INL/CON-21-65507-Revision-0

Production of Xenon-135 from isotopically enriched Xenon-134 and Xenon-136 Targets

April 2022

Jared J Horkley, Jacob L Brookhart, Edna S Cardenas, Kevin P Carney, Tracy P Houghton, Troy A Robinson, Matthew G Watrous

Changing the World's Energy Future

INL is a U.S. Department of Energy National Laboratory operated by Battelle Energy Alliance, LLC

INL/CON-21-65507-Revision-0

Production of Xenon-135 from isotopically enriched Xenon-134 and Xenon-136 Targets

Jared J Horkley, Jacob L Brookhart, Edna S Cardenas, Kevin P Carney, Tracy P Houghton, Troy A Robinson, Matthew G Watrous

April 2022

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

Production of Xenon-135 from Isotopically Enriched Xenon-134 and Xenon-136 Targets

J.J. Horkley*[1], J.L. Brookhart[1], E.S. Cardenas[1], K.P. Carney[1], C. Hines[2], T.P. Houghton[1], T.A. Robinson[1], M.G. Watrous[1]
1) Idaho National Laboratory 2) Washington State University
*Corresponding author

Goal: Explore alternative methods to produce monoisotopic xenon reference standards for atmospheric CTBTO monitoring.

Overview

- Atmospheric radioxenon analyses conducted under the Comprehensive Test Ban Treaty Organization (CTBTO) necessitates the collection and analysis of air samples to quantify ^{135}Xe, an indicator for nuclear explosions and reactor operations.
- High quality, monoisotopic reference standards that are typically short lived, are used for instrument calibration and must be produced and shipped promptly to monitoring stations worldwide.
- This work aims to develop a method to produce pure ^{135}Xe, minimizing ^{133}Xe contaminants that are often generated during production.

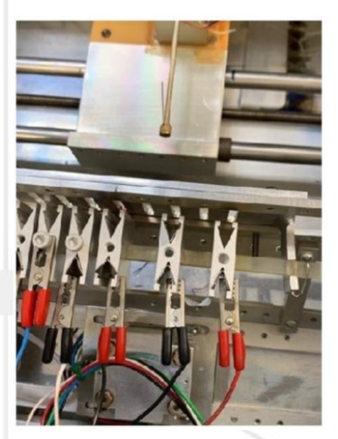
Approach

- Electromagnetic isotope separation is used to produce monoisotopic targets of ^{134}Xe and ^{136}Xe for accelerator or reactor irradiation to produce the intended product.
- This work examines 1) the preparation of ^{135}Xe by neutron capture (n, γ) in a TRIGA reactor using a ^{134}Xe target and 2) ^{135}Xe production using high energy photon (photonuclear) irradiation (γ, n) of ^{136}Xe. Results from each process are compared for ^{135}Xe yield and purity.

Target Production

Stable isotope mass separator

-Scanditronix 1.5 m radius, 5 kGauss magnet
-6 Faraday cup collectors monitor beam current and collect stable Xe isotopes simultaneously.
- >99% first pass enrichment for ^{134}Xe and ^{136}Xe.

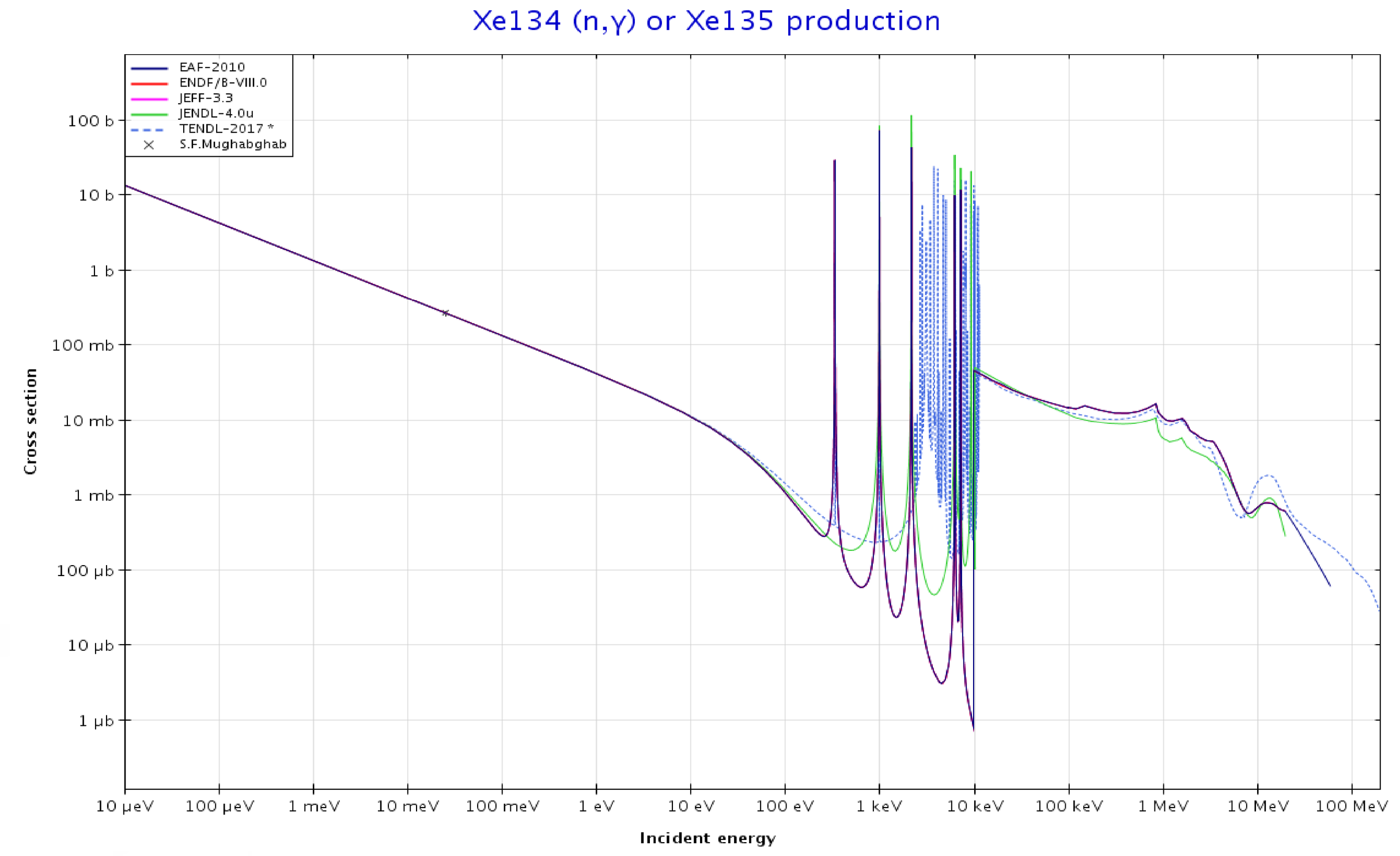


- ^{134}Xe and ^{136}Xe targets were produced from natural Xe feed gas.
- Xe ions were generated in a thermal type source based on the "Colutron" design from Beam Imaging Solutions Inc. The source is floated at the acceleration potential (35-40 kv) and operates steadily producing 1-3 μA of current. Accelerated ions pass through a bending magnet and are separated by mass and collected on high purity v-shaped aluminum foils mounted to individual Faraday cup detectors.
- Total ion collection for ^{134}Xe (n, γ) experiment: 1.5 μAh or ~ 0.43 μg ^{134}Xe. (Assumes 100% collection efficiency).
- Total ion collection for ^{136}Xe (γ , n) experiment: 1.3 μAh or ~ 0.37 μg ^{136}Xe. (Assumes 100% collection efficiency).

Irradiations

^{134}Xe (n, γ):

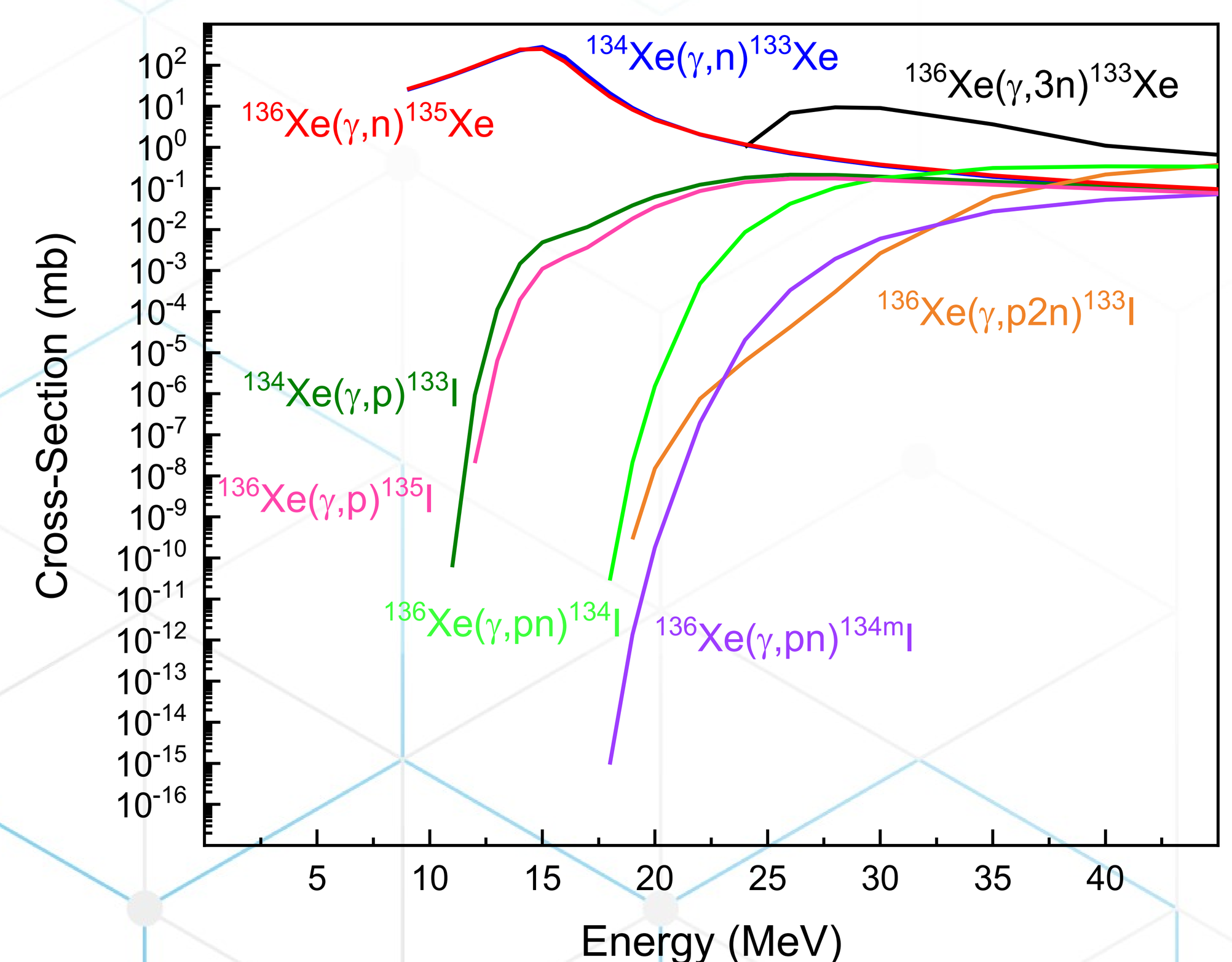
- ^{134}Xe target irradiated at Washington State University 1 MW TRIGA reactor for 6 hours to produce an estimated 2500Bq.
- 89 Bq ^{135}Xe produced indicating target mass was ~5 % of theoretical collection.
- ^{133}Xe detected after 48-hour decay. 133Xe/135Xe ratio: 0.0043 ± 0.004558



Examining data from previous irradiations and assuming a neutron flux of 1 *10^{13} n /cm^{2}s, estimated average cross section for the ^{134}Xe(n,γ)^{135}Xe reaction in the WSU light water reactor= 0.151 Barns.

^{136}Xe (ɣ, n):

- ^{136}Xe target irradiated at Idaho State University Idaho Accelerator Center's 25 MeV S-band linear accelerator for 6.5 hours at 21.8 MeV.
- Sample was allowed to cool and was measured by HPGe for approximately 47 hours.
- 592 Bq ^{135}Xe produced.
- ^{133}Xe was not detected in the sample.



Reaction Energy Thresholds
^{136}Xe(γ,n)^{135}Xe - 8.1 MeV
^{136}Xe(γ,3n)^{133}Xe - 23.0 MeV
^{136}Xe(γ,p)^{135}I - 10 MeV
^{136}Xe(γ,pn)^{134}I - 18 MeV
^{136}Xe(γ,p2n)^{133}I - 24.0 MeV
^{134}Xe(γ,n)^{133}Xe - 8.6 MeV
^{134}Xe(γ,p)^{133}I - 9.6 MeV
^{134}Xe(γ,pn)^{132}I - 17.8 MeV
^{136}Xe(γ,p3n)^{132}I - 32 MeV

Photonuclear cross-sections obtained from TENDL-2015. Reaction energy thresholds obtained from the National Nuclear Data Center.

Analysis

- Targets produced from the mass separator provide adequate near-monoisotopic samples for both reactor and photon irradiation experiments.
- Ion beams can be monitored with adequate accuracy, but the efficiency of collection is not well-known, and needs to be investigated further.
- Reactor-irradiated targets produced significant ^{133}Xe in the final product, likely from the ^{134}Xe(n,2n)^{133}Xe reaction from higher energy neutrons.
- Accelerator-irradiated targets yielded approximately 6x the product ^{135}Xe for the equivalent irradiation time and did not show measurable ^{133}Xe at the time of analysis.

Next steps

- Determine target masses as a function of integrated beam current by mass spectrometric analysis of unirradiated targets.
- Determine alternative target collection substrates.
- Compare irradiation efficiency of substrate-impinged target vs. gaseous target:

- Upscaled target production would require up to 1000x more target material and would require mA-level beam current and higher efficiency collection in the isotope separator.
- Efforts are ongoing to increase beam currents on the isotope separator.

Acknowledgements

-Idaho State Universities Idaho Accelerator Center
-Washington State University Nuclear Science Center
-Beam Imaging Solutions, Inc.
-DOE Funding: DE-AC07-05ID14517