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

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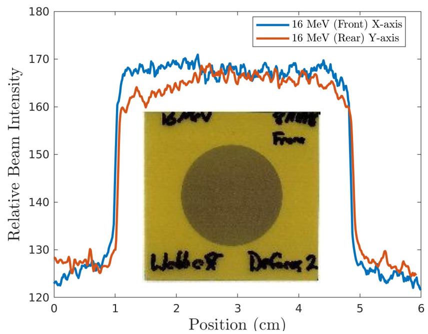
Isotope Dilution Mass Spectrometry: Standard Reference Material ^{236}Np

- Neptunium-236g ($t_{1/2}$=1.5X10^5 a) is used for IDMS determination of ^{237}Np ($t_{1/2}$=2.14X10^6 a).
  - *^{237}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}Np stockpile limited to **10's of µg** of material.
- U.S. interagency **Np working group*** (currently 10 members) formed in August 2015 to coordinate Np production R&D and address metrology community needs and purity requirements.
- ^{236}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}Np is on the **DHS "High Priority**" List for future funding.
- Data for production is extremely sparse!

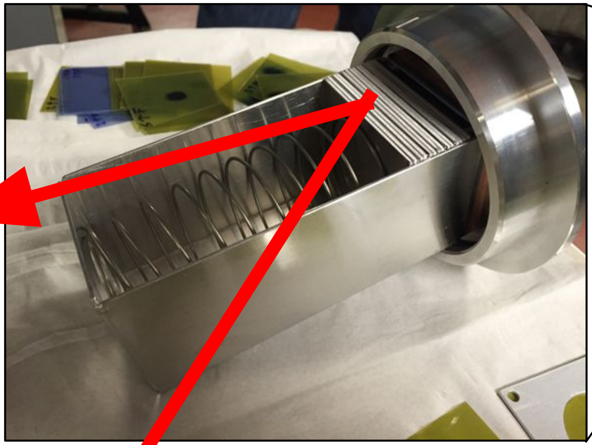
*SM Jerome, K Carney, R Essex, ME Fassbender, S Goldberg, M Kinlaw, SP LaMont, D Mackney, JJ Morrison, FM Nortier, Reference materials for neptunium determination, *Applied Radiation and Isotopes*, 126 (2017) 44-48.

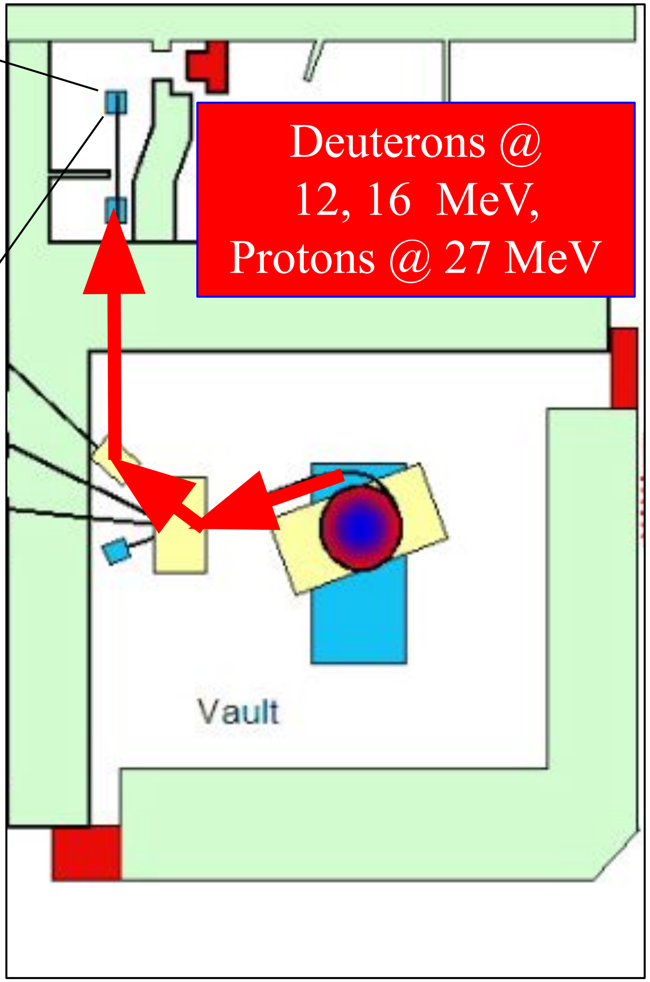
Measurements at the LBNL 88-Inch cyclotron: “thick-target” ^{235}U(d,n)^{236m}Np and “thin target” ^{238}U(p,3n)^{236m}Np cross sections

Beam profile measured Using GAFChromic film



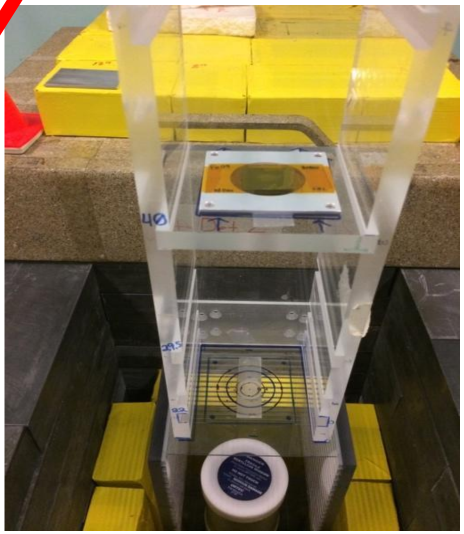
Stacked Target Holder



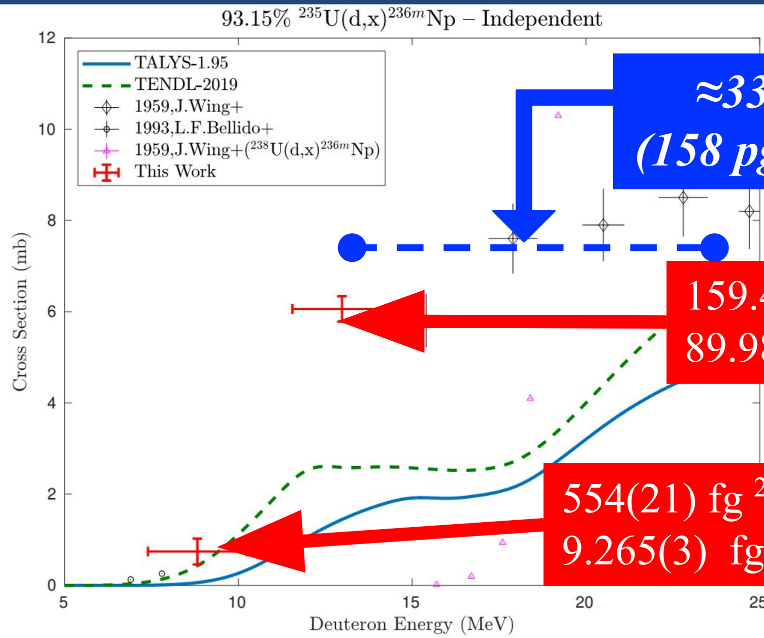


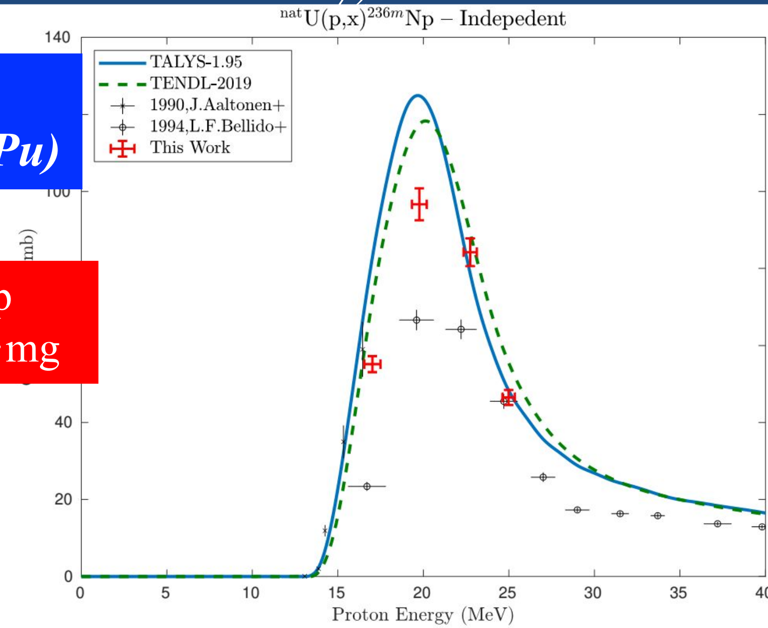
The ^{235}U sample was “overfilled” with beam

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)

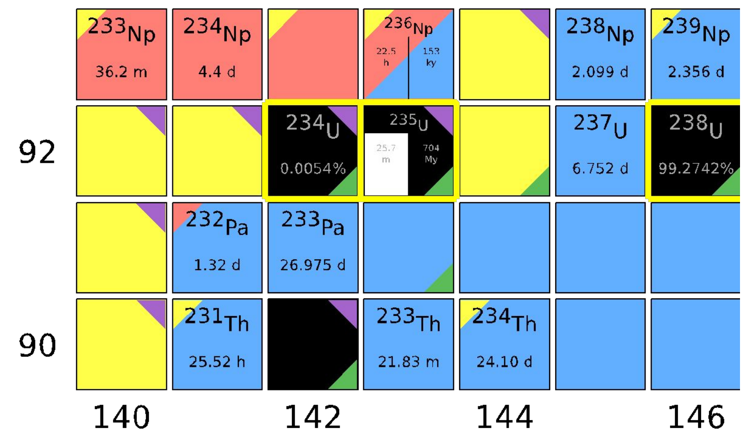




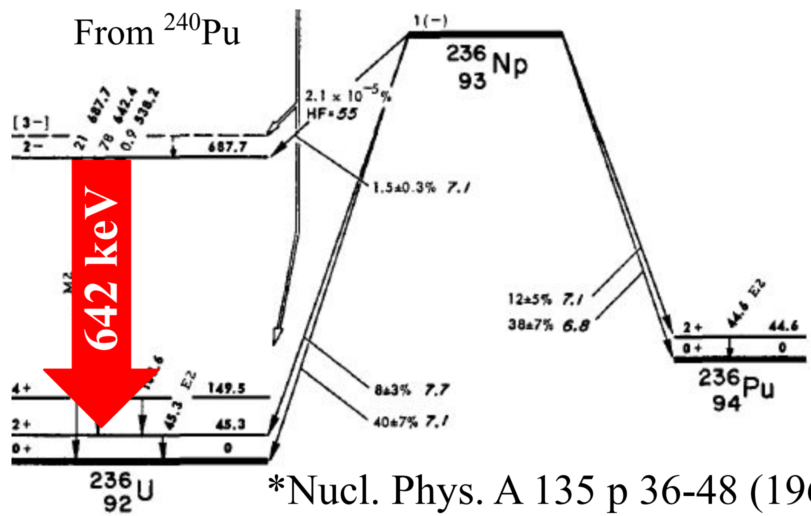
≈335 pg/μA•day
(158 pg/μA•day ^{236}Pu)

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



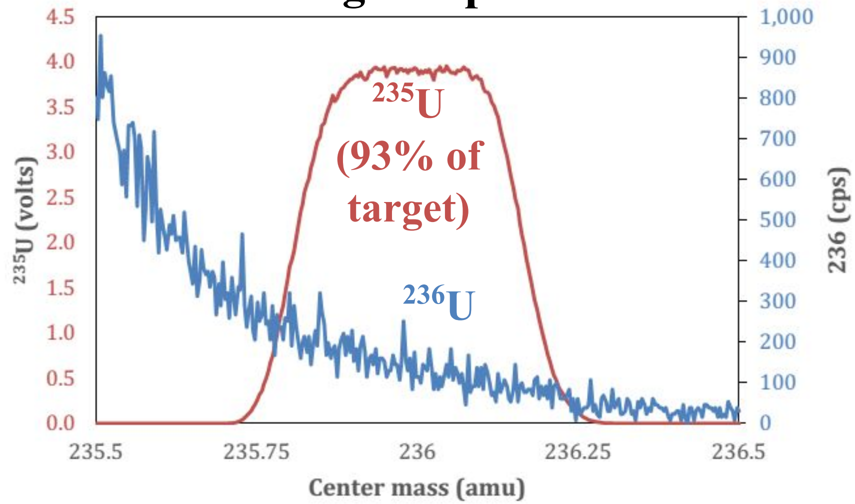
*Nucl. Phys. A 135 p 36-48 (1969)

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, 2.14×10^6 y, respectively).
- The ≈200 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 α-spec
- ^{238}U(p,3n) targets awaiting ICP-MS

ICP-MS data for (d,n) following U separation



LANL α-spect results

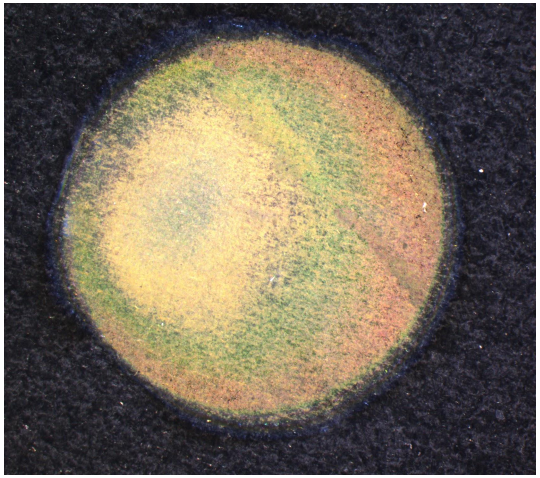
Sample	^{236}Pu (fg)	^{238}Pu (fg)
Un-irradiated Target	—	—
UO-1	75.7(5)	96(2)
UO-2	305(2)	630(8)

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,γ)^{237}Np characterization
  - 99.94% targets fabricated by LLNL (Gharibyan) with an irradiation planned for 2-3/21



12 targets made by LLNL with ρR_{areal} from 80-160 mg/cm^2

Target	Beam	Energy	^{236m}Np (γ-spec)	^{236g}Np (ICP-MS)	^{236}Pu (α-spec)
^{235}U (93%)	D	12 MeV	✓	✓	✓
^{235}U (93%)	D	16 MeV	✓	✓	✓
^{nat}U	p	16-25 MeV	✓		
^{235}U (99.4%)	*D*	*14 MeV*	*Scheduled for Feb-Mar 2021*		

Collaborators

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