From the stainless steel inner container the heat is transported by radiation and conduction to the NORYL 5104 outer container. Heat conducted through the NORYL 5104 is then dissipated to the outside ambient air by a natural convection heat transfer process.

The thermal model of the $^{238}\text{PuO}_2$ container is modeled as a set of concentric cylinders. A schematic of the container design model is shown in Figure 5.1. Heat transfer out the ends of the container was not considered for this thermal evaluation.

Figure 5.1: EP-3 Plutonium Storage Container Design Model - A set of Concentric Cylinders

Proceeding from inside out the following annular layers are found:

- oxide powder
- quartz glass container
- air gap
- stainless steel inner container
- air gap
- NORYL 5104 outer container
- outside ambient air

Heat transfer occurs by conduction only through the container materials and by both conduction and thermal radiation through the air gaps. Heat rejection from the NORYL 5104 outer surface to the ambient air is by natural convection.

To evaluate the heat transfer process, three modes of heat transfer were considered. They were: conduction heat transfer, convection heat transfer, and thermal radiation heat transfer. The following section describes each of the heat transfer processes.

Conduction Heat Transfer

The rate of heat removal by conduction through a stationary medium (container materials) is:

$$Q_K = \frac{KA}{Th}(T_i - T_o) = \frac{(T_i - T_o)}{R_K}$$

$$\text{where } R_K = \frac{Th}{KA}$$

Th = material thickness

K = thermal conductivity of the material

A = heat transfer area

$T_i - T_o$ = temperature difference between inner and outer walls

Thermal Radiation Heat Transfer

The rate of heat removal by thermal radiation is:

$$Q_R = \frac{\sigma A_i F_{io} (T_i^4 - T_o^4)}{\frac{1}{\epsilon_i} + \left(\frac{A_i}{A_o} \right) \left(\frac{1}{\epsilon_o} - 1 \right)} = H_R A_i (T_i - T_o) = \frac{(T_i - T_o)}{R_R}$$

$$\text{where } R_R = \frac{1}{H_R A_i}$$

$$H_R = \frac{\sigma F_{io} (T_i^2 + T_o^2)(T_i + T_o)}{\frac{1}{\epsilon_i} + \left(\frac{A_i}{A_o} \right) \left(\frac{1}{\epsilon_o} - 1 \right)}$$

σ = Stefan-Boltzman constant = 5.67×10^{-8}
 F_{io} = geometric view factor
 T_i = inner cylinder temperature
 T_o = outer cylinder temperature
 A_i = inner cylinder heat transfer area
 A_o = outer cylinder heat transfer area
 ϵ_i = emissivity of inner surface
 ϵ_o = emissivity of outer surface

Convection Heat Transfer

The rate of heat removal by natural convection is:

$$Q_c = H_c A (T_s - T_\infty) = \frac{(T_s - T_\infty)}{R_c}$$

where $R_c = \frac{1}{H_c A}$

$$H_c = \frac{K Nu}{d} = \frac{K [0.53 (Gr Pr)^{0.25}]}{d}$$

$$Gr = \frac{g \beta (T_s - T_\infty) d^{1/3}}{\nu^2}$$

H_c = convective heat transfer coefficient

A = convective heat transfer area

Nu = Nusselt number

d = diameter

K = thermal conductivity

Gr = Grashof number

Pr = Prandtl number

g = acceleration due to gravity

β = temperature coefficient of volume expansion

ν = kinematic viscosity

To compute the steady state temperature distribution, a thermal circuit analog model was developed. This model along with the calculated thermal resistance values across each node allowed the utilization of a commercially available PSpice circuit analysis program to determine temperature profiles at each node of the circuit. The thermal circuit analog model is shown in Figure 5.2.

Figure 5.2: Thermal Circuit Analog Model of the $^{238}\text{PuO}_2$ Container Design

Using the PSpice program the following temperatures could be computed:

- node 1 - quartz glass inner wall (T_{qi})
- node 2 - quartz glass outer wall (T_{qo})
- node 3 - stainless steel container inner wall (T_{si})
- node 4 - stainless steel container outer wall (T_{so})
- node 5 - NORYL 5104 container inner wall (T_{pi})
- node 6 - NORYL 5104 container outer wall (T_{po})

The thermal resistance values used in the circuit analog model were calculated in the following way (proceeding from left to right in Figure 5.2):

²³⁸PuO₂ heat source

Heat from radioactive decay of ²³⁸Pu

$$Q_{rd} = 0.569 \text{ W/gram of } ^{238}\text{Pu}$$

Mass of Pu per container

$$M_{pu} = 300 \text{ gram}$$

Heat generated from ²³⁸Pu powder

$$Q_{pu} = 0.569 \text{ W/gram} * 300 \text{ gram} = 170.7 \text{ W}$$

R/k,glass

Conduction through quartz glass

$$R/k, \text{glass} = T_h/k, \text{glass} / (K/k, \text{glass} A/k, \text{glass})$$

where $T_h/k, \text{glass}$ = quartz glass thickness

$K/k, \text{glass}$ = thermal conductivity of glass

$A/k, \text{glass}$ = heat transfer area

R/rad,air1

Thermal radiation in the air gap between quartz and stainless steel containers

$$R/\text{rad,air1} = 1 / (H/r, \text{air1} A/r, \text{air1})$$

$$\text{where } H_{r, \text{air1}} = \frac{\sigma F_{qo-si} (T_{qo}^4 + T_{si}^4) (T_{qo} + T_{si})}{\frac{1}{\epsilon_{qo}} + \left(\frac{A_{qo}}{A_{si}} \right) \left(\frac{1}{\epsilon_{si}} - 1 \right)}$$

$A/r, \text{air1}$ = heat transfer area

R/k,air1

Conduction through air gap between quartz and stainless steel containers

$$R/k, \text{air1} = T_h/k, \text{air1} / (K/k, \text{air1} A/k, \text{air1})$$

where $T_h/k, \text{air1}$ = thickness of air gap 1

$K/k, \text{air1}$ = thermal conductivity of air gap 1

$A/k, \text{air1}$ = heat transfer area

R/k,ss

Conduction through stainless steel container

$$R/k, \text{ss} = T_h/k, \text{ss} / (K/k, \text{ss} A/k, \text{ss})$$

where $T_h/k, \text{ss}$ = stainless steel container thickness

$K/k, \text{ss}$ = thermal conductivity of stainless steel container

$A/k, \text{ss}$ = heat transfer area

R/rad,air2

Thermal radiation in the air gap between stainless steel and NORYL 5104 containers

$$R/\text{rad,air2} = 1 / (H/\text{r,air2} \ A/\text{r,air2})$$

$$\text{where } H_{\text{r,air2}} = \frac{\sigma F_{\text{so-pi}} (T_{\text{so}}^4 + T_{\text{pi}}^4) (T_{\text{so}} + T_{\text{pi}})}{\frac{1}{\epsilon_{\text{so}}} + \left(\frac{A_{\text{so}}}{A_{\text{pi}}} \right) \left(\frac{1}{\epsilon_{\text{pi}}} - 1 \right)}$$

A/r,air2 = heat transfer area

R/k,air2

Conduction through air gap between stainless steel and NORYL 5104 containers

$$R/\text{k,air2} = \text{Th/k,air2} / (K/\text{k,air2} \ A/\text{k,air2})$$

where Th/k,air2 = thickness of air gap 2

K/k,air2 = thermal conductivity of air gap 2

A/k,air2 = heat transfer area

R/k,plstc

Conduction through NORYL 5104 container

$$R/\text{k,p} = \text{Th/k,p} / (K/\text{k,p} \ A/\text{k,p})$$

where Th/k,p = NORYL 5104 container thickness

K/k,p = thermal conductivity of NORYL 5104 container

A/k,p = heat transfer area

R/hc

Natural convection from NORYL 5104 outer surface to the ambient air

$$R/\text{hc} = 1 / (H_c \ A)$$

$$\text{where } H_c = \frac{K \text{Nu}}{d} = \frac{K [0.53 (Gr \text{Pr})^{0.25}]}{d}$$

$$Gr = \frac{g \beta (T_s - T_\infty) d^{1/3}}{\nu^2}$$

A = heat transfer area

The $^{238}\text{PuO}_2$ container design features are listed in Table 5.1.

Table 5.1: $^{238}\text{PuO}_2$ Container Design Features

Container	ID (inches)	OD (inches)	Length (inches)	K (W/m K)	Purpose
Quartz Glass Container	1.312 5	1.5125	6.0	1.381	• helium permeation material
Stainless Steel Inner Container	1.76	2.0	7.5	1.63	• containment vessel for glass breakage • metal frit at one end for He permeation
NORYL 5104 Outer Container	2.75	3.5	8.0	0.186	• neutron shielding material • insulator - eliminates rad. heat transfer • max. temp = 145°C • \$2.20 per pound

The thermal properties of the container materials and other significant values used in the thermal resistance calculations are shown in Table 5.2.

Table 5.2: Thermal Properties of Air¹⁰

Temp	Temp	Temp	Density	Thermal Conductivity	Specific Heat	Thermal Diffusivity $\alpha \times 10^6$	Kinematic Viscosity $\nu \times 10^6$	Prandtl Number
(F)	(K)	(C)	(kg/m ³)	(W m K)	(J/kg K)	(m ² /s)	(m ² /s)	
32	273	0	1.252	0.0237	1011	19.2	13.9	0.71
68	293	20	1.164	0.0251	1012	22.0	15.7	0.71
104	313	40	1.092	0.0265	1014	24.8	17.6	0.71
140	333	60	1.025	0.0279	1017	27.6	19.4	0.71
176	353	80	0.968	0.0293	1019	30.6	21.5	0.71
212	373	100	0.916	0.0307	1022	33.6	23.6	0.71
392	473	200	0.723	0.0370	1035	49.7	35.5	0.71
572	573	300	0.596	0.0429	1047	68.9	49.2	0.71
752	673	400	0.508	0.0485	1059	89.4	64.6	0.72
932	773	500	0.442	0.0540	1076	113.2	81.0	0.72
1832	1273	1000	0.268	0.0762	1139	240.0	181.0	0.74

The following section describes the thermal evaluation results. Five different cases that were used to study the thermal effects are shown in Tables 5.4 through 5.8.

Table 5.3: Table for the Acronyms Used

T _{po}	NORYL 5104 container outer wall temperature
T _{pi}	NORYL 5104 container inner wall temperature
T _{so}	stainless steel container outer wall temperature
T _{si}	stainless steel container inner wall temperature
T _{qo}	quartz glass outer wall temperature
T _{qi}	quartz glass inner wall temperature
T _{amb}	ambient air temperature
H _c	convective heat transfer coefficient from T _{po} to T _{amb} (environment)
D _{po}	NORYL 5104 container outer diameter

In thermal model #1, the convective heat transfer coefficient (H_c) was first calculated using the assumptions that the ambient temperature was maintained at 20 °C and the desired temperature for the outermost skin of the container (T_{po}) was 40 °C. The calculated convective heat transfer coefficient was 5.3 W/m² which occurred in the natural convection region¹⁰. However, when the value of 5.3 W/m² was used to calculate the actual skin temperature of the container, it turned out to a much higher value of 502 °C. This demonstrated that a higher convective coefficient should be used in order to reduce the skin temperature.

Table 5.4: Results of Thermal Model #1

Assume:	T _{po} /desired = 40 °C T _{amb} = 20 °C D _{po} = 3.5 in
Calculate:	T _{po} ; T _{pi} ; T _{so} ; T _{si} ; T _{qo} ; T _{qi} ; and H _c
Results:	H _c = 5.3 W/ m ² K T _{po} = 775.0 K = 502.0 °C T _{pi} = 944.1 K = 671.1 °C T _{so} = 944.2 K = 671.2 °C T _{si} = 945.2 K = 672.2 °C T _{qo} = 945.3 K = 672.3 °C T _{qi} = 950.0 K = 677.0 °C
Remark:	T _{po} = 502 °C is too hot; Need to increase H _c

In thermal model #2, the highest natural convective heat transfer coefficient ($H_c = 30 \text{ W/m}^2$) listed in reference [10] was used to generate the container temperature profiles. This model predicted a much lower outer skin temperature than the previous model (from 502°C to 107°C). However, this values of 107°C was still higher than the desired 40°C temperature. From this model it was concluded that in order to achieve a skin temperature of 40°C , one cannot rely entirely on the natural convection process, but must consider a forced convection process to cool the containers.

Table 5.5: Results of Thermal Model #2

Assume:	$T_{po}/\text{desired} = 40^\circ\text{C}$ $T_{amb} = 20^\circ\text{C}$ $D_{po} = 3.5 \text{ in}$ $H_c = 30 \text{ W/m}^2 \text{ K}$ • highest value listed in ref. (Kreith) for natural convection
Calculate:	$T_{po}; T_{pi}; T_{so}; T_{si}; T_{qo}; T_{qi}$
Results:	$T_{po} = 380.0 \text{ K} = 107.0^\circ\text{C}$ $T_{pi} = 533.2 \text{ K} = 260.2^\circ\text{C}$ $T_{so} = 533.6 \text{ K} = 260.6^\circ\text{C}$ $T_{si} = 534.5 \text{ K} = 261.5^\circ\text{C}$ $T_{qo} = 534.7 \text{ K} = 261.7^\circ\text{C}$ $T_{qi} = 550.0 \text{ K} = 277.0^\circ\text{C}$
Remark:	$T_{po} = 107^\circ\text{C}$ is still too hot; Need to increase H_c to reduce T_{po} ; $T_{po}/\text{desired} = 40^\circ\text{C}$ will not be reached by natural convection process;

In thermal model #3, a forced convection process was investigated. Since the previous model (model #2) demonstrated that the desired skin temperature of 40 °C was not achievable using only the natural convection process, an investigation on which convective heat transfer coefficient value would provide the desired skin temperature was conducted. This analysis resulted in a convective heat transfer coefficient which occurred in the forced convection region ($H_c = 118 \text{ W/m}^2\text{K}$).

Table 5.6: Results of Thermal Model #3

Assume:	$T_{po}/\text{desired} = 40 \text{ }^\circ\text{C}$ $T_{amb} = 20 \text{ }^\circ\text{C}$ $D_{po} = 3.5 \text{ in}$ $H_c > 30 \text{ W/ m}^2 \text{ K}$	
Calculate:	$T_{po}; T_{pi}; T_{so}; T_{si}; T_{qo}; T_{qi};$ and H_c required to get $T_{po}/\text{desired} = 40 \text{ }^\circ\text{C}$	
Results:	$T_{po} = 313.0 \text{ K} = 40 \text{ }^\circ\text{C}$ $T_{pi} = 467.1 \text{ K} = 194.1 \text{ }^\circ\text{C}$ $T_{so} = 467.8 \text{ K} = 194.8 \text{ }^\circ\text{C}$ $T_{si} = 468.8 \text{ K} = 195.8 \text{ }^\circ\text{C}$ $T_{qo} = 469.0 \text{ K} = 196.0 \text{ }^\circ\text{C}$ $T_{qi} = 485.0 \text{ K} = 212.0 \text{ }^\circ\text{C}$ $H_c = 118 \text{ W/m}^2 \text{ K}$	
Remark:	$T_{po} = 40 \text{ }^\circ\text{C} = T_{po}/\text{desired}$; this meets the requirement. $H_c = 118 \text{ W/ m}^2 \text{ K}$ is outside the natural (free) convection range; Therefore, a forced air convection is required to keep $T_{po} = 40 \text{ }^\circ\text{C}$. (Ref.: Kreith, p20 Free Air Convection: 6 - 30 $\text{W/m}^2 \text{ K}$ Forced Air Convection: 30 - 300 $\text{W/m}^2 \text{ K}$)	

In thermal model #4, an investigation on the effect of doubling the wall thickness of the NORYL 5104 was conducted. This study assumed a passive cooling process (within the natural convection range using $H_c = 30 \text{ W/m}^2$). The results of the study showed that doubling the wall thickness of the outer skin would reduce the temperature down to 93°C , but this was still hotter than the desired temperature.

Table 5.7: Results of Thermal Model #4

Assume:	$T_{po}/\text{desired} = 40^\circ\text{C}$ $T_{amb} = 20^\circ\text{C}$ $D_{po} = 3.5 \text{ in}$ $H_c = 30 \text{ W/m}^2 \text{ K}$ (Stay within natural convection range) Double the NORYL 5104 wall thickness; $D_{po} = 4.25''$	
Calculate:	$T_{po}; T_{pi}; T_{so}; T_{si}; T_{qo}; T_{qi}$	
Results:	$T_{po} = 366.0 \text{ K} = 93.0^\circ\text{C}$ $T_{pi} = 636.3 \text{ K} = 363.3^\circ\text{C}$ $T_{so} = 636.6 \text{ K} = 363.6^\circ\text{C}$ $T_{si} = 637.6 \text{ K} = 364.6^\circ\text{C}$ $T_{qo} = 637.7 \text{ K} = 364.7^\circ\text{C}$ $T_{qi} = 653.0 \text{ K} = 380.0^\circ\text{C}$	
Remark:	$T_{po} = 93^\circ\text{C} > T_{po}/\text{desired};$ Doubling the wall thickness of the NORYL 5104 container reduced T_{po} by 14°C ; but T_{po} is still too hot.	

In thermal model #5, an investigation on the effect of doubling the lengths of all the containers was conducted. This study also assumed that the cooling process was passive heat transfer (within the natural convection range using $H_c = 30 \text{ W/m}^2$). The results of this study demonstrated that the doubling of the container lengths reduced the skin temperature by 44°C (from 107°C to 63°C), but this was still higher than the desired 40°C .

Table 5.8: Results of Thermal Model #5

Assume:	$T_{po}/\text{desired} = 40^\circ\text{C}$ $T_{amb} = 20^\circ\text{C}$ $D_{po} = 3.5 \text{ in}$ $H_c = 30 \text{ W/m}^2 \text{ K}$ (Stay within natural convection range) Double the lengths of all containers.	
Calculate:	$T_{po}; T_{pi}; T_{so}; T_{si}; T_{qo}; T_{qi};$ and	
Results:	$T_{po} = 336.0 \text{ K} = 63.0^\circ\text{C}$ $T_{pi} = 413.0 \text{ K} = 140.0^\circ\text{C}$ $T_{so} = 414.7 \text{ K} = 141.7^\circ\text{C}$ $T_{si} = 415.2 \text{ K} = 142.2^\circ\text{C}$ $T_{qo} = 416.0 \text{ K} = 143.0^\circ\text{C}$ $T_{qi} = 424.0 \text{ K} = 151.0^\circ\text{C}$	
Remark:	$T_{po} = 63^\circ\text{C} > T_{po}/\text{desired};$ Doubling the lengths of the containers reduced T_{po} by 44°C ; but T_{po} is still too hot.	

6.0 Permeation Calculations

Helium is generated from the radiolytic decay of $^{238}\text{PuO}_2$ at a rate of 6.37×10^{11} atoms/sec/gm Pu^{238} . For a 300 gram of Pu^{238} , the helium generation rate is:

$$Q_{\text{gen}} = 3.17 \times 10^{-10} \frac{\text{mol}}{\text{sec}} \quad (\text{Eq. 6.1})$$

Permeation rate of helium through the fused quartz is described by the equations (6.2) and (6.3) depending on the gas pressure¹⁰.

Equation (6.2) describes the steady state permeation rate at low pressure, assuming the gas obeys the ideal gas law:

$$Q_{\text{perm}} = K \left(\frac{A}{L} \right) \Delta P \quad (\text{Eq. 6.2})$$

where Q = permeation rate of gas, mol/sec
 K = permeability coefficient, mol/cm-atm-sec
 A/L = effective area to thickness ratio, cm
 ΔP = pressure differential across fused silica, atm

$$K = \frac{K_0}{\Lambda} T \exp \left(\frac{-Q_k}{RT} \right)$$

where K = permeability, mol/s cm atm
 K_0 = pre-exponential factor, atom/s cm atm K
 T = absolute temperature, K
 Q_k = activation energy, kcal/mol
 R = gas constant
 Λ = Avagadro's number

At high pressure, a "fugacity" term must be introduced since gas can no longer be treated as an ideal gas. Equation (6.3) represents the steady state permeation rate at high pressure:

$$Q_{\text{perm}} = K \left(\frac{A}{L} \right) \phi P \quad (\text{Eq. 6.3})$$

where Q = permeation rate of gas, mol/sec
 K = permeability coefficient, mol/cm-atm-sec
 A/L = effective area to thickness ratio, cm
 ϕ = fugacity coefficient
 (@ each pressure, P from compressibility data)
 P = pressure at high pressure side, atm

To obtain pressure versus time profile for the storage container, the net pressure build-up must be calculated. This is done by subtracting the permeation rate of helium through the fused quartz from the helium generation rate as shown in equation (6.4).

$$PV = nRT, \quad P = \frac{nRT}{V}$$

where P = pressure
V = volume
n = # of moles of He
T = absolute temperature
R = gas constant

$$\dot{P} = \frac{\dot{n} \times R \times T}{V}$$

where $\dot{n}$ = # of moles of He/sec

$$\dot{n} = Q_{net} = Q_{gen} - Q_{perm}$$

where $Q_{GEN} = 3.17 \times 10^{-10}$ mol/sec
 $Q_{PERM} = K P A/L$

by substituting and rearranging we have

$$\dot{P} + \left(\frac{KART}{LV\Lambda} \right) P = \frac{Q_{gen} RT}{V\Lambda}$$

differentiating we get¹³

$$P = \frac{Q_{gen} L}{K A \Lambda} \left(1 - e^{-\frac{t K A R T}{L V}} \right) \quad (\text{Eq. 6.4})$$

where t is time in seconds

Figures (6.1), (6.2), and (6.3) show the helium partial pressures for the conceptual designs discussed in Section 4.0.

Figure 6.1. Pressure history for the EP-61 filtered endcap concept.

Figure 6.2. Pressure history for the EP-60 filtered endcap concept.

Figure 6.3. Pressure history for the EP-3 concept.

7.0 Stress Calculations

The stresses for the concepts presented in this report are calculated from the helium partial pressure, radius of the quartz, and the thickness of the quartz¹³. The stress for the EP-61 filter is

$$\sigma_{EP-61} = \frac{P \times R_2}{2 \times t}$$

where: P is pressure in lb/in² tangential to edge support, R_2 is the spherical radius in inches, and t is the thickness in inches. The stress for the EP-60 and EP-3 filters are

$$\sigma_{EP-60} = \sigma_{EP-3} = \frac{P \times R_2}{t}$$

where: P is pressure in lb/in², R_2 is the cylinder radius in inches, and t is the thickness in inches.

The EP-61 filter has a maximum helium partial pressure of approximately 118 psi. The corresponding stress is 885 psi (the compressive and tensile strengths given here are for room temperature, the actual strength at temperature is higher). The compressive strength of the quartz is 160,000 psi, thus the potential for fracture due to internal gas pressure is unlikely. Higher internal pressures were not considered because they would exceed the extensive qualification testing already performed on the rest of the EP-60 and EP-61 designs. The EP-61 calculation assumes uniform wall thickness and tangential edge support. Failure modes for glass are typically governed by flaws and stress concentrations thus the calculated wall stress is only a check to ensure the filter is a plausible design.

The EP-60 filter has a maximum helium partial pressure of approximately 45 psi. The corresponding stress is 180 psi. Based on the compressive strength of quartz the EP-60 filter has a pressure safety factor of over 800. This assumes a uniform wall thickness of .05 inches. The actual wall will have taper for added strength. This will increase the internal pressure and may reduce the safety factor. The most likely failure mode for this design and the EP-3 design is shock, since the tubes are cantilevered, the same failure modes described for the EP-61 apply for all the filter designs.

The EP-3 filter has a maximum helium pressure of approximately 20 psi. The corresponding stress is 130 psi. This vessel is in tension. The tensile strength of quartz is 7,000 psi, thus the pressure safety factor is over 50. The failure of glass due to flaws and stress concentrations particularly in a radioactive environment are increased when the glass is in tension as with this design.

8.0 Conclusion

Long term plutonium storage in a vessel utilizing a quartz filter for helium release is a viable option. The conceptual designs reviewed in this report are feasible storage vessel candidates. They meet the design criteria of low cost and manufacturability, and should provide safe long term storage of plutonium oxide. Although the radiolytic effects of these designs have not been addressed due to planned programmatic scope, their effects can be incorporated into the vessel design.

The EP60 Cap Concept is the most robust, manufacturable and inexpensive design concept considered in this study. The attributes of the design are:

- Quartz filter is in tension, the most robust condition for a brittle material.
- The glass transition region takes place gradually along the cylinder. This is an advantage over the disk design where the glass transition is performed place radially at the edge of the disk.
- The impact to the existing - qualified design is minimized. Modifications to existing hardware can incorporate the filter. Minimal re-qualification is required compared to a new design such as the EP3 Tube Concept.

The drawbacks to the design are that it does not meet the goal of a 40 °C skin temperature, and the EP60 does not accommodate the 1 inch diameter plutonium pellets. If these design requirements are overriding the EP3 Tube Concept would be the best concept.

The study conducted and summarized in this report completes the conceptual design phase. Future work should include detailed analysis of thermal, stress, pressure, shock and vibration environments. Also, investigation of radiolytic degradation of the quartz and NORYL need to be conducted. Although the safety factors incorporated into the designs are substantial, careful consideration is required for manufacturing of the quartz if it is used in tension. Small inclusions and stress concentrations can initiate crack growth for quartz in tension, and the effects of these imperfections can be magnified by the temperature and radiation effects.

REFERENCES

1. D. Horrell and T. George, Plutonium-238 Stabilization and Packaging, Los Alamos National Laboratory proposal
2. R. H. Doremus, *Mod. Aspects Vitreous State* **2**, 1 (1962)
3. D. E. Sets, R. W. Lee, and R. C. Grank, *J. Chem. Phys.* **34**, 17 (1961)
4. J. E. Shelby, *J. Appl. Phys.* **43**, 3068 (1972)
5. W. G. Perkins and D. R. Begeal, *J. Chem. Phys.* **54**, 1683 (1971)
6. J. E. Shelby, Molecular Solubility and Diffusion. In: M. Tomozawa, R. H. Doremus (eds) *Treatise on Materials Science and Technology*, Vol. 17, Academic Press, New York, pp 1-40.
7. F. J. Norton, *J. Am. Ceram. Soc.*, **36**, 90, (1953)
8. M. A. Whitney, Calculations of Internal Pressure Under Normal Maximum Operating Conditions, Westinghouse Savannah River Company, 1991
9. N. P. Bansal, R. H. Doremus, *Handbook of Glass Properties*
10. F. Kreith and M.S. Bohn, Principles of Heat Transfer , (West Publishing Co., St. Paul, 1973)
11. S. Glasstone, K. J. Laidler, and H. Eyring, The Theory of Rate Processes, McGraw-Hill, New York, 1941
12. Quartz Scientific, Inc. Laboratory Products Catalog QS-1010, Revised 6/90
13. Spiegel, M.R., *Advanced Mathematics for Engineers and Scientists*, pp.51, Mc Graw Hill
14. Roark and Young, Formulas for Stress and Strain, Mc Graw Hill, New York

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