236\text{Np}/236\text{Pu} production via the $^{235}\text{U}(d,n)$
and $^{238}\text{U}(p,3n)$ channels

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Isotope Dilution Mass Spectrometry: Standard Reference Material $^{236}\text{Np}$

- Neptunium-236g ($t_{1/2}=1.5\times10^5$ a) is used for IDMS determination of $^{237}\text{Np}$ ($t_{1/2}=2.14\times10^6$ a).
  - $^{237}\text{Np}$ co-production has to be minimal!
- It is neither anthropogenic nor primordial in occurrence and can thus function as an isotope dilution tracer.
- Current world-wide $^{236}\text{Np}$ stockpile limited to 10’s of $\mu$g of material.
- U.S. interagency Neptunium working group* (currently 10 members) formed in August 2015 to coordinate Np production R&D and address metrology community needs and purity requirements.
- $^{236}\text{Np}$ is an NSAC-I (2015) recommended isotope associated with “research opportunities in the physical sciences and engineering [...] where a shortage [...] is a challenge”
- $^{236}\text{Np}$ is on the DHS “High Priority” List for future funding.
- Data for production is extremely sparse!

Measurements at the LBNL 88-Inch cyclotron: “thick-target” $^{235}\text{U}(d,n)^{236m}\text{Np}$ and “thin target” $^{238}\text{U}(p,3n)^{236m}\text{Np}$ cross sections

Beam profile measured Using GAFChromic film

The $^{235}\text{U}$ sample was “overfilled” with beam

Stacked Target Holder

Deuterons @ 12, 16 MeV, Protons @ 27 MeV

HPGe counter
Post-irradiation counting shows the 642.3 keV γ-ray from the decay of the $^{236m}$Np isomeric state ($t_{1/2} = 22.5$ h)

- $159.4(52)$ fg $^{236m}$Np
- $89.98(20)$ fg/uAh·mg
- $554(21)$ fg $^{236m}$Np
- $9.265(3)$ fg/uAh·mg

Multiple channels observed via γ-spec

From $^{240}$Pu

Quantification of $^{236g}$Np / $^{237}$Np at LANL

- Quantification of $^{236g}$Np / $^{237}$Np production requires chemical workup and ICP-MS – based analysis, due to lifetimes ($t_{1/2}=1.5\times10^5$ y, $2.14\times10^6$ y, respectively).
- The $\approx200$ mg 93% $^{235}$U samples irradiated with 12 and 16 MeV deuterons underwent analysis at LANL in Spring 2019.
- **Optimal $^{236g}$Np production rates** (assuming a 550 mg/cm$^2$ $^{235}$U target):
  - $17.01(8)$ pg/uA•hr for 16 MeV.
  - $1.78(1)$ pg/uA•hr for 12 MeV.
- $^{236}$Np:$^{237}$Np production ratio (atom basis):
  - $1.05(38)$ for 16 MeV
  - $1.55(18)$ for 12 MeV.
  
  *No $^{236}$U produced!*
- $^{236}$Pu from $^{236m}$Np observed via $\alpha$-spec
- $^{238}$U(p,3n) targets awaiting ICP-MS

![ICP-MS data for (d,n) following U separation](image)

**LANL $\alpha$-spect results**

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{236}$Pu (fg)</th>
<th>$^{238}$Pu (fg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Un-irradiated Target</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>UO-1</td>
<td>75.7(5)</td>
<td>96(2)</td>
</tr>
<tr>
<td>UO-2</td>
<td>305(2)</td>
<td>630(8)</td>
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</tbody>
</table>
Next Steps

- Goals met so far:
  - Irradiation and γ-spectroscopy at LBNL
  - Chemical/Mass Spec analysis at LANL for $^{236g}$Np
  - Up to 40 ng can be made in 10 days @ 10 µA
  - Significant $^{237}$Np observed in the $^{235}$U targets
    - ~60% present in un-irradiated targets
    - Remainder co-produced from $^{238}$U(d,3n)
  - High-purity $^{235}$U targets needed for $^{235}$U(d,$\gamma$)$^{237}$Np characterization
    - 99.94% targets fabricated by LLNL (Gharibyan) with an irradiation planned for 2-3/21

<table>
<thead>
<tr>
<th>Target</th>
<th>Beam</th>
<th>Energy</th>
<th>$^{236m}$Np (γ-spec)</th>
<th>$^{236g}$Np (ICP-MS)</th>
<th>$^{236}$Pu (α-spec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}$U (93%)</td>
<td>D</td>
<td>12 MeV</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
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<tr>
<td>$^{235}$U (93%)</td>
<td>D</td>
<td>16 MeV</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
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<tr>
<td>natU</td>
<td>p</td>
<td>16-25 MeV</td>
<td>✓</td>
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<tr>
<td>$^{235}$U (99.4%)</td>
<td>D</td>
<td>14 MeV</td>
<td>✓</td>
<td></td>
<td>Scheduled for Feb-Mar 2021</td>
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</table>
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