Mitigation of $^{208}$Tl Gamma Dose from $^{236}$Pu Decay Chain via Chemical Removal of $^{232}$U and $^{228}$Th

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Radioisotope Thermoelectric Generators

Generator that works off of heat that is produced from radioactive decay of $^{238}\text{Pu}$. Missions are typically labeled in required $W_e$ (electric watts).
Background: The Advanced Test Reactor

- Dwindling stockpiles.
  - New production at HFIR, but more is needed.
- Production of $^{238}\text{Pu}$ in the Advanced Test Reactor
  - Produced by neutron irradiation of $^{237}\text{Np}$ by $(n,\gamma)$ reactions.
  - Undesired biproduct: $^{236}\text{Pu}$ from fast neutron $(n,2n)$ reactions.
  - Outer I positions result in $^{236}\text{Pu}$ content of ~2 ppm.
  - Higher $^{236}\text{Pu}$ concentrations from B positions ~ 6 ppm.
Background: Pu-236 Buildup

Reaction schemes for transmuting Np into Pu (Credit: Patent US 6896716 B1)
Background: Pu-236 Decay Chain

- Why is $^{236}\text{Pu}$ undesired?
  - Decay daughter $^{208}\text{Tl}$. 

- How do we mitigate the hazards from $^{208}\text{Tl}$?
  - Material aging and chemical removal of higher decay daughters $^{232}\text{U}$ and $^{228}\text{Th}$.

Methodology: Software and Parameters

- SCALE 6.2: ORIGEN Module
  - Ordinary Differential Equation/Bateman Equation solver.
  - Point depletion and decay calculations.
  - Capable of simulating material processing through specifying material removal between cases.
- Simulate a chemical processing procedure.
  - Material removed from reactor, allowed to age 150 days.
  - First chemical processing.
  - Material allowed to age additional period of time.
  - Second chemical processing. Identical process as first.
Methodology: Parameters to Test

- Starting $^{236}\text{Pu}$ Concentrations:
  - 1 ppm, 2 ppm, 4 ppm, 6 ppm, 8 ppm, 10 ppm, 12 ppm

- Processing Aging Times:
  - Initial processing at 150 days after removal from ATR.
  - Second processing after a varying time interval:
    - 1 year to 8 years.

- Decontamination Factor or Fraction Removed or Retained:
  - Chemical processing to remove uranium and thorium. All other elements retained.
  - Testing the removal fractions:
    - 97% removal
    - 99% removal
    - 99.99% removal.
  - Assume same element fraction removed for both uranium and thorium.
  - Identical process for both 1st and 2nd processing steps.
Methodology: Procedure

- Set up initial ORIGEN input with initial Pu isotopics for a given $^{236}$Pu concentration, processing times, and decontamination factors.
  - Assume 1 gram of elemental Pu.
- Run ORIGEN. Save the results.
- Repeat for each $^{236}$Pu concentration, aging time, and removal fraction.
- Desired Result: Find $^{236}$Pu concentration, removal fractions and aging time that results in:
  - Less than 1.7 microcuries $^{208}$Tl per gram of Pu.
  - $^{208}$Tl activity must not rise above 1.7 microcuries before two years after second processing. Provides two year working window.
  - Baseline comparison: Equivalent to Pu with 2 ppm $^{236}$Pu with only the initial 150 day processing for two years.
Results: $^{236}$Pu Concentration

- Higher $^{236}$Pu concentration results in higher equilibrium levels for other isotopes including $^{208}$Tl.
- $^{208}$Tl concentration doesn’t need to increase as much.
  - Leads to less time below 1.7 μCi $^{208}$Tl limit.
- Results from irradiation position in ATR.
  - Change aging time and removal fraction of U and Th to accommodate.
Results: Aging Time before 2\textsuperscript{nd} Processing

- Longer aging time leads to slower rate of $^{208}$Tl increase after second processing.
- Lower $^{236}$Pu activity at time of processing.
- Resulting $^{208}$Tl concentration is lower over time.
- More time below 1.7 $\mu$Ci $^{208}$Tl limit after processing.
Results: Increasing Uranium and Thorium Removal

- More $^{232}\text{U}$ and $^{228}\text{Th}$ removed.
- $^{208}\text{Tl}$ concentration drops to new immediate equilibrium with respect to $^{232}\text{U}$.
  - $^{208}\text{Tl}$ has to increase more.
  - Minimize rate of increase in $^{208}\text{Tl}$
  - May lead to more time below 1.7 $\mu$Ci $^{208}\text{Tl}$ limit.
Analysis

• Summary factors of concentration, aging and rate of increase.
  • $^{236}$Pu Concentration
    • More $^{236}$Pu $\rightarrow$ more $^{208}$Tl.
  • Second Processing Aging Time
    • Slower $^{208}$Tl increase due to lower $^{236}$Pu activity after aging.
    • Increased aging $\rightarrow$ less $^{208}$Tl after processing.
  • Increased Removal Uranium and Thorium
    • Lower $^{208}$Tl after processing.
    • Increased removal $\rightarrow$ less $^{208}$Tl

Which combinations stay below 1.7 μCi $^{208}$Tl limit for 2 years?
Second Reprocessing Aging Time Results

The graph illustrates the relationship between the aging time (in years) and the Pu-236 concentration (in ppm) for different removal percentages: 99.99%, 99%, and 97%. The graph shows that as the Pu-236 concentration increases, the aging time also increases. The curves for each removal percentage diverge, indicating that higher removal percentages result in longer aging times for the same concentration.
Conclusion

• Increasing removal of uranium and thorium is important.

• Chemical processing procedures that remove more uranium and thorium make plutonium with higher $^{236}\text{Pu}$ concentrations viable materials with respect to our $^{208}\text{Tl}$.

• Higher removal of uranium and thorium also reduces the required aging time. Makes plutonium ready to use earlier. Free up storage space.

• Future Work?
  – Could more stages of chemical processing be worthwhile?
  – Vary removal between 1st and 2nd processing, or between uranium and thorium?
  – Finer time steps for second aging time?
  – Optimization necessary.
  – Still limited by willingness to allow plutonium to age for several years. Storage space needed.
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References


Questions?