Document 65
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244Pu target manufacturing
January 14-16, 2008

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This work performed under the auspices of the U.S. Department of Energy by
Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

UCRL-XXXX-12345
We can make the required targets

Diamond-turned targets

Rippled target

Stepped target

We are extending our existing capabilities to build Pu targets

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Using $^{244}\text{Pu}$ poses unique challenges

We are using $^{244}\text{Pu}$ because of its low specific radioactivity

- Material is currently in solution
  - We need metal
- Oxidation of metal needs to be controlled
  - We need inert atmospheres
- Total supply is 1 gram
  - We need to recover used material

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Relative radioactivity of available material</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>1</td>
</tr>
<tr>
<td>$^{242}\text{Pu}$</td>
<td>$1/14$ (8 grams available)</td>
</tr>
<tr>
<td>$^{244}\text{Pu}$</td>
<td>$1/64$ (1 gram available)</td>
</tr>
</tbody>
</table>

Further isotopic separation is not necessary

We are addressing these challenges

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We have begun the development to produce metal at the milligram scale
We will process $^{244}$Pu ingots and produce fully assembled targets.

1. **Input: $^{244}$Pu metal**
   - Receive purified "ingots"
   - Size: ~2 mm

2. **Form a blank**
   - Re-melt metal, form near-net shape
   - Grain size refinement, polishing

3. **Shaping operations**
   - Manufacture parts with diamond turning lathe

4. **Layering and metrology**
   - Apply materials to back and front.
   - Polishing and interferometry

5. **Assembly**
   - Assemble final package

6. **Ship to experimental site**
   - Release part in a container for transport

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Inert atmospheres are required for sample processing and target assembly

- A dedicated clean glovebox line is required
  - Equipment contaminated with $^{239}$Pu compromises the ES&H benefits of $^{244}$Pu

90% of target components can and will be built in existing facilities
$^{244}$Pu recovery is a requirement

$\sim 100\%$ returned

Processing from solution yields 90% to metal

40% unused material from manufacturing is returned

Target manufacturing yields 60% of metal to target

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We have made progress

**We can diamond turn α-phase Pu**

- [Graph showing raw data with annotations: Data filtered to remove machine noise. Surface finish (RMS) = 0.014 µm.]
- [Image showing a 2 mm scale with a surface finish.

**We can control and characterize grain size**

- [Image showing an area with a 0.01 mm scale.
- Average grain size = 23.8 µm.

- [Image showing another area with a 0.01 mm scale.
- Average grain size = 8.7 µm.

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We plan to be producing targets in FY12

<table>
<thead>
<tr>
<th>FY08</th>
<th>FY09</th>
<th>FY10</th>
<th>FY11</th>
<th>FY12</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glovebox design</td>
<td>Glovebox manufacture</td>
<td>Glovebox installation</td>
<td>Commission gloveboxes and equipment</td>
<td></td>
</tr>
<tr>
<td>Room construction</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Purchase/Install major equipment</td>
<td></td>
<td></td>
<td>Receive material</td>
</tr>
<tr>
<td>Pu micrometallurgy development</td>
<td>Metrology development</td>
<td>Coating process development</td>
<td>Target fabrication with surrogate material</td>
<td>Assemble Pu target and deliver</td>
</tr>
</tbody>
</table>
A detailed resource-loaded plan exists

- Approximate costs over 3.5 years
  - Hardware: $10 million
  - Process development: $5 million

$^{244}$Pu target fabrication represents 25% of the total cost of the campaign
$^{244}$Pu target production team

- Pyrochemistry
- Pu micrometallurgy
- Glovebox design
- Facility construction
- Target integration
Backup slides
We have expertise in processing small-scale Pu samples

- Crucible for microscale Pu melts
- Mechanical- and electro-polished surfaces

2.5 mm

8.3 mg δ Pu

1 mm
We are assessing Superblock as a possible location for the target facility

- Cost
  - No space charges in Superblock, but additional training costs incurred for workers
- Schedule
  - Superblock operates with limited hours, limited certified personnel
- Capabilities
  - Most small-scale actinide tools in Superblock are contaminated with higher-activity isotopes

The small amount of $^{244}\text{Pu}$ and its low activity allow us to work outside of Superblock

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We have expertise in handling small Pu samples at LLNL

- We have capabilities for small-scale actinide work
- Development for $^{244}\text{Pu}$ micrometallurgy will be done with $^{239}\text{Pu}$ in existing facilities

<table>
<thead>
<tr>
<th>Sample</th>
<th>Dimensions</th>
<th>Mass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diamond anvil cell</td>
<td>20 - 200 μm disc, 10 - 30 μm thick</td>
<td>0.05 - 15 μg</td>
</tr>
<tr>
<td>EXAFS</td>
<td>3 mm disc, 15 μm thick</td>
<td>1.7 mg</td>
</tr>
<tr>
<td>Resistometry</td>
<td>5 mm x 5 mm, 10 μm thick</td>
<td>4 mg</td>
</tr>
<tr>
<td>Transmission electron microscopy</td>
<td>3 mm disc, 20 nm - 150 μm thick</td>
<td>&lt; 17 mg</td>
</tr>
<tr>
<td>X-ray diffraction</td>
<td>3 mm disc, 150 μm thick</td>
<td>17 mg</td>
</tr>
<tr>
<td>Magnetic measurements</td>
<td>1 - 2 mm$^3$</td>
<td>16 - 127 mg</td>
</tr>
<tr>
<td>Differential scanning calorimetry</td>
<td>3 mm x 2 mm cylinder</td>
<td>225 mg</td>
</tr>
<tr>
<td>Target (for comparison)</td>
<td>3-6 mm disc, 100 μm thick</td>
<td>14-56 mg</td>
</tr>
</tbody>
</table>
Parts will be characterized with interferometry

- Parallelism
- Flatness
- Thickness
- Step height, ripple dimensions
- 10 nm resolution

Zygo interferometer

Interferometer data from a rippled sample
We are improving electropolishing techniques for characterizing microstructure

Initial work

Mechanically- and electro-polished surfaces

Recent work
We plan to explore 3 pyrochemistry metallization routes

Pu solution → Ion exchange → Precipitation/Calcination → Fluoride → Fluorinate → Bomb reduction

Oxide → Direct Oxide Reduction → Metal Product

Li Reduction volatilization

Methods development with $^{242}$Pu will allow us to determine the best process path for $^{244}$Pu

Goal: high yield and high purity with minimal inclusions
Renovation of wet chemistry glovebox for pyrochemistry operations completed

Before: Wet chemistry glovebox
After: Pyrochemistry glovebox
Materials usage and recovery in production mode

- Available: 1g of $^{244}$Pu in solution
- Goal: Metal produced in lots of 100-500 mg
- Goal: Process time ~14 days

- Plan: 20 to 55mg per target
- Goal: Process time 180 days for 5 targets

- >99% material returned
- 20 Shots per year for 5 years

Pyrochemistry → Target Fabrication → Experiment

10% Return → 40% Return → ~100% Return
Sample recovery: The catcher

Requirements for the catcher

- Should be able to collect >99% of sample materials moving at velocities of up to 20 km/s
- Transparent to 58 keV backlighter
- Rugged housing
- Holding fixture from DIM90-45 image plate housing
- As close as possible to the sample
The degree of purity has to be determined by analytical methods.

We have demonstrated the ability to perform optical metallography on mg-sized specimens of $\alpha$ Pu and to characterize inclusions from the pyrochemistry.
## Plutonium isotopes

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life (years)</th>
<th>Specific Activity (Ci/g)</th>
<th>Decay mode</th>
<th>Radiation Energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\alpha$</td>
</tr>
<tr>
<td>$^{236}\text{Pu}$</td>
<td>2.9</td>
<td>540</td>
<td>$\alpha$</td>
<td>5.8</td>
</tr>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>88</td>
<td>17</td>
<td>$\alpha$</td>
<td>5.5</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>24,000</td>
<td>0.063</td>
<td>$\alpha$</td>
<td>5.1</td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>6,500</td>
<td>0.23</td>
<td>$\alpha$</td>
<td>5.2</td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>14</td>
<td>100</td>
<td>$\beta$</td>
<td>$&lt;$</td>
</tr>
<tr>
<td>$^{242}\text{Pu}$</td>
<td>380,000</td>
<td>0.0040</td>
<td>$\alpha$</td>
<td>4.9</td>
</tr>
<tr>
<td>$^{244}\text{Pu}$</td>
<td>83,000,000</td>
<td>0.000018</td>
<td>$\alpha$</td>
<td>4.6</td>
</tr>
</tbody>
</table>

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## Isotopes of available materials

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Specific Activity (Ci/g)</th>
<th>Atomic %</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{244}\text{Pu})</td>
<td>0.000018</td>
<td>98.6701</td>
</tr>
<tr>
<td>(^{242}\text{Pu})</td>
<td>0.0040</td>
<td>0.9949</td>
</tr>
<tr>
<td>(^{241}\text{Pu})</td>
<td>100</td>
<td>0.0133</td>
</tr>
<tr>
<td>(^{240}\text{Pu})</td>
<td>0.23</td>
<td>0.3143</td>
</tr>
<tr>
<td>(^{239}\text{Pu})</td>
<td>0.063</td>
<td>0.0060</td>
</tr>
<tr>
<td>(^{238}\text{Pu})</td>
<td>17</td>
<td>0.0013</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Specific Activity (Ci/g)</th>
<th>Atomic %</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{242}\text{Pu})</td>
<td>0.0040</td>
<td>99.958</td>
</tr>
<tr>
<td>(^{244}\text{Pu})</td>
<td>0.000018</td>
<td>0.002</td>
</tr>
<tr>
<td>(^{241}\text{Pu})</td>
<td>100</td>
<td>0.01</td>
</tr>
<tr>
<td>(^{240}\text{Pu})</td>
<td>0.23</td>
<td>0.022</td>
</tr>
<tr>
<td>(^{239}\text{Pu})</td>
<td>0.063</td>
<td>0.005</td>
</tr>
<tr>
<td>(^{238}\text{Pu})</td>
<td>17</td>
<td>0.003</td>
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</tbody>
</table>
## Isotopics of available materials

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Atomic %</th>
<th>Radioactivity in 1 g of solution (Ci)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{244}$Pu</td>
<td>98.6701</td>
<td>$1.776 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>0.9949</td>
<td>$3.947 \times 10^{-5}$</td>
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<tr>
<td>$^{241}$Pu</td>
<td>0.0133</td>
<td>$1.316 \times 10^{-2}$</td>
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<tr>
<td>$^{240}$Pu</td>
<td>0.3143</td>
<td>$7.112 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>0.0060</td>
<td>$3.730 \times 10^{-6}$</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>0.0013</td>
<td>$2.237 \times 10^{-4}$</td>
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<td>$3.998 \times 10^{-3}$</td>
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<tr>
<td>$^{244}$Pu</td>
<td>0.002</td>
<td>$3.630 \times 10^{-10}$</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>0.01</td>
<td>$9.959 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>0.022</td>
<td>$5.018 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>0.005</td>
<td>$3.111 \times 10^{-6}$</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>0.003</td>
<td>$5.016 \times 10^{-4}$</td>
</tr>
</tbody>
</table>
Fluoride reduction: metallothermic (bomb) reduction

- The reaction is performed in an inductively heated pressure vessel in a non-vitrified crucible
  - Non-vitreous crucible necessary to withstand the sharp temperature rise caused by the large heat of reaction
- The reaction reaches high enough temperatures to melt the calcium fluoride slag that is formed in the reaction allowing the product metal to coalesce into a metal button

\[
2\text{Ca} + \text{PuF}_4 \rightarrow \text{Pu}^0 + 2 \text{CaF}_2 \quad \text{with iodine initiator}
\]

or

\[
2\text{Ca} + \frac{1}{4} \text{PuO}_2 + \frac{3}{4} \text{PuF}_4 \rightarrow \text{Pu}^0 + \frac{3}{2}\text{CaF}_2 + \frac{1}{2} \text{CaO}
\]
Fluoride reduction: preparation of PuF$_4$

- Plutonium tetrafluoride is prepared by several methods, including:
  - Fluorination of plutonium peroxide with HF gas through a platinum frit
  - Fluorination of low fired plutonium dioxide with HF gas through a platinum or Inconel frit
  - Precipitation of plutonium trifluoride by HF and then the subsequent conversion of the PuF$_3$ to 3/4PuF$_4$ and 1/4PuO$_2$ by drying

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Direct oxide reduction

PuO$_2$ + 2Ca$^0$ → Pu$^0$ + 2CaO (sol'n in CaCl$_2$)

- PuO$_2$ is reduced to metal by direct oxide reduction (DOR)
  - Primary reaction takes place in molten CaCl$_2$ at ~900°C
  - Reaction is spontaneous with $\Delta G^0_r = -47$ kcal/mole PuO$_2$
Lithium reduction

\[ \text{PuO}_2 + 4 \text{Li} \rightarrow \text{Pu}^0 + 2\text{Li}_2\text{O} \]

- \( \text{PuO}_2 \) is reduced by lithium vapor to produce Pu metal
  - Lithium oxide formed in the reaction can be removed in vacuum at temperature
  - \( \text{Li}_2\text{O} \) will volatilize away from the Pu metal under vacuum after the reaction is completed

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Metal preparation recovery: 
*sustainability of program*

- Recovery methods
  - Bomb reduction residues are primarily calcium fluoride slag and MgO crucible. These can be dissolved in nitric acid and aluminum nitrate to complex the fluoride
  - DOR residues are the calcium chloride salt and the MgO crucible. This material can be recovered by HCl dissolution
  - Li reduction residues require flow-sheet development. This method could result in minimum inclusions
- All processes can be followed by ion exchange and precipitations