Optimization of Pu-238 Production in the Advanced Test Reactor

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Objective

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Background
Create an ATR irradiation design meeting radiological exposure and safety standards which, with HFIR, can produce 1.5 kg Pu-238 annually by 2025.
Commonly Used Terms

- ATR - Advanced Test Reactor at Idaho Natl. Lab
- HFIR - High Flux Isotope Reactor at Oak Ridge Natl. Lab
- MMRTG - Multi-Mission Radioisotope Thermoelectric Generator
- Target Rod - Pellet stack assembly with cladding, plenum, and spacers
- Pellet - $\text{NpO}_2$ - Al powder mixed and pressed together
- Plenum - Space for fission gases to go during irradiation
- Cladding - Outer aluminum coating of pellet stack
- Basket - Aluminum casing that holds targets in place
- Irradiation Facility - Aluminum container holding target baskets
- I-Position/Channel - Where target rods are placed on ATR periphery
- B-Position/Channel - Where target rods are placed closer to ATR fuel lobes
- Np-237 - Neptunium isotope that decays into plutonium when irradiated
- Pu-238 - Plutonium isotope used in RTGs for spacecraft missions
- Pu-236 - Plutonium isotope, decay daughters include 2.62 MeV gammas
Pu-238 Reaction Scheme

Reaction schemes for transmuting Np into Pu (Credit: Patent US 6896716 B1 (2005))
MMRTGs

Multi-Mission Radioisotope Thermoelectric Generators

Generator running on heat produced from radioactive decay of $^{238}\text{Pu}$

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

Credit: NASA
The ATR

- Irradiation of Np-237 at ATR & HFIR
- HFIR limited to 300-500g (using all positions)
- ATR I and B positions most readily available
Design Specs & Geometry
Design Positions

- **Small I**: 4 Positions, 4 Targets
- **Medium I**: 16 Positions, 128 Targets
- **Large I**: 4 Positions, 88 Targets
- **Large B**: 4 Positions, 4 Targets
- **Small B**: 7 Positions, 7 Targets
Design Positions Cont.

- Water Jacket
- Basket
- Water Jacket
- Irradiation Facility
- Cladding
- Pellet Stack
# Design Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Target Rod Length</td>
<td>40 inches MAX</td>
</tr>
<tr>
<td>Pu-236 Concentration</td>
<td>2 ppm MAX</td>
</tr>
<tr>
<td>NpO$_2$- Al Pellet Concentration (volumetric)</td>
<td>30% MAX</td>
</tr>
<tr>
<td>Pu-238 Quality</td>
<td>87% MIN</td>
</tr>
<tr>
<td>Pu-238 Conversion Ratio</td>
<td>10% MIN</td>
</tr>
</tbody>
</table>
Methodology
Method

- Used previous results to define optimization scope
- Modeled in Serpent
  - Used depletion analysis to observe material levels
  - Simplified lattice structures for ease of editing
- Advantages of Serpent over MCNP
  - Runs faster
  - Advanced lattice types
- Idaho National Laboratory’s High Performance Computing system used for modeling
Results
Range of Analysis

- Varied target length from 35”, 40”, and 48”
- Increased initial NpO₂ conc. from 20% to 50%
  - 2% step increments
- Added targets in alternative positions in ATR
  - Small and Large B-Positions
  - Small I-Positions
Longer targets have superior:

- Annual Pu-238 Yield (200-300 g), Pu-236 Concentration (<2 ppm), and Pu Quality (96-98%)
Small I-Positions

- Low annual Pu-238 production
- 6-9% Conversion ratio
- 87-91% Quality
Large B-Positions

- Higher annual Pu-238 production than small I-positions
- Pu-236 concentration increase to 3-4 ppm, closer to reactor fuel
- 10-14% conversion ratio, Quality only acceptable at high pellet conc.
Small B-Positions

- Significant annual Pu-238 production from only 7 rods
- Pu-236 concentration unacceptable at 6-8 ppm
- 16-20% Conversion Ratio, but <83% Quality Unacceptable
Individual Position Yield

40” Rod, 30% NpO₂

Note: Large I positions have 22 rods per position and medium I positions have 8. All other positions have 1 rod/position. Production increases as positions are used together.
### Pu-238 Production/Position

*40” Rod, 30% NpO₂*

<table>
<thead>
<tr>
<th>Position</th>
<th>Target Quantity</th>
<th>(^{238}\text{Pu}) Production (g)</th>
<th>(^{236}\text{Pu}) Conc. (ppm)</th>
<th>(^{238}\text{Pu}) Quality (%)</th>
<th>(^{238}\text{Pu}) Conversion Ratio (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Large I</td>
<td>22</td>
<td>27.05</td>
<td>1.50</td>
<td>97.01</td>
<td>2.16</td>
</tr>
<tr>
<td>Medium I, inner</td>
<td>8</td>
<td>12.19</td>
<td>1.33</td>
<td>96.07</td>
<td>2.68</td>
</tr>
<tr>
<td>Medium I, outer</td>
<td>8</td>
<td>12.80</td>
<td>1.39</td>
<td>96.09</td>
<td>2.81</td>
</tr>
<tr>
<td>Small I</td>
<td>1</td>
<td>3.86</td>
<td>1.54</td>
<td>90.07</td>
<td>6.79</td>
</tr>
<tr>
<td>Large B</td>
<td>1</td>
<td>6.55</td>
<td>3.43</td>
<td>86.94</td>
<td>11.50</td>
</tr>
<tr>
<td>Small B</td>
<td>1</td>
<td>10.34</td>
<td>6.46</td>
<td>80.01</td>
<td>18.15</td>
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</tbody>
</table>
Fission Rate/Thermal Flux

Neutron Capture
Heat distribution from Serpent

- Track total heating deposition for each target rod
- Calculate decay heat in each target rod
Analysis & Recommendations
**Design Recommendations**

**Max Yield Design:**

778.04 g of Pu-238 @ 96.72%, 3.34 ppm (Pu-236)

- 48” Rods, 50% NpO₂, All I and B positions
- Ran full core instead summing individual positions; interactions are significant

**Conservative Yield Design:**

350.58 g of Pu-238 @ 96.32%, 1.63 ppm (Pu-236)

- 40” Rods, 30% NpO₂, All I and Large B positions (no Small B)
Stockpile Enrichment

Assuming 16 kg of usable 78% Pu available in current stockpile:

<table>
<thead>
<tr>
<th>Design</th>
<th>Annual 87% Pu-238 (kg)</th>
<th>Pu to Add from Stockpile (kg)</th>
<th>Stockpile Gone (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>All I and B Positions</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>48” Rods, 50% NpO₂</td>
<td>1.456</td>
<td>0.869</td>
<td>18</td>
</tr>
<tr>
<td>I and Large B Positions</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>40” Rods, 30% NpO₂</td>
<td>0.6445</td>
<td>0.377</td>
<td>42</td>
</tr>
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</table>
Conclusion
Conclusion & Future Work

ATR/HFIR production can meet NASA’s goal, but with serious concessions in quality and $^{236}\text{Pu}$. In addition, ATR/HFIR positions are not always available.

1. Flux Traps and other High-Priority positions
   a. Deal with high Pu-236 levels, low Pu-238 quality

2. Pure Np-237 Pellets
   a. Deal with high Pu-236 levels, low conversion ratio
Acknowledgements

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References

Questions?

Scrapped Design 5

Scrapped Design 6

Background  Design  Results  Analysis  Future Work
Extra Slides
Pu-236 Daughters

Pu-236 Daughters Cont.

Production of Hazardous Decay Daughters of Pu-236

- TI-208
- Pb-212
- Bi-212
Pu Trends Over Cycle Length

48” Rod, 30% NpO₂

Note: Small B positions do not include B7 (for HSIS).
**What about the 1993 study***?

<table>
<thead>
<tr>
<th></th>
<th>1993 Study</th>
<th>2019 Analysis</th>
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</thead>
<tbody>
<tr>
<td>ATR Positions Utilized</td>
<td>3 modified Flux Traps, Large B, Small B, Small I</td>
<td>Large I, Medium I, Small I, Large B</td>
</tr>
<tr>
<td>Operational Cycle</td>
<td>288 days at power, 72 day shutdown (24/6)</td>
<td>186 days at power, 180 day shutdown (62/60)</td>
</tr>
<tr>
<td>Power Level</td>
<td>200 MW</td>
<td>105 MW</td>
</tr>
<tr>
<td>237Np Irradiated</td>
<td>102.1 kg</td>
<td>12.76 kg</td>
</tr>
<tr>
<td>238Pu Produced</td>
<td>11.35 kg</td>
<td>350 g</td>
</tr>
<tr>
<td>238Pu &lt; 2 ppm</td>
<td>1.07 kg</td>
<td>~315 g</td>
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</tbody>
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- PFS-3 Design Specification for ATR not incorporated in previous designs. Increases target size by 90%.
- Only Design 2 from previous year runs successfully.
- MCNP did not include $^{237}\text{Np}(\gamma,\text{n})^{236}\text{Np} \rightarrow ^{236}\text{Pu} + \beta^-$ process.
## 2018 Results

<table>
<thead>
<tr>
<th>Design</th>
<th>#1</th>
<th>#2</th>
<th>#3</th>
<th>#4</th>
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<tbody>
<tr>
<td>Conversion Ratio</td>
<td>0.038</td>
<td>0.030</td>
<td>0.033</td>
<td>0.025</td>
</tr>
<tr>
<td>Pu-238 (g)</td>
<td>129</td>
<td>204</td>
<td>193</td>
<td>267</td>
</tr>
<tr>
<td>Quality (%)</td>
<td>0.95</td>
<td>0.96</td>
<td>0.96</td>
<td>0.97</td>
</tr>
<tr>
<td>Pu 236 (ppm)</td>
<td>1.17</td>
<td>1.08</td>
<td>1.28</td>
<td>1.65</td>
</tr>
<tr>
<td>Number of rods</td>
<td>104</td>
<td>208</td>
<td>184</td>
<td>332</td>
</tr>
<tr>
<td>Analysis Factor</td>
<td>1.0000</td>
<td>0.6001</td>
<td>0.6783</td>
<td>0.3088</td>
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