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

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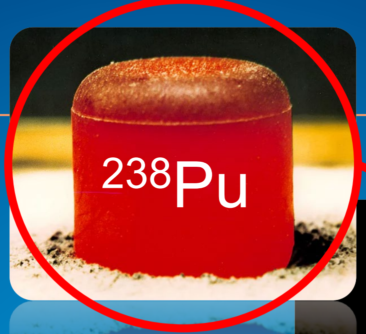
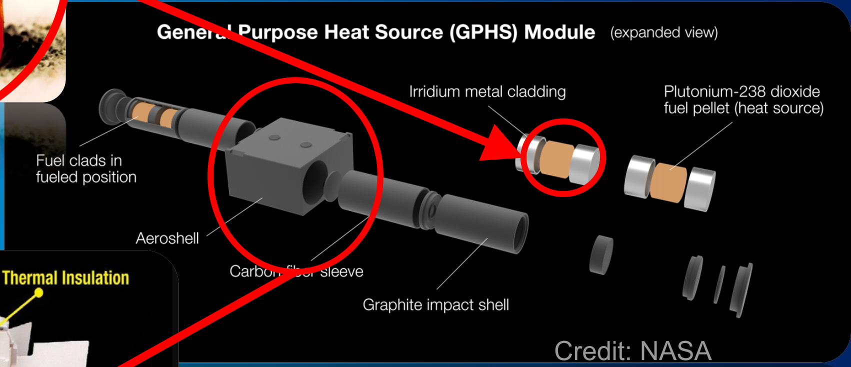
Outline

- Background and Objectives
- Methodology
- Results
- Analysis
- Conclusions

Background: RTGs

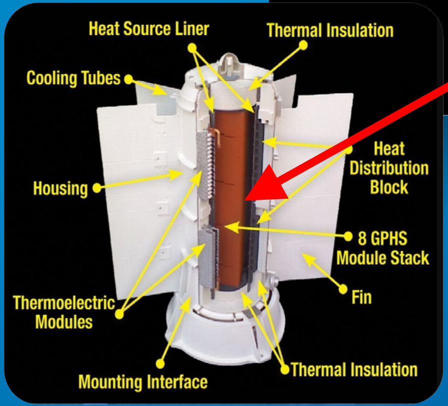




***R**adioisotope*
***T**hermoelectric*
***G**enerators*

Generator that works off of heat that is produced from radioactive decay of ^{238}Pu.

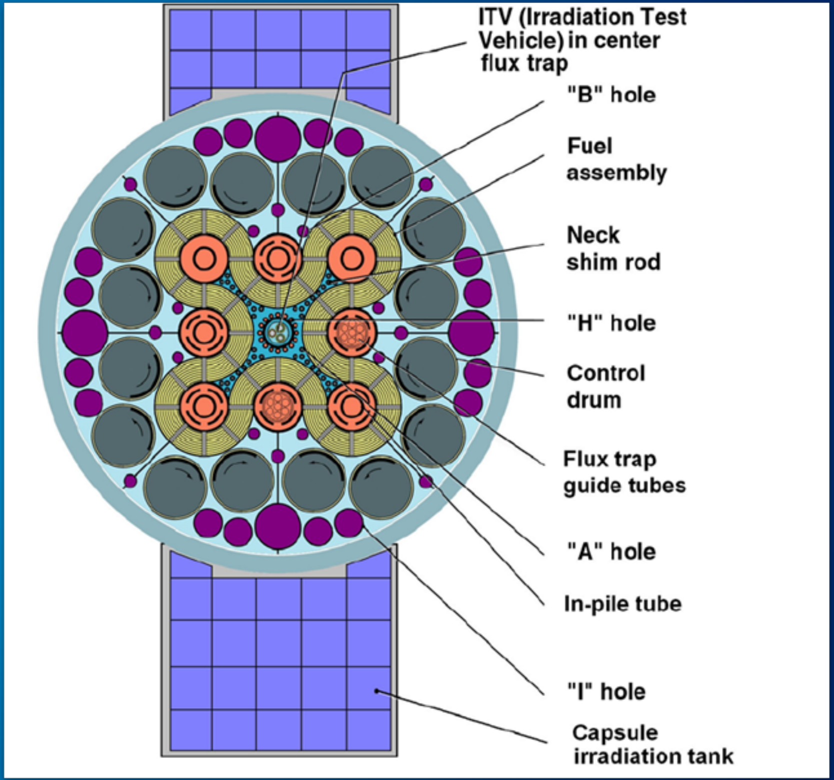
Missions are typically labeled in required W_e(electric watts).



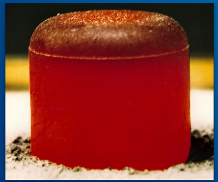
Credit: NASA

Background: The Advanced Test Reactor

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



Credit: Idaho Nat. Lab



Credit: Los Alamos Nat. Lab

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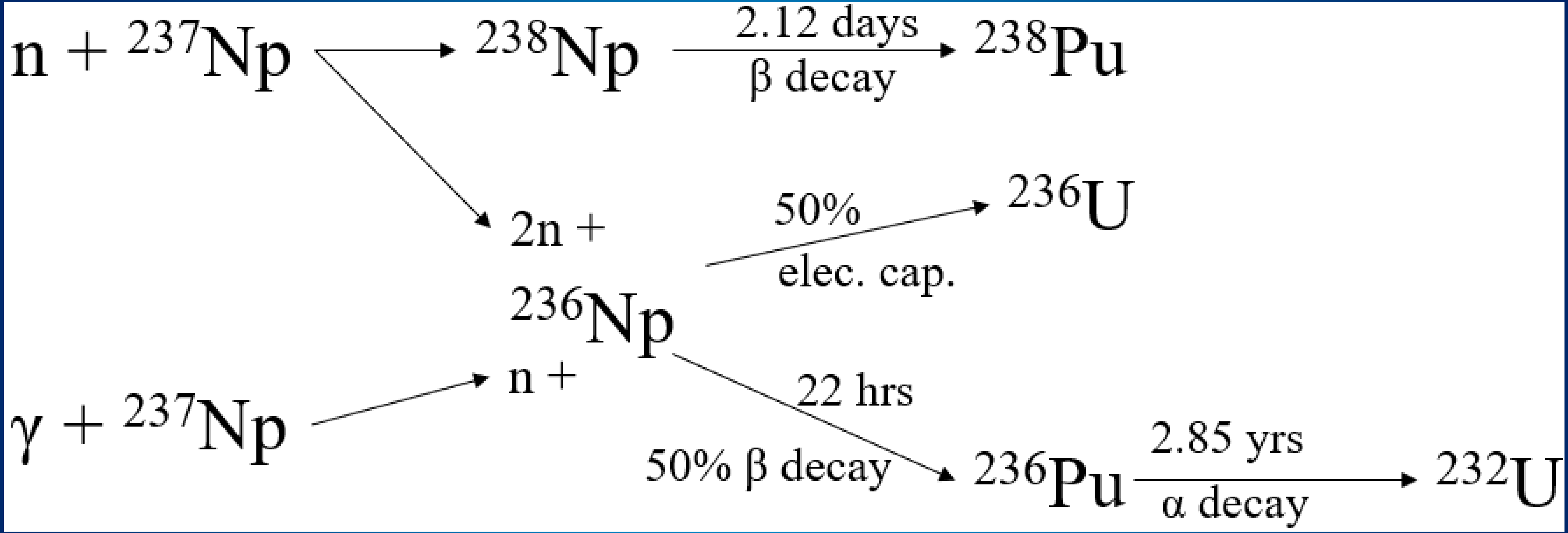
Background: Pu-236 Buildup



Reaction schemes for transmuting Np into Pu (Credit: Patent US 6896716 B1)

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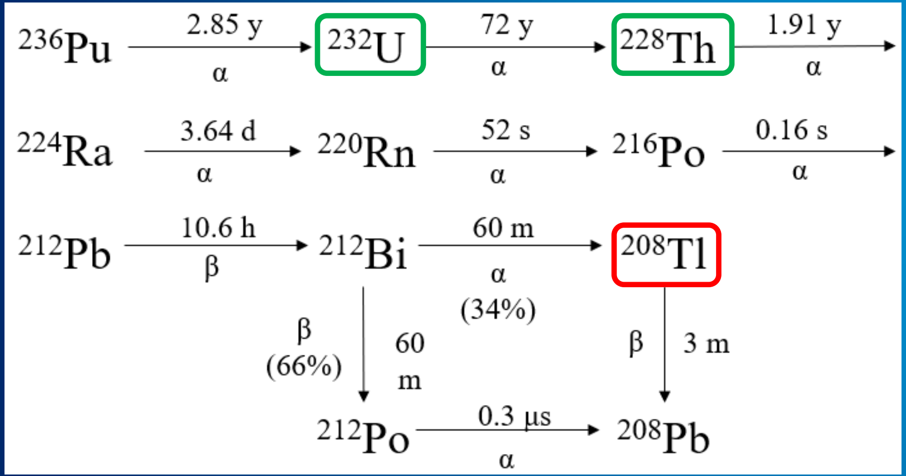
Background: Pu-236 Decay Chain

- Why is ^{236}Pu undesired?
  - Decay daughter ^{208}Tl.
- How do we mitigate the hazards from ^{208}Tl?
  - Material aging and chemical removal of higher decay daughters ^{232}U and ^{228}Th.



Principal Gamma Ray Abundances From ^{236}Pu Daughters

Nuclide	Gamma, Mev.	% Abundance
^{212}Pb	0.239	82
^{212}Bi	0.727	6
^{208}Tl	0.277	3
	0.511	8
	0.583	30
	0.860	3
	2.62	34

Matlack, G., and Metz, F. “Radiation Characteristics of Plutonium-238,” 1967. Los Alamos Scientific Laboratory.

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.

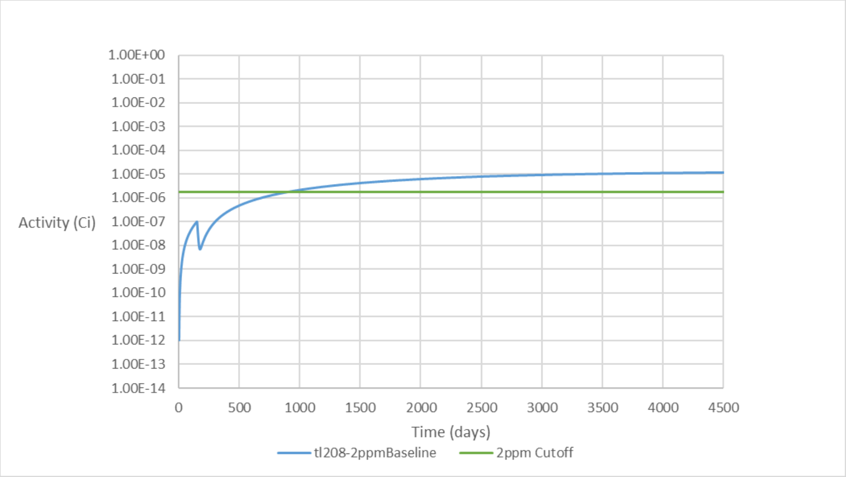
Methodology: Parameters to Test

- Starting ^{236}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.

```
processing{
        retained=[ U=0.0001
                   Th=0.0001
                   Pu=1
                   Ra=1
                   Rn=1
                   Po=1
                   Pb=1
                   Bi=1
                   Tl=1]
              }
```

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.



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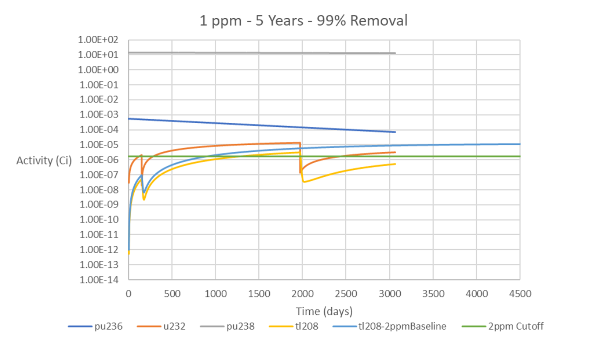
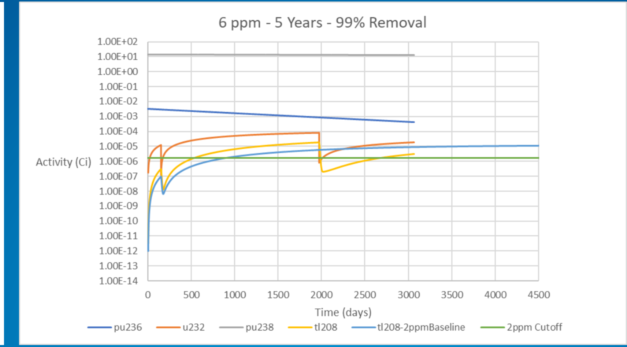
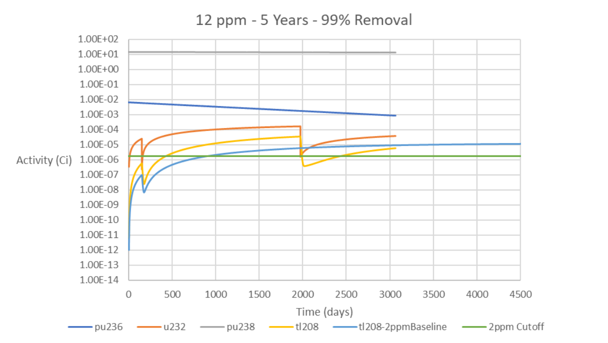
Results: ^{236}Pu Concentration







- 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.

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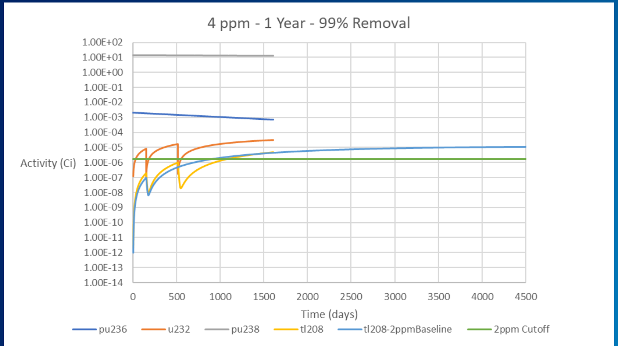
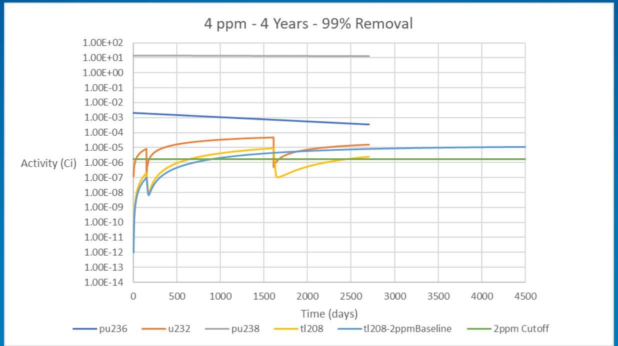
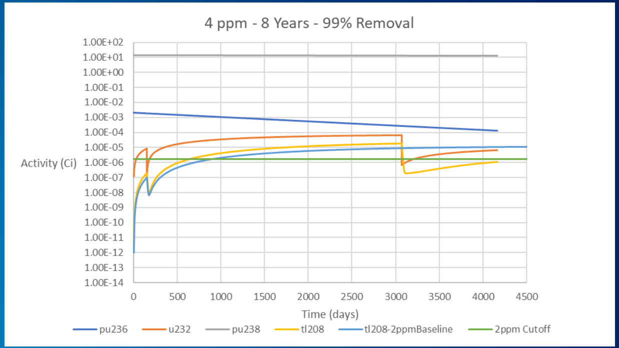
Results: Aging Time before 2nd Processing







- 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 μCi ^{208}Tl limit after processing.

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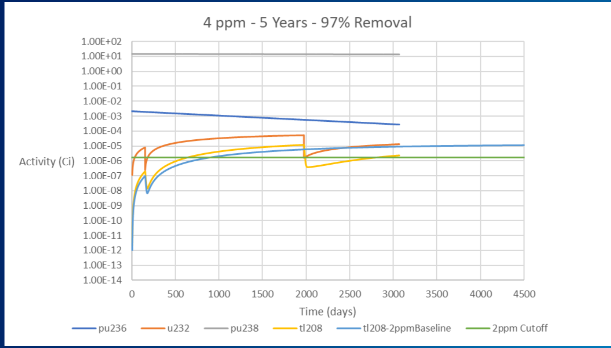
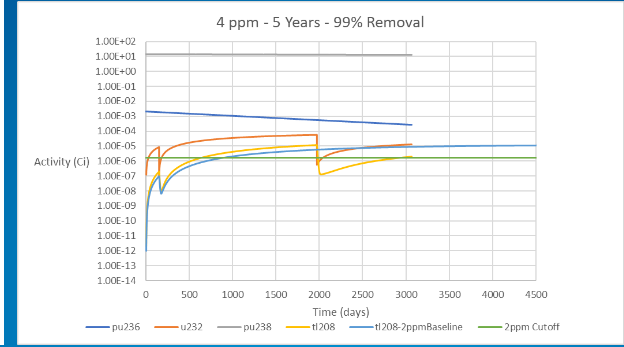
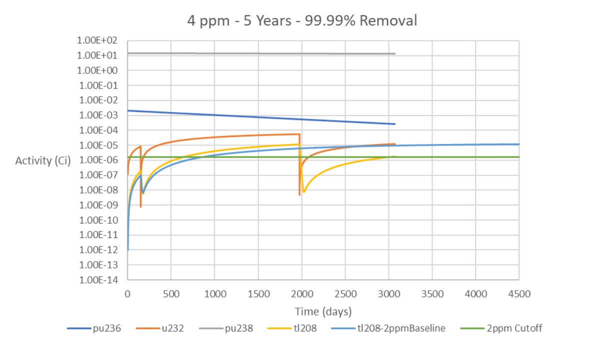
Results: Increasing Uranium and Thorium Removal

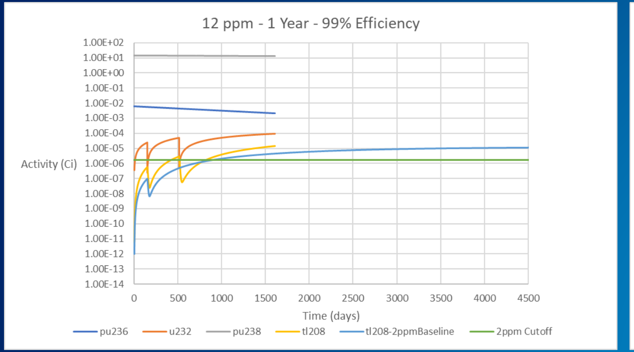


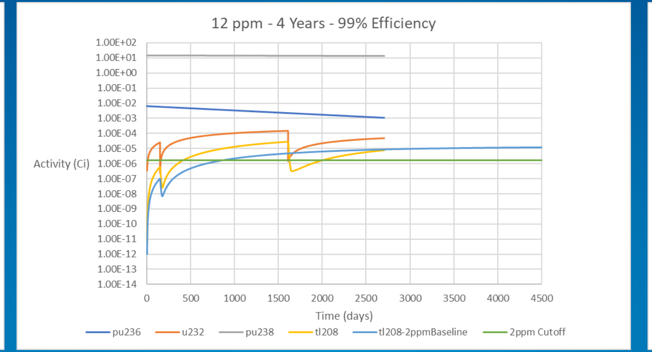




- More ^{232}U and ^{228}Th removed.
- ^{208}Tl concentration drops to new immediate equilibrium with respect to ^{232}U.
  - ^{208}Tl has to increase more.
  - Minimize rate of increase in ^{208}Tl
  - May lead to more time below 1.7 µCi ^{208}Tl limit.

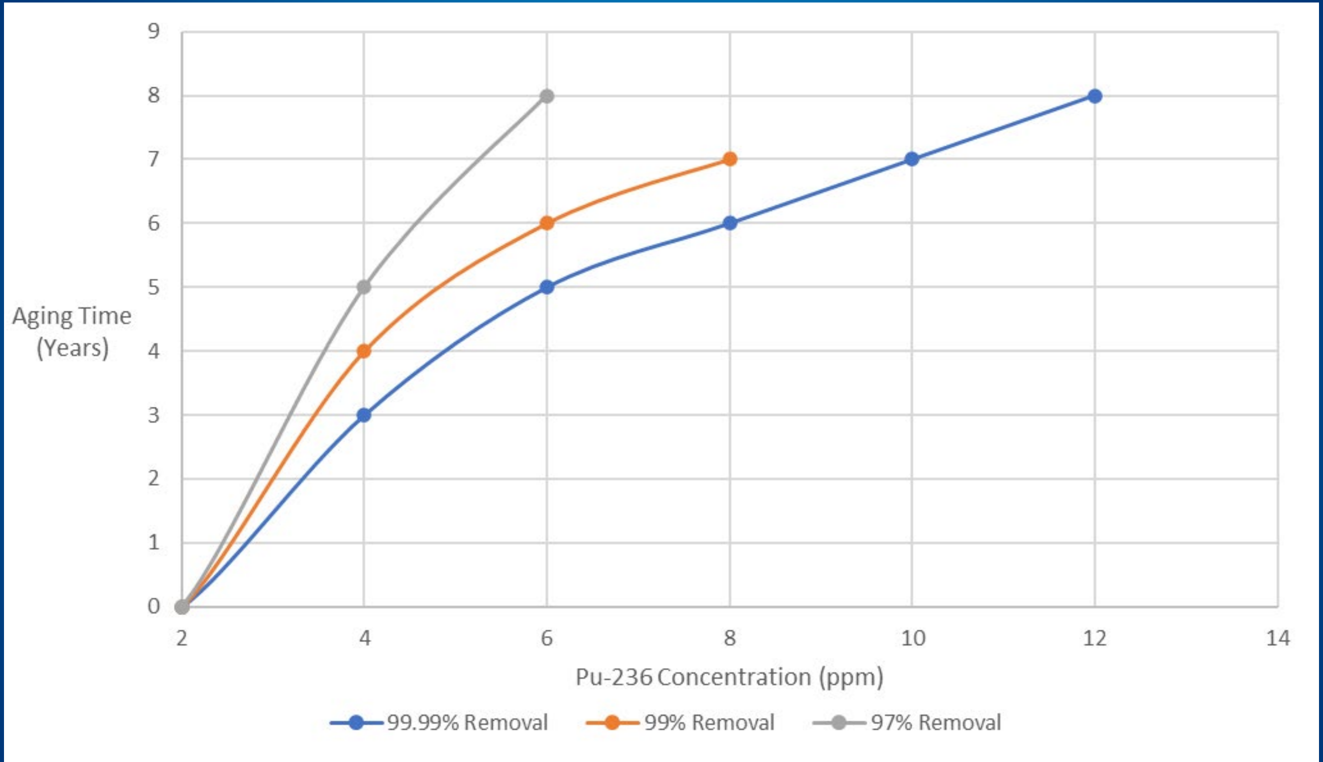
Analysis





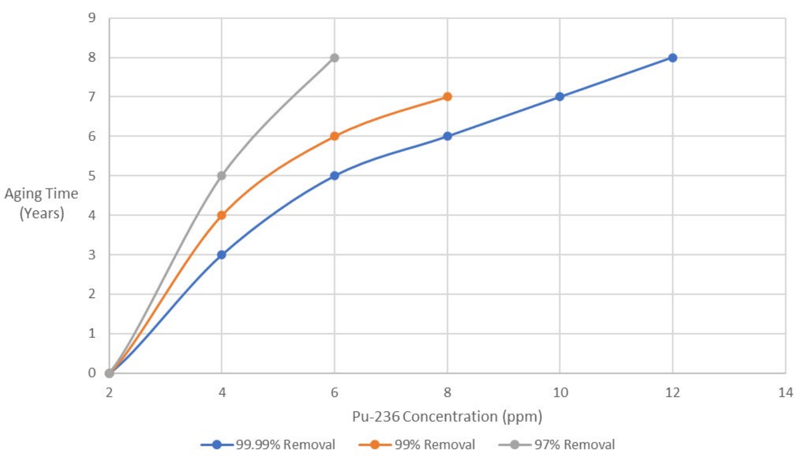
- Summary factors of concentration, aging and rate of increase.
- ^{236}Pu Concentration
  - More ^{236}Pu → more ^{208}Tl.
- Second Processing Aging Time
  - Slower ^{208}Tl increase due to lower ^{236}Pu activity after aging.
  - Increased aging → less ^{208}Tl after processing.
- Increased Removal Uranium and Thorium
  - Lower ^{208}Tl after processing.
  - Increased removal → less ^{208}Tl
- Which combinations stay below 1.7 μCi ^{208}Tl limit for 2 years?

Second Reprocessing Aging Time Results



Conclusion

- Increasing removal of uranium and thorium is important.
- Chemical processing procedures that remove more uranium and thorium make plutonium with higher ^{236}Pu concentrations viable materials with respect to our ^{208}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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Questions?