Fission Product Yield Measurements of ^{239}Pu Irradiated at the USGS TRIGA Reactor

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Introduction

Understanding fuel utilization, nuclear forensics, and the behavior of aging weapons requires the use of fission observables to generate theoretical models. The uncertainty in current fission product yield (FPY) measurements restricts computational models to accurately predict the fission behavior at varying neutron energies.

Current FPY measurements suggest neutron-induced fission will favor symmetric fission as the energy of the incident neutron increases. Improving the nuclear database calls for additional FPY measurements to better understand this neutron energy-dependence. The production of valley nuclides, like Ag-111 and Cd-115, are particularly sensitive to the incident neutron energy and can help elucidate this behavior.

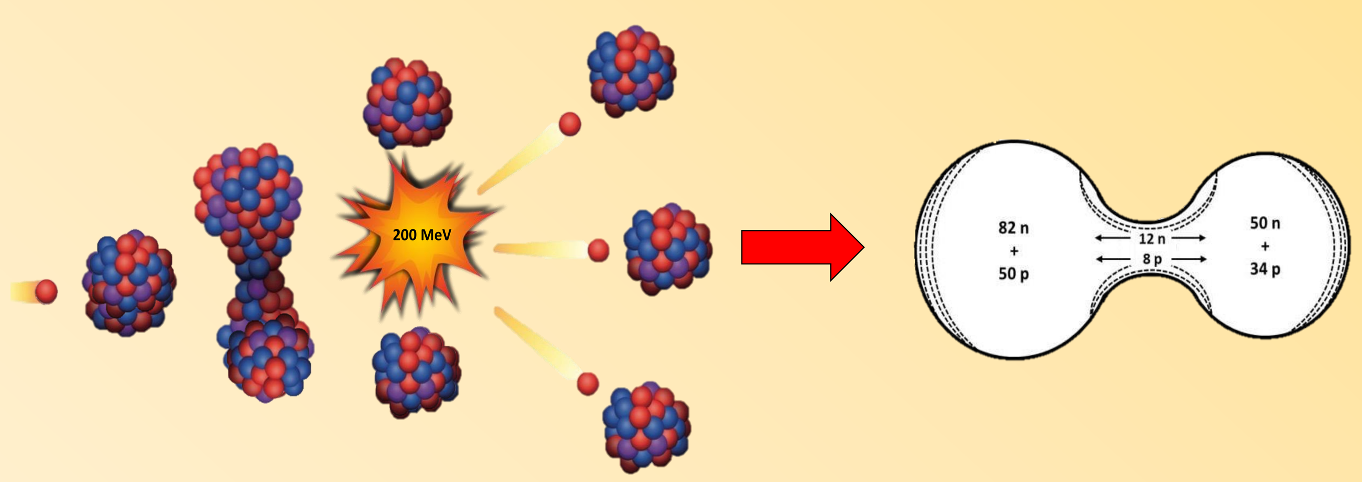


Figure 1: General process of a nucleus undergoing neutron-induced fission (left) and the preferred configuration during nuclear deformation (right).

Objective: Irradiate Pu-239 targets under distinct neutron environments to evaluate the neutron energy dependence of a fissioning Pu-239 nucleus. Measurements from these integral experiments aim to help reduce measurement uncertainties in the nuclear database used for nuclear applications.

Pu-239 Irradiations at GSTR

Figure 2: PuO_2 powder wrapped in Al foil (left) and neutron flux monitors mounted onto quartz tubing (right).

Two weapons-grade (type MT-52) Pu-239 targets were individually irradiated under distinct neutron environments.

Each PuO_2 target was wrapped in high purity aluminum foil and sealed in quartz tubing under argon gas.

The target was mounted alongside Ni and Au neutron flux monitors.

The target was mounted along the vertical centerline of a customized irradiation can and irradiated in the Central Thimble.

Bare irradiation: The target excluded any neutron shield and was exposed to the entire neutron spectrum.

Hardened spectrum: The target was wrapped in Cd to shield the target from thermal neutrons and expose the target to a harder neutron spectrum.

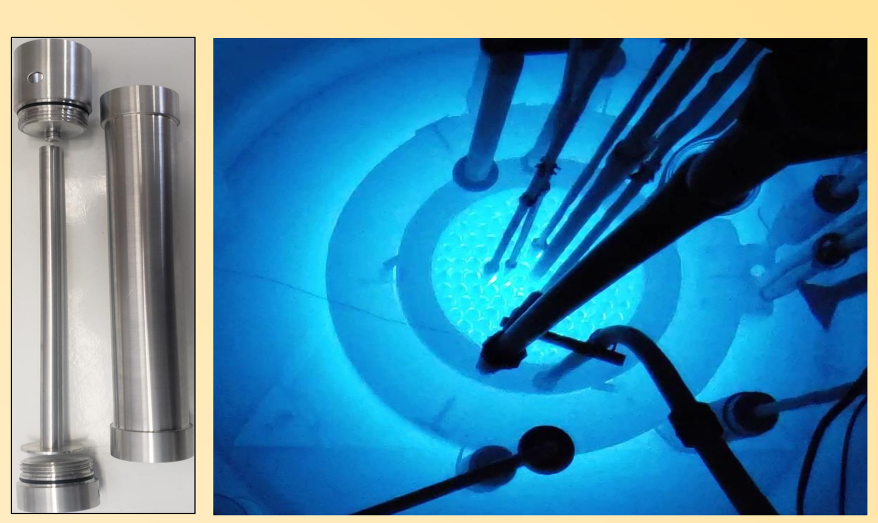
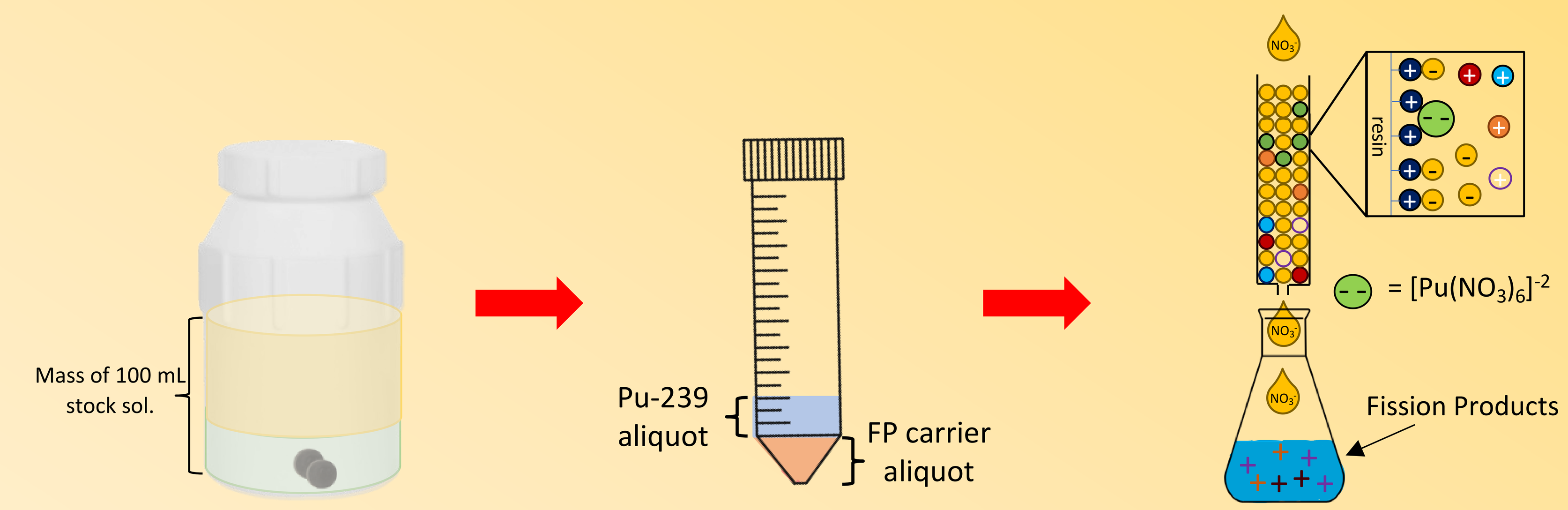


Figure 3: Customized Central Thimble irradiation can (left) Geological Survey TRIGA Reactor at 800 kW (right).

Radiochemical Separations



Target dissolution

Al-wrapped target was individually dissolved under a hot solution of 8M HNO_3/50mM HF. Once fully dissolved, the solution was brought up in 8M HNO_3 to generate a 100 mL stock solution.

Replicate preparation

An aliquot of the Pu-239 stock solution was spiked into centrifuge tubes containing spiked carrier solutions of select fission products.

Fission product purification

Each replicate was first purified from Pu-239 via anion-exchange in 8M HNO_3. The recovered fission products were then purified from byproduct material and separated from each other using established separation procedures.

Sample Deposition

Replicates were mounted via filtration or electrodeposition onto a filter paper or stainless steel planchette.

- Sample thickness: 0.95 mm to 1.07 mm
- Sample diameter: 17.15 mm (electrodeposition)
- Sample diameter: 17.60 mm (filtration)

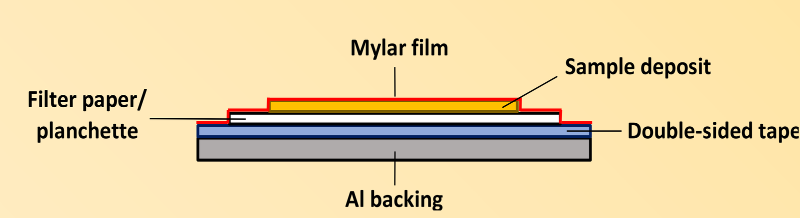


Figure 4: Sample mounting configuration for all replicates prepared.

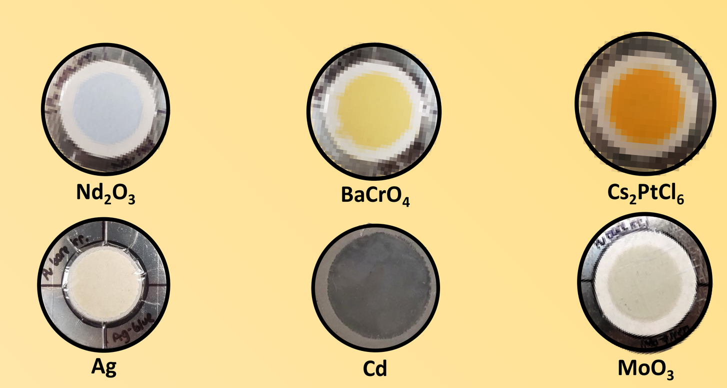


Figure 5: Deposition of each type of fission product measured.

HPGe efficiency corrections

Gamma-sensitive samples were counted on a high-purity germanium detector (HPGe). Button sources were used to generate an efficiency curve at the furthest sampling position (position 6).

Relative standard sources were prepared according to figure 4 to match the solid angle coverage of gammas emitted from the fission product replicates. These sources served to correct for prevalent geometry effects at close distances that are not accounted for with button sources.

Position 1

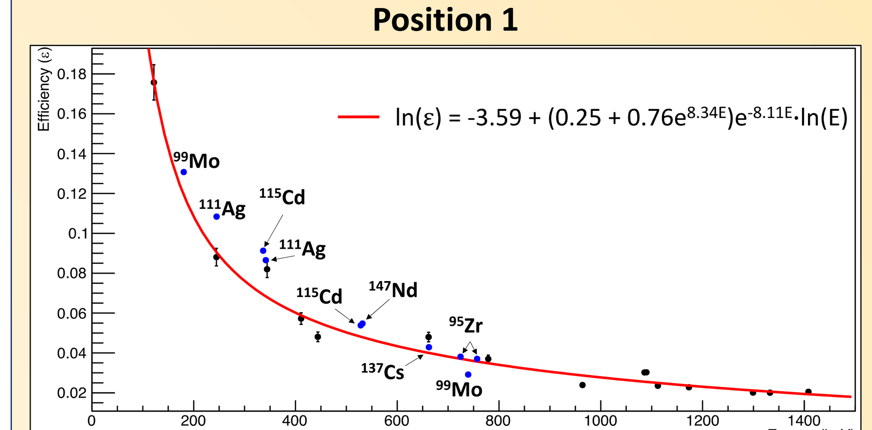


Figure 6: Comparison between the efficiency curve generated from button sources and relative standard sources at the closest position.

Position 6

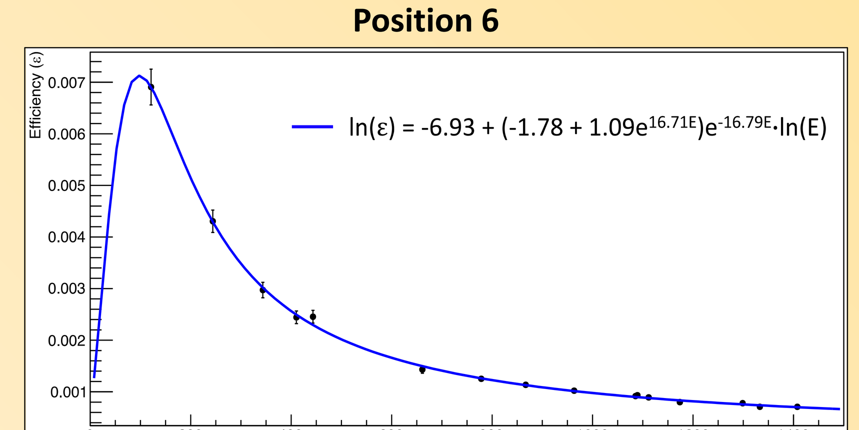


Figure 7: Efficiency curve generated from button sources at the furthest position.

Cumulative Fission Product Yields

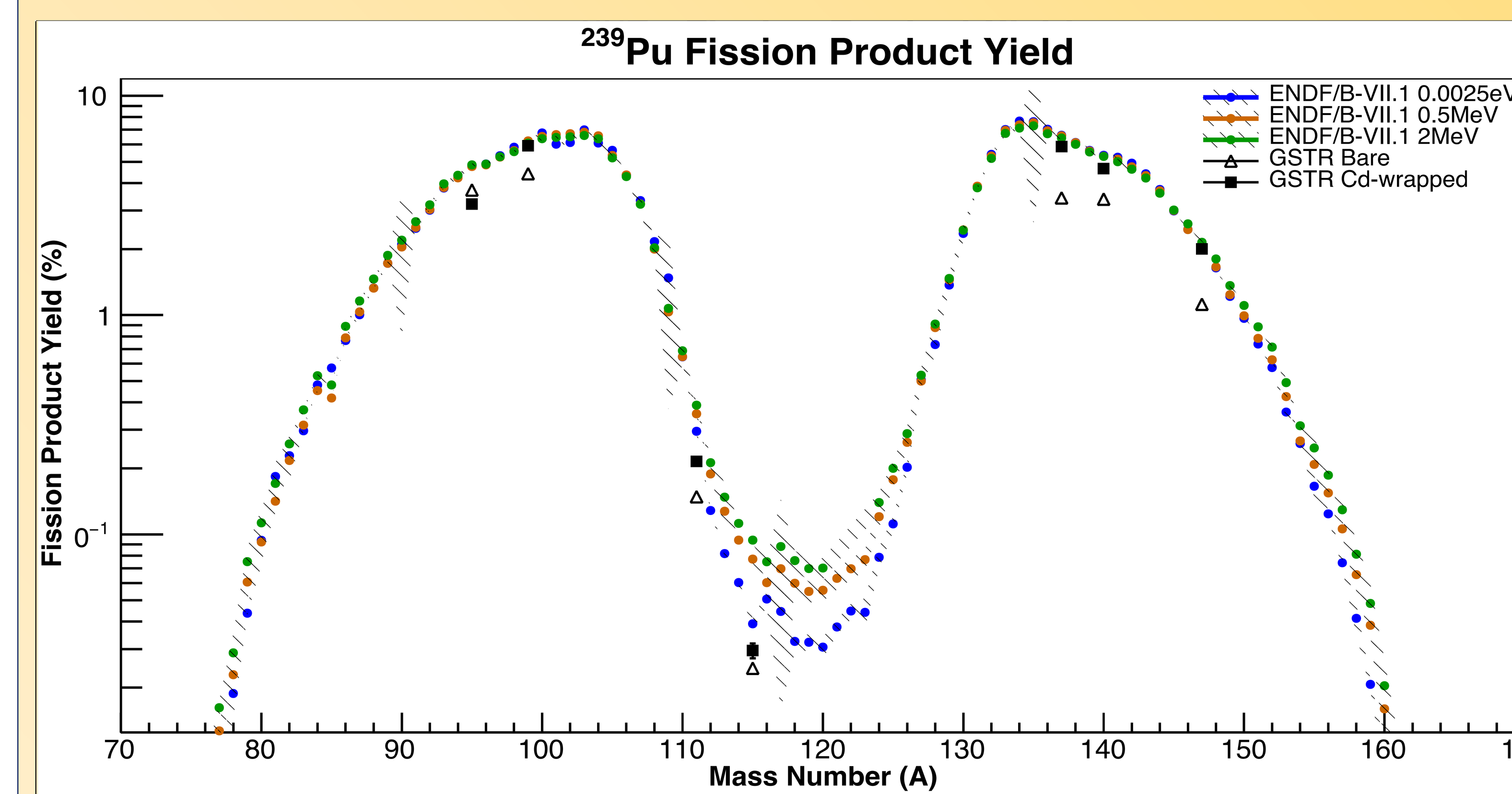


Figure 8: Calculated fission product yield measurements of select fission products for Pu-239

Table 1: Calculated fission product yields (%)

Fission Product	Bare	Cd-wrapped	0.0025 eV	0.5 MeV	2MeV
Zr-95	3.72(7)	3.21(5)	4.82(7)	4.76(3)	4.8(1)
Mo-99	4.41(6)	5.9(2)	6.21(9)	6.23(4)	6.1(2)
Ag-111	0.148(3)	0.215(3)	0.295(8)	0.355(7)	0.388(8)
Cd-115	0.0245(5)	0.030(2)	0.039(2)	0.077(3)	0.094(4)
Cs-136	0.054(4)	0.085(4)	0.1(6)	0.06(3)	0.06(4)
Cs-137	3.42(3)	5.9(2)	6.61(3)	6.57(5)	6.4(1)
Ba-140	3.4(1)	4.7(2)	5.35(7)	5.30(4)	5.3(2)
Nd-147	1.12(2)	2.00(8)	2.00(6)	2.01(1)	2.15(4)

Conclusions and Future Work

Using relative standards with the same isotopes as the select fission products allowed for proper corrections of the inherent summing effects that are unique to each fission product.

The experimental yields for both experiments were lower than anticipated despite exhibiting the expected trend with increasing neutron energy. The Pu-240 present in the type MT-52 targets may have reduced the overall neutron flux and contributed to the lower experimental yields.

Future work: measure the isotopic composition of the MT-52 Pu target using mass spectrometry and evaluate Pu-240's contribution towards the overall flux depreciation

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