INL/CON-23-75792-Revision-0

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Idaho National Laboratory

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Final Edge of Poster

Recovery of Neptunium-236g from Photon and Proton-Irradiated Actinide Targets

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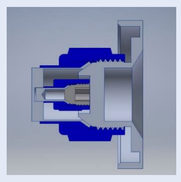
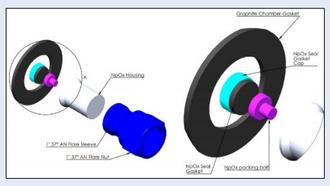
Introduction

Neptunium-236g is a rare radionuclide used as a tracer for neptunium-237 analyses. The availability of ^{236}gNp is limited and the viable production routes are costly, time consuming, and only produce trace quantities of the desired product. For this work, two known production methods were tested to determine product recovery, purity, and viability for use as a tracer. The first method utilizes a photon-irradiated ^{237}Np target to produce ^{236}gNp by the ^{237}Np(ɣ,n)→^{236}Np reaction. The second method utilizes the ^{238}U(p,3n) →^{236}Np reaction. These production routes were evaluated previously, and the former was considered ineffective without isotope separation and the latter was not well-characterized for the ^{236}mNp/^{236}gNp production ratio. Recent resurgence of electromagnetic isotope separation technology has enabled at least partial recovery of ^{236}gNp from part-per-million abundance feeds produced by the photonuclear reaction. To address the lack of production data for the second method, a proton-irradiated depleted uranium target was chemically processed to recover and purify the Np for abundance and ratio analyses. The status and current analytical results for each production method are presented.

Photon Irradiated ^{237}Np





- 0.5 g NpO_2
  - Test Sample
- Irradiated June 2018- Idaho State University Idaho Accelerator Center
  - 5 hours at 38 MeV
  - ~5x10^{14} total fissions
  - Contact dose (+8 days): 450 mR/hr
  - ^{236m}Np: 97 ± 9 mCi (measured)
  - ^{236g}Np: ~0.17 µg (estimated)
  - ^{236}Pu: 97 ± 9 mCi (measured)

- 2.5 g NpO_2
  - Production Sample
- Irradiated May-July 2020- Idaho State University Idaho Accelerator Center
  - 5 hours at 38 MeV
  - ~5x10^{14} total fissions
  - Contact dose (+8 days): 450 mR/hr
  - ^{236g}Np: ~100 µg (estimated)
  - ^{236}Pu: 97 ± 9 mCi (measured)

Proton Irradiated ^{238}U

-32 g depleted uranium target irradiated at Los Alamos National Laboratory Isotope Production Facility.

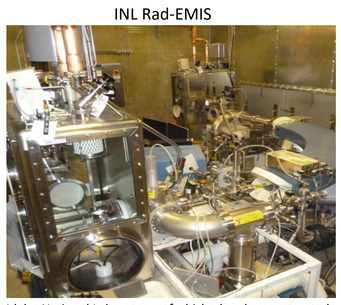
-Proton irradiation (^{238}U (p,3n)) for 103.9 hours to produce an estimated 44 µg ^{236g}Np.

-The target was shipped to INL for chemical processing to recover the and analyze Np to determine ^{236}Np:^{237}Np ratio.

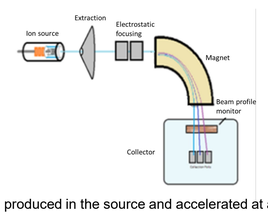
-Possible feed for isotope separation pending ratio results.

Electromagnetic Isotope Separation

INL Rad-EMIS



Idaho National Laboratory refurbished and reconstructed two 90-degree sector isotope separators[1,2]. The first one competed was originally designated for stable isotope production but is now used primarily as a non-radioactive platform to test methods and components. The second separator is currently designated for radioisotope enrichment and is undergoing significant component upgrades under the stewardship of the U.S. Department of Energy Isotopes Program (DOE-IP). The primary purpose of this effort is to establish a standing capability to produce high purity, highly enriched radioisotope spike solutions that can be used as reference materials for isotope dilution mass spectrometry (IDMS) analyses.



-Ions are produced in the source and accelerated at a variable potential of 30-40 kv.

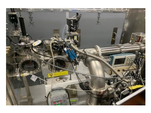
-An extraction lens referenced to the acceleration potential, as well as electrostatic focusing lenses shape the ion beam before it enters the magnet.

-The magnet separates isotopes according to mass.

-The separated beam is monitored via profilometer and Faraday cup measurements.

-Ion species from a given feed material can be determined by scanning mass ranges by adjusting the magnetic field.

Upgrades:
Collector chamber and focusing lens



Old collector chamber

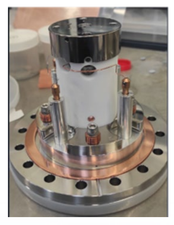
New collector chamber

Quadrupole lens: DTE Research and Design, LLC.

Source Material Preparation

Electrochemical plating cell

Electrodeposition of Np metal



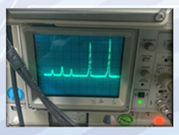
Liquid metal ion source- AuSiEr load.
Constructed by IonOptika Inc.

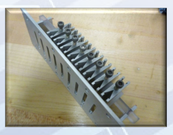
-The LMIS is typically used in FIB instruments and ion thrusters.
  Low M.P. metals such as Ga and In are typically used as feed.
    -Ga: 30º C
    -In: 157º C
    -Np: 644º C
    -Pu: 639º C
-Relatively low beam currents but high efficiency and run times.
  -Up to 20µA
  -Efficiency >50%
  -Can operate for several hundred hours
-Drawbacks
  -Basic design is simple but requires sharp, specialized W needle emitter
  -Difficult to load
  -Adaptation to actinides

Recovery and Analyses

-Isotopically separated nuclides are collected simultaneously on V-shaped Faraday cup collectors fitted with high purity foils (Al, Ti, W, Au).

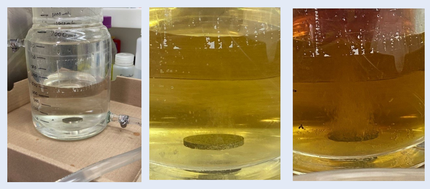
-The foils are removed, and the product is leached into acid solution to be analyzed for purity and enrichment by ICP-MS.

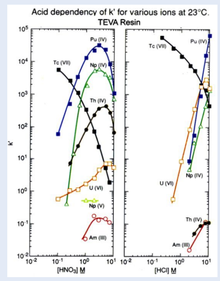




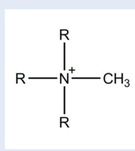
Chemical Separation

- **Surrogate tests**: Small scale batch contacts with TEVA showed:
  - Fast kinetics – 5 min adequate to attain equilibrium
  - Np uptake is very high: 96.6 %
  - Optimal uptake from 3 M HNO_3
  - Np uptake decreases due to aging of blended recipe, i.e. it is best to perform separation 1 hour after blending all ingredients
  - Recipe, scaled up to 1 L:
    - 500 mL dissolver solution
    - 20 mL of 1.5 M sulfamic acid with 20 mg of $Fe(NO_3)_3$
    - 100 mL of 1 M ascorbic acid
    - 380 mL of 0.01 M HNO_3
    - Wait 1 hour
    - Add 40 g TEVA resin
    - Run the resin contact
    - Am-243 added to trace Np

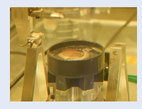


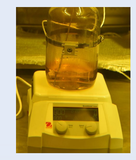


K' values, Eichrom Inc.



- **Irradiated Target Processing**:
  - Recovered target using "can opener"
  - Target was dissolved in hot cell
  - Dissolver solution was transferred out of cell for subsequent processing in fume hood
  - Aliquots were taken from each fraction to determine Np recovery and other radionuclides present.
  - Samples submitted for HPGe and MC-ICP-MS analyses (results are pending).





Conclusions and Future Work

The ^{237}Np(ɣ,n)→ ^{236}Np production route yielded less product than predicted with a total yield of approximately 6 micrograms of ^{236}Np. Currently, this feed is considered inadequate for a mass separator feed. Pending instrument upgrades with enhanced optics and a liquid metal ion source tailored for actinide feeds, this production route may be feasible with longer feed irradiation time, a different target geometry and chemical matrix, and with modification to the accelerator end station. The ^{238}U(p,3n) →^{236}Np production route is currently the most practical and economical. Isobaric interferences can be greatly reduced by chemical separation, however isotopic enrichment is still challenging considering the low quantities produced. Predicted yields for this production route were approximately 44 micrograms. Tracer analyses with yields over 90% indicate that approximately 30-40 micrograms of ^{236g}Np will be isolated from the DU target, depending on the required polishing steps for chemical purification. ^{236}Np/ ^{237}Np ratio analyses are underway. Results from this work will determine a path forward for reference material production and future irradiation efforts.

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