



HOUGHTON
UNIVERSITY

The Effects of Radiation Escape on Accuracy and Precision in Isotopic Composition Determination of Uranium Samples with Decay Energy Spectroscopy

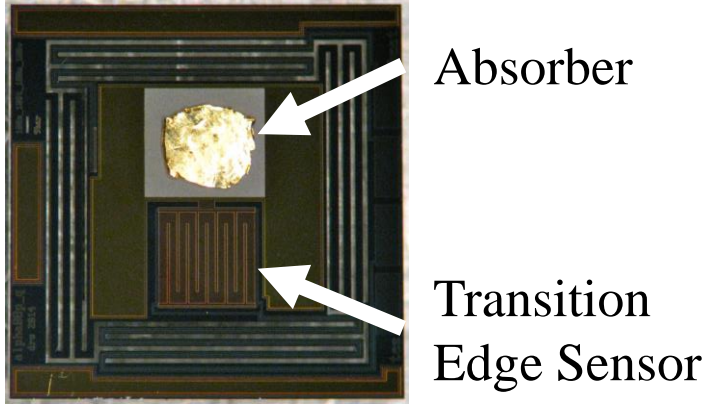
Timothy Ockrin
Houghton University
April 20, 2024

Nuclear Safeguards



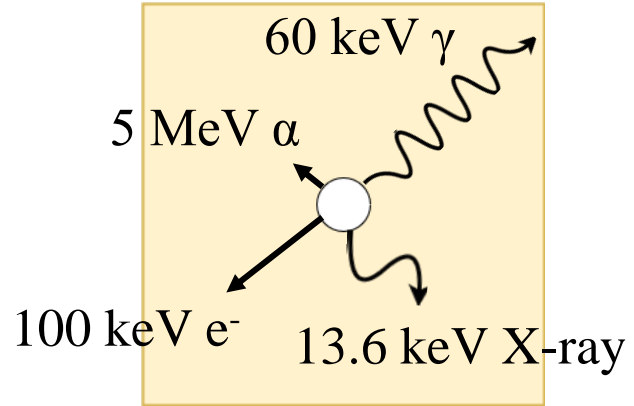
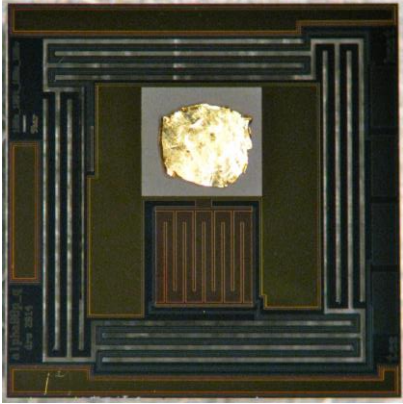
Require precise isotopic composition measurements of samples

Decay Energy Spectroscopy (DES)



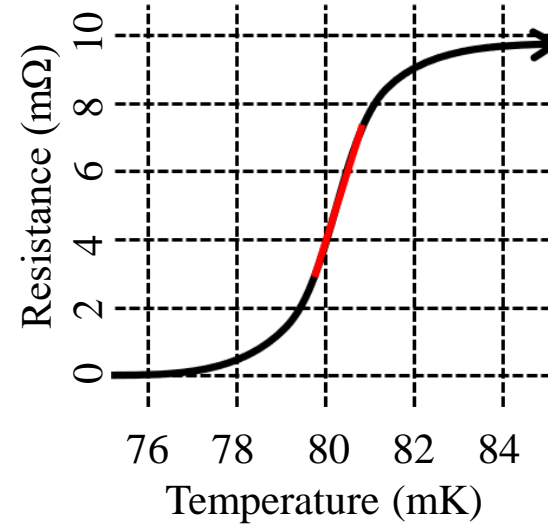
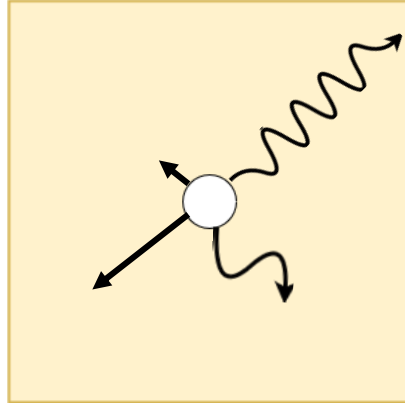
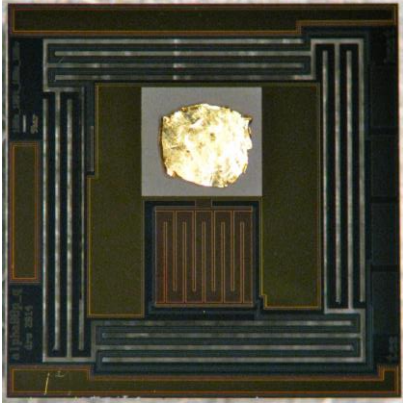
DES detector with gold absorber
thermally linked to sensor

Decay Energy Spectroscopy (DES)



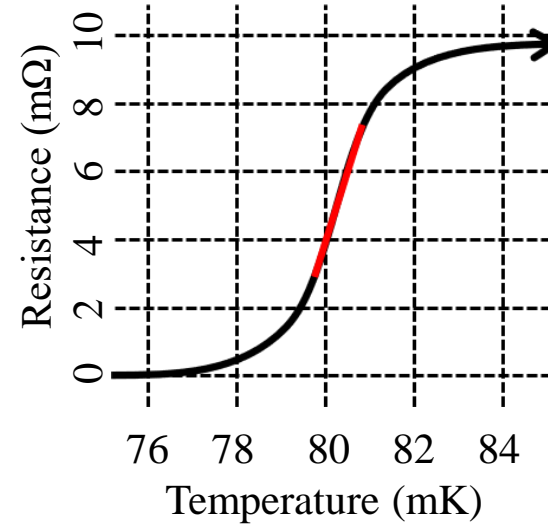
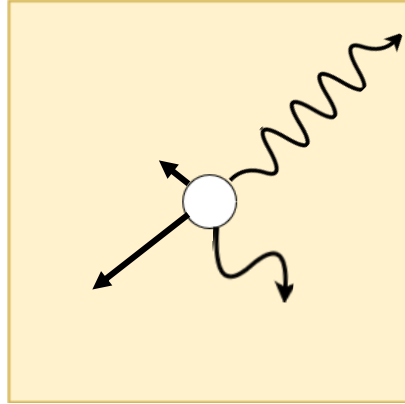
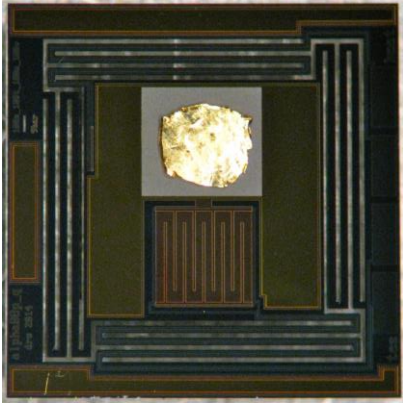
The energy of all particles released in a decay is thermalized simultaneously

Decay Energy Spectroscopy (DES)



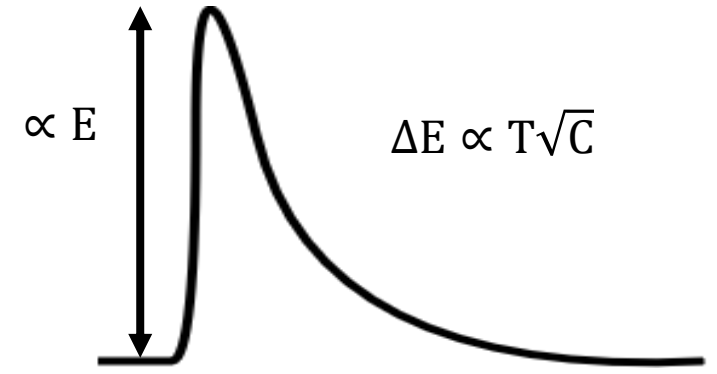
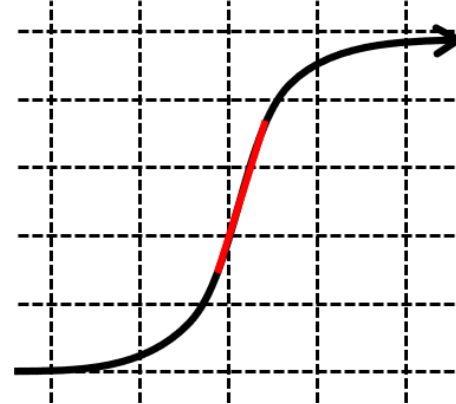
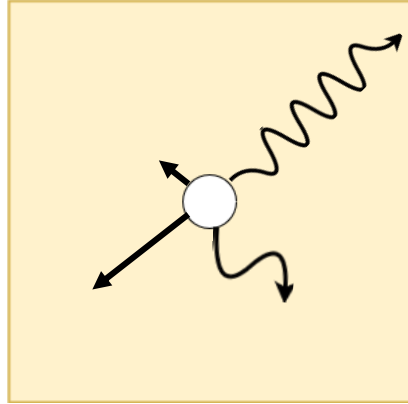
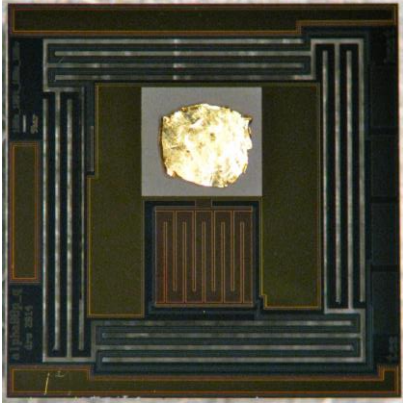
Sensor on transition between normal and superconducting

Decay Energy Spectroscopy (DES)



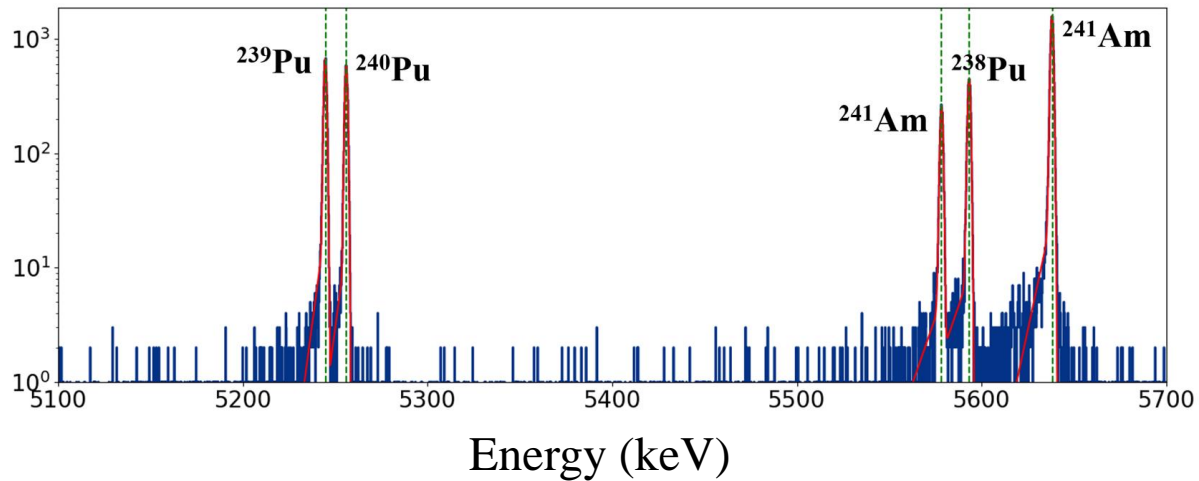
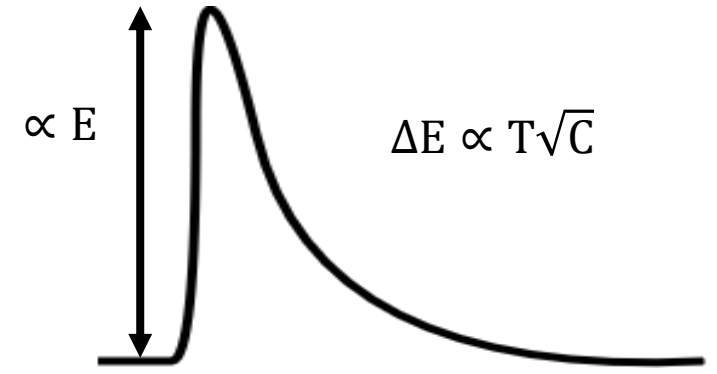
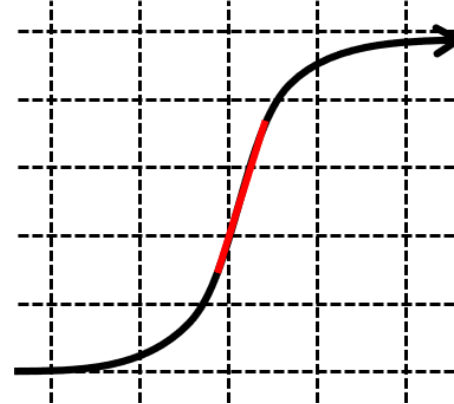
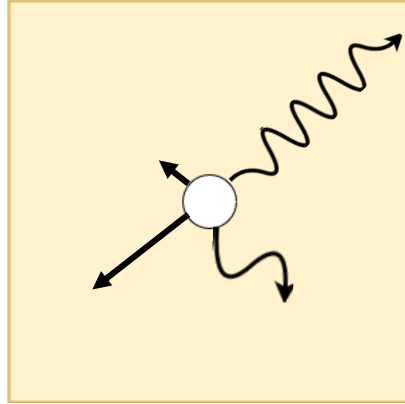
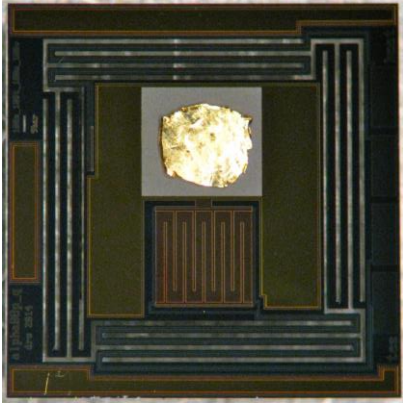
Sensor on transition between normal and superconducting

Decay Energy Spectroscopy (DES)



The height of a pulse is directly proportional to the energy of the decay

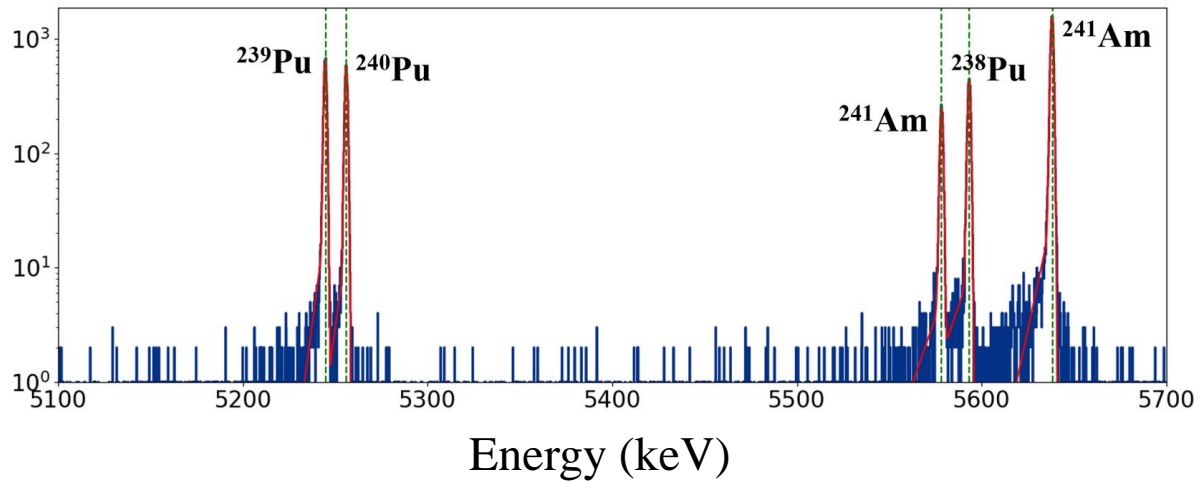
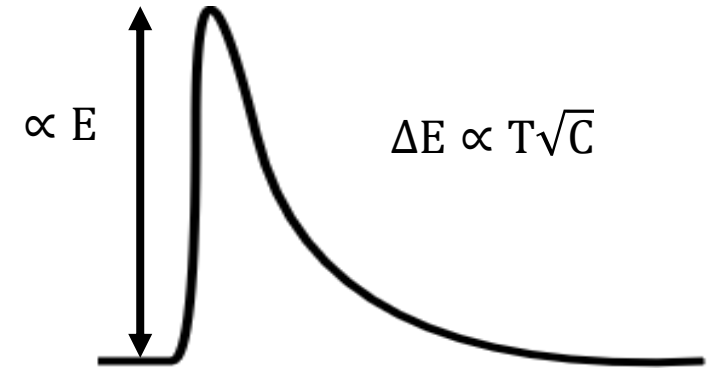
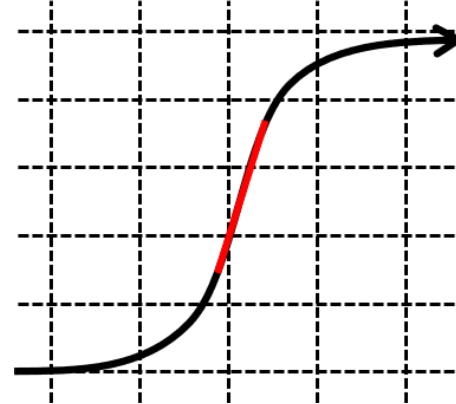
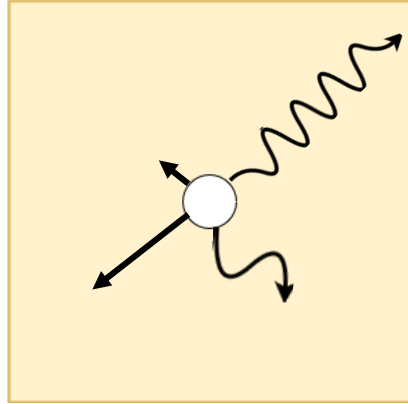
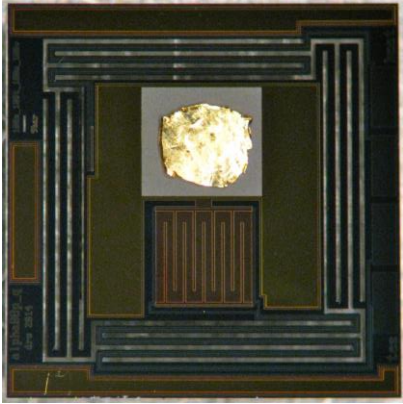
Decay Energy Spectroscopy (DES)



Advantages of Decay Energy Spectroscopy:

- Simplified Spectra

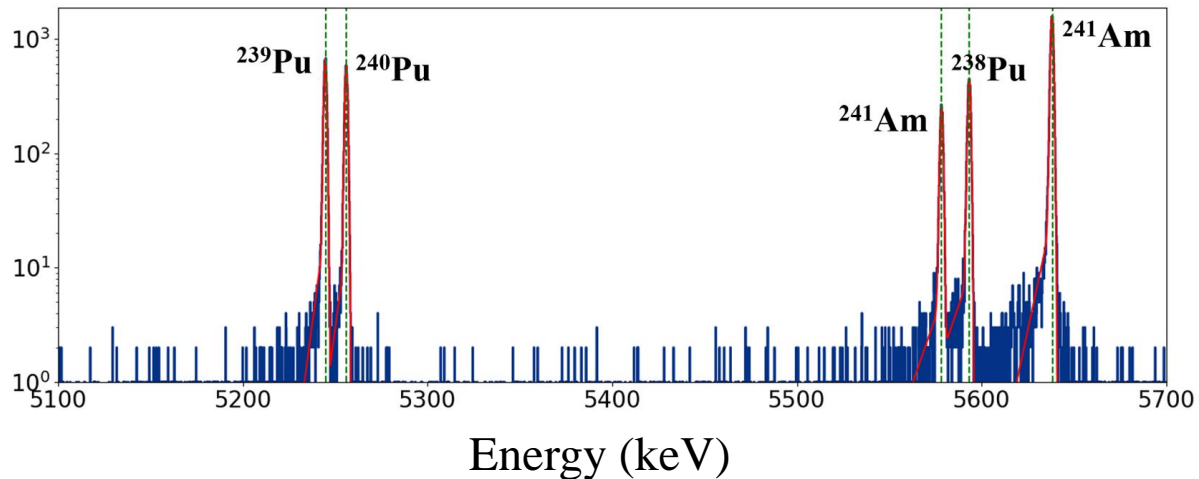
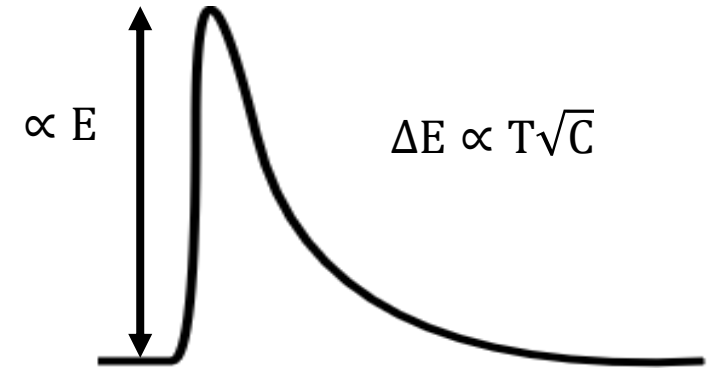
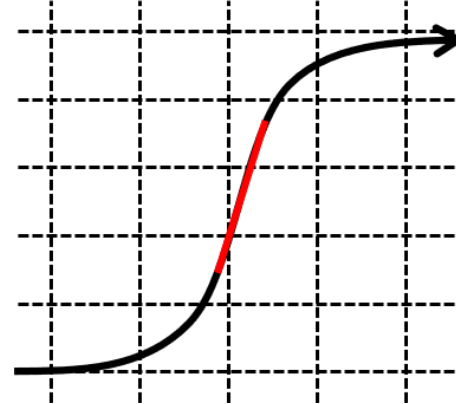
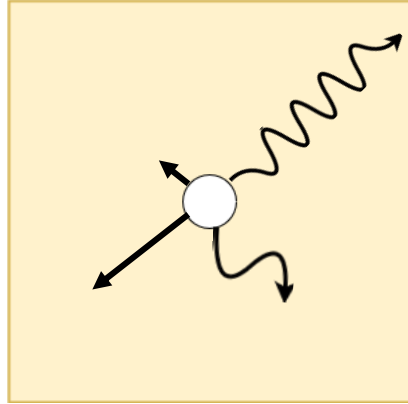
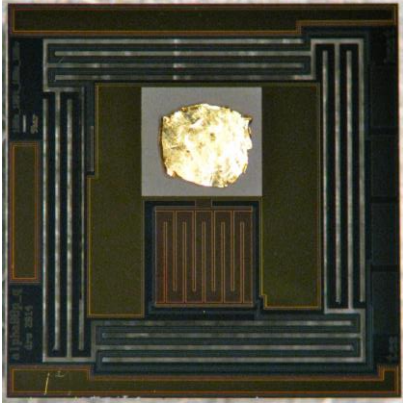
Decay Energy Spectroscopy (DES)



Advantages of Decay Energy Spectroscopy:

- Simplified Spectra
- High Energy Resolution (1-5 keV @ 5 MeV)

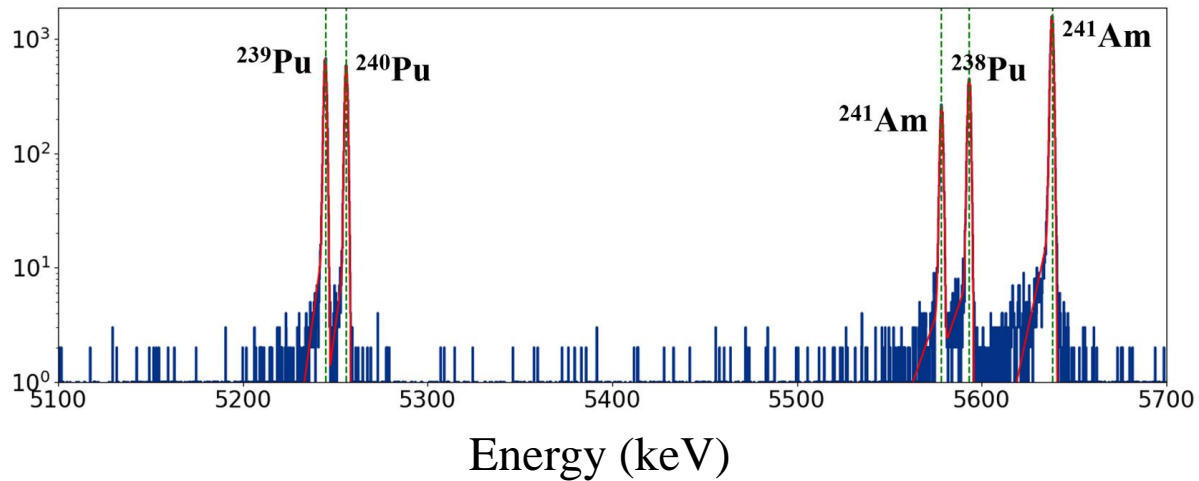
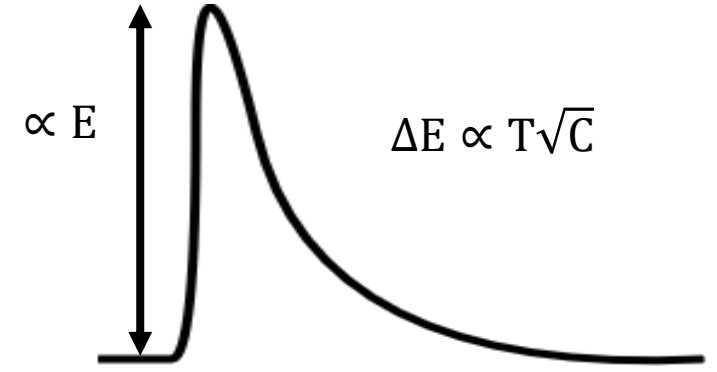
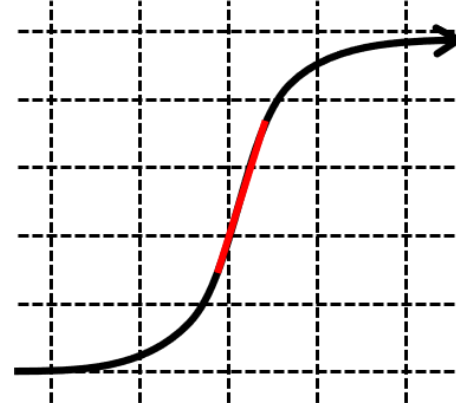
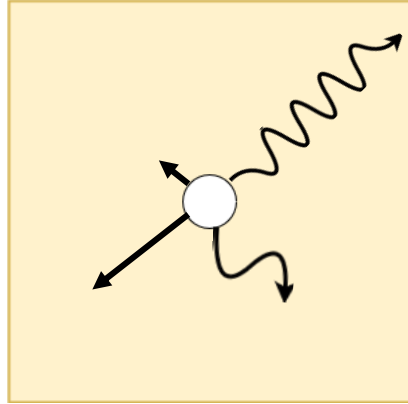
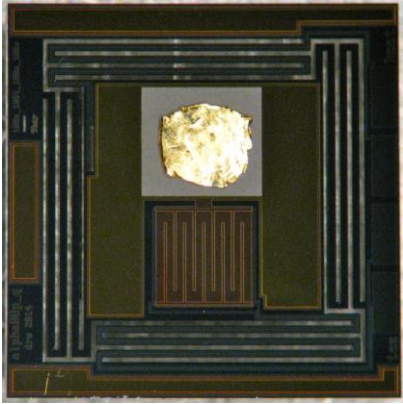
Decay Energy Spectroscopy (DES)



Advantages of Decay Energy Spectroscopy:

- Simplified Spectra
- High Energy Resolution (1-5 keV @ 5 MeV)
- Can measure several isotopes simultaneously without chemical separation

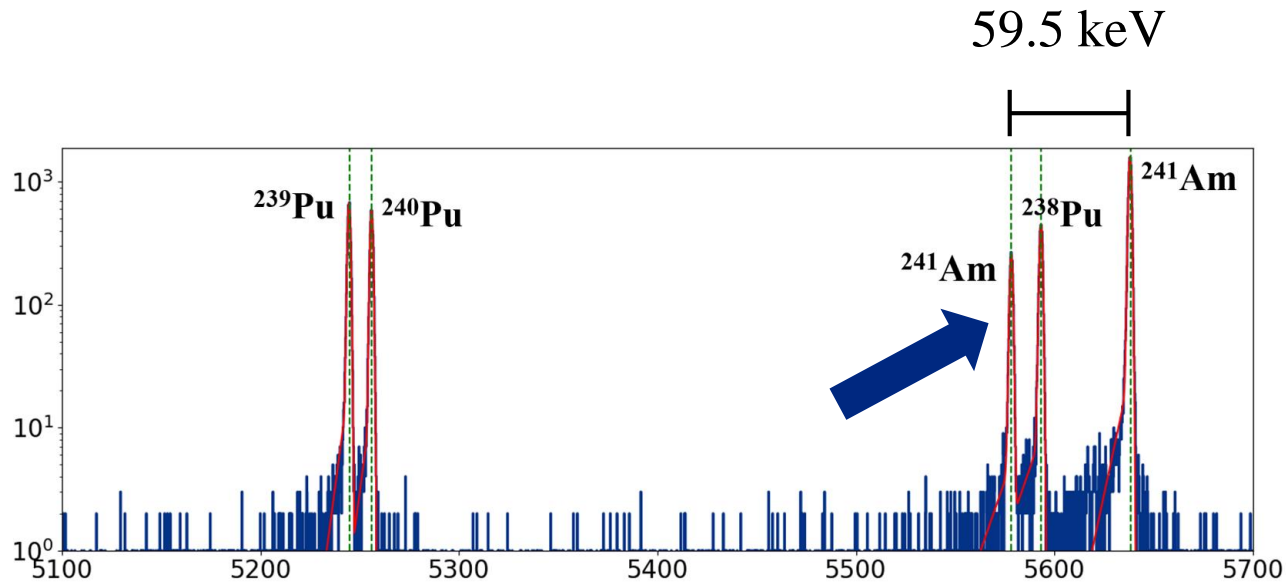
Decay Energy Spectroscopy (DES)



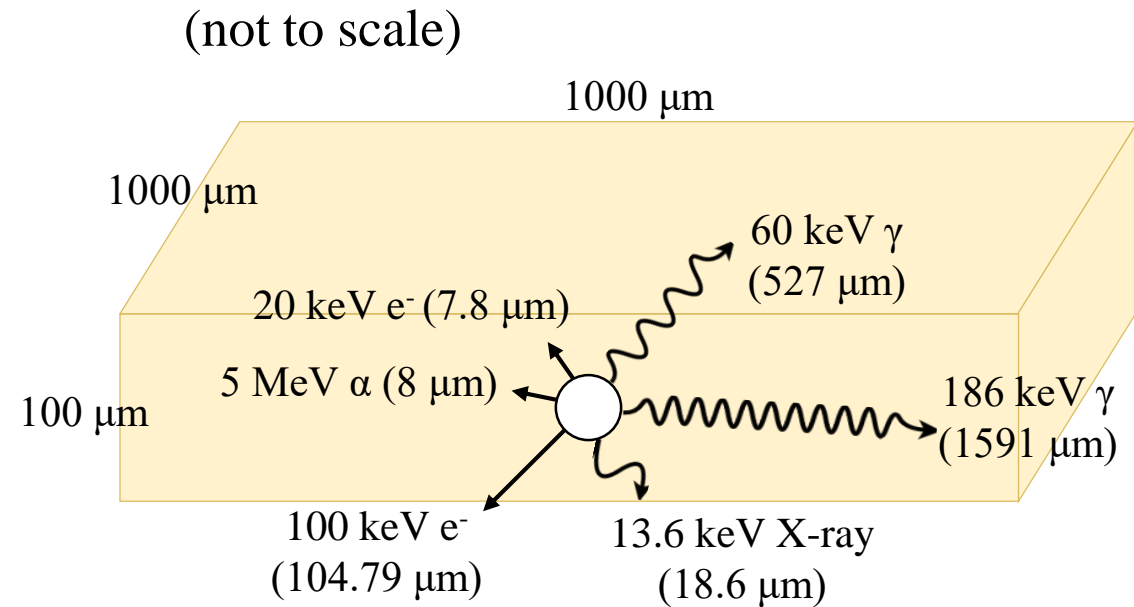
Advantages of Decay Energy Spectroscopy:

- Simplified Spectra
- High Energy Resolution (1-5 keV @ 5 MeV)
- Can measure several isotopes simultaneously without chemical separation
- Low activity requirement (~ 1 Bq)

Decay Energy Spectroscopy: Energy Escape

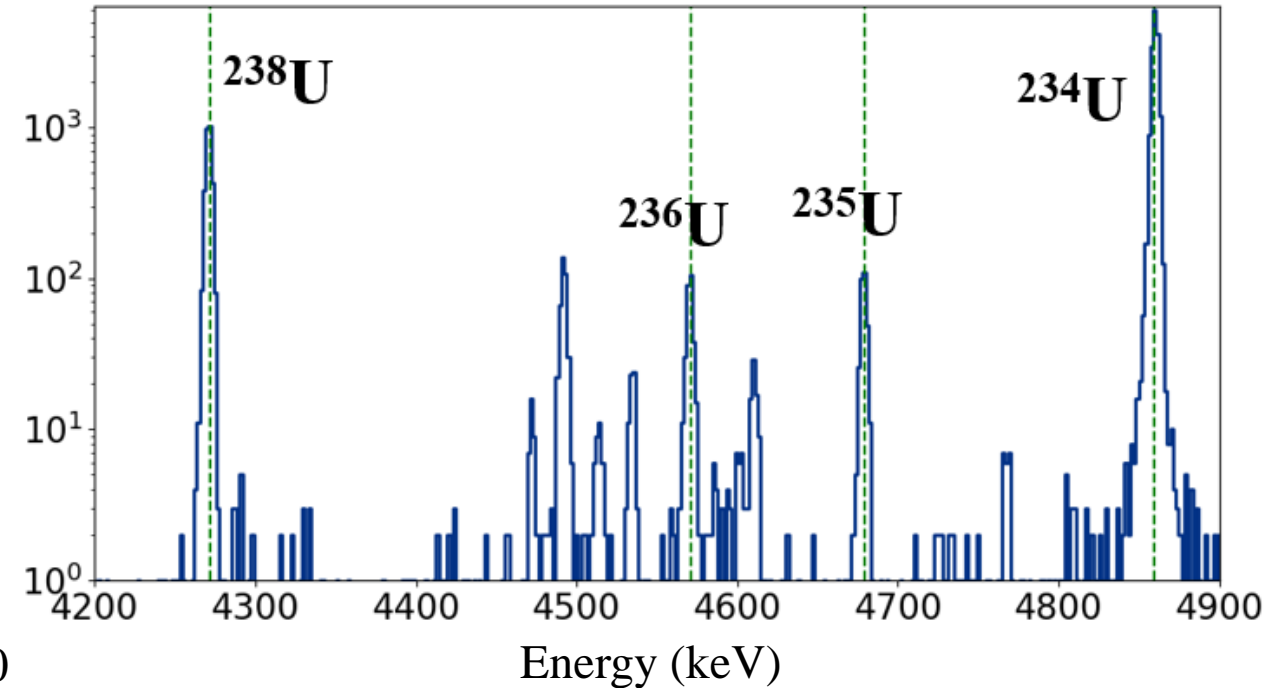
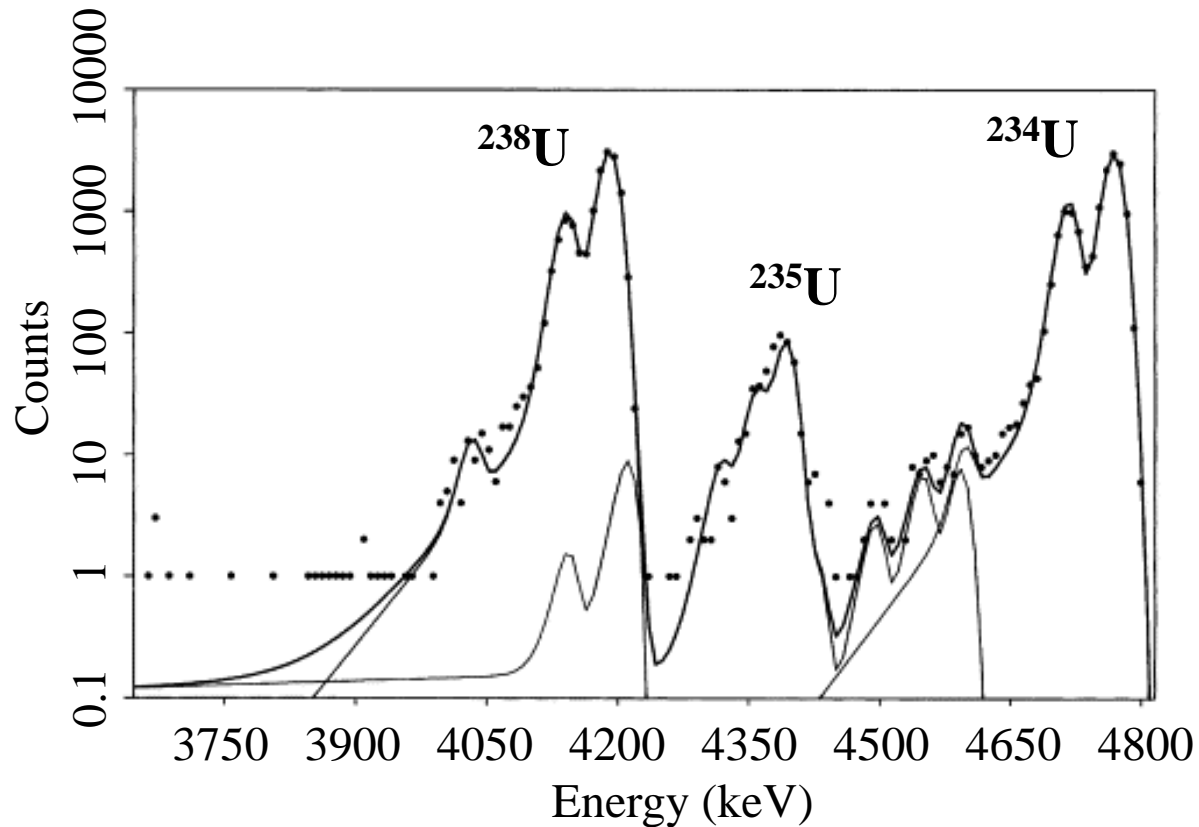


35.9% of ^{241}Am decays result in a 59.5 keV gamma, which has about a 90% chance to escape the absorber.



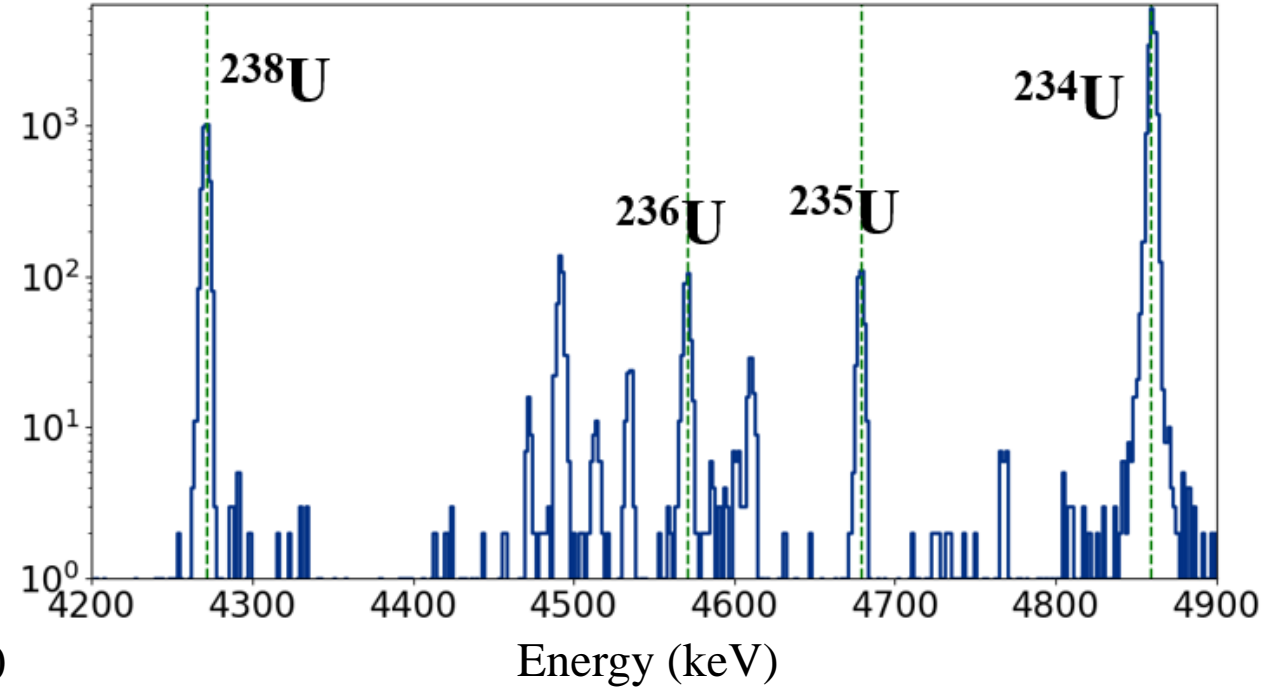
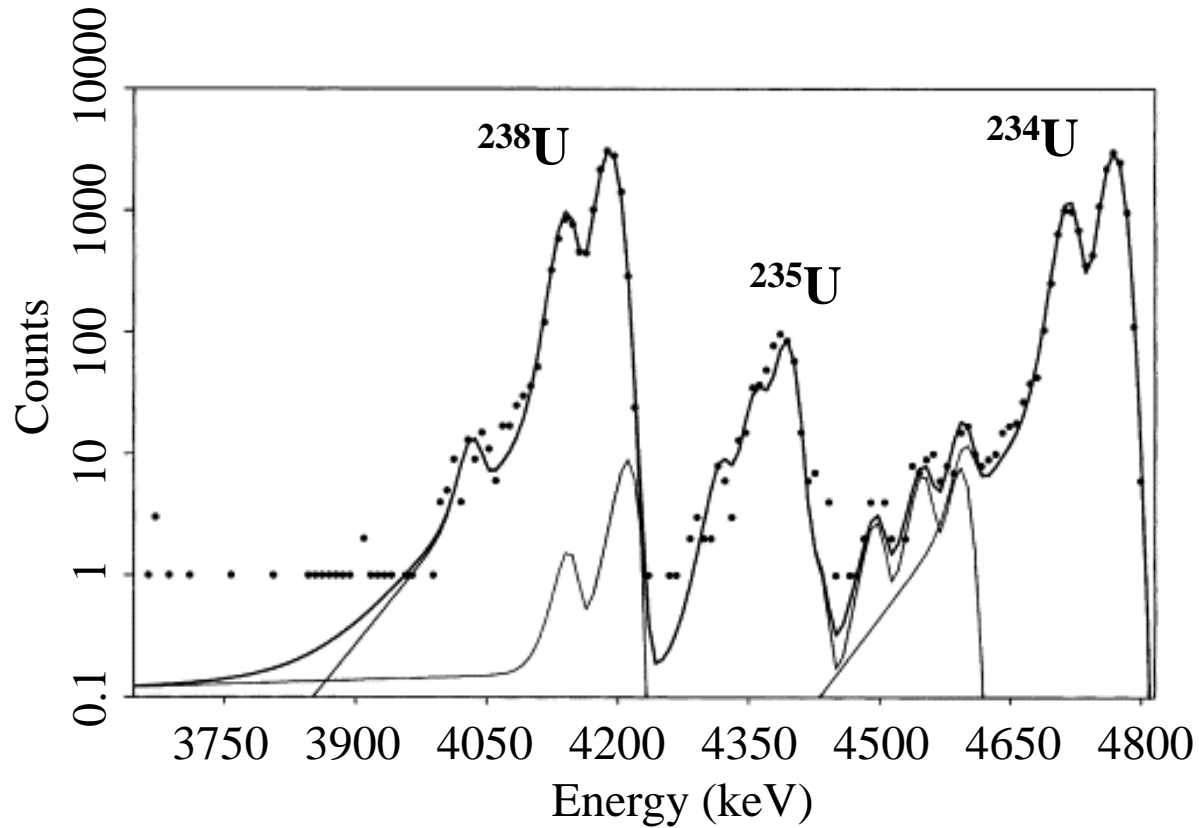
Radiation from source embedded in absorber has different probabilities of escape

Uranium Analysis Challenges



M.P. Montero, et al. "Isotopic uranium and plutonium analysis by alpha-particle spectrometry", NIM-B (2004).

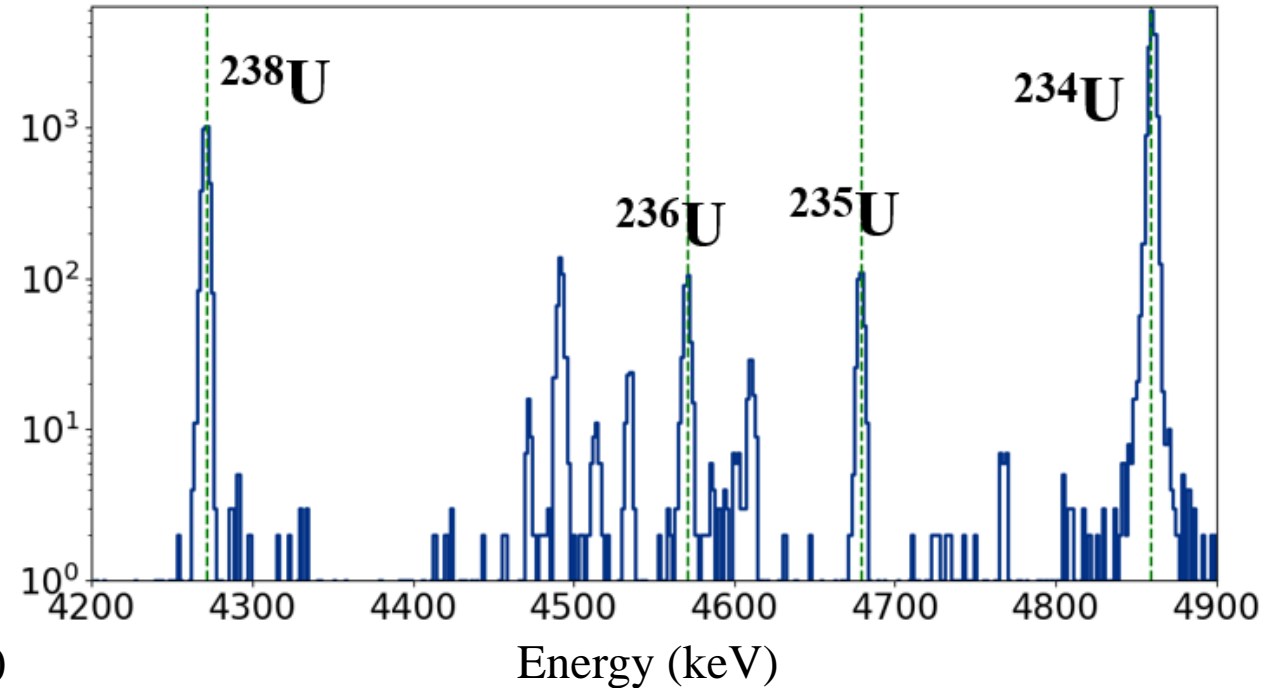
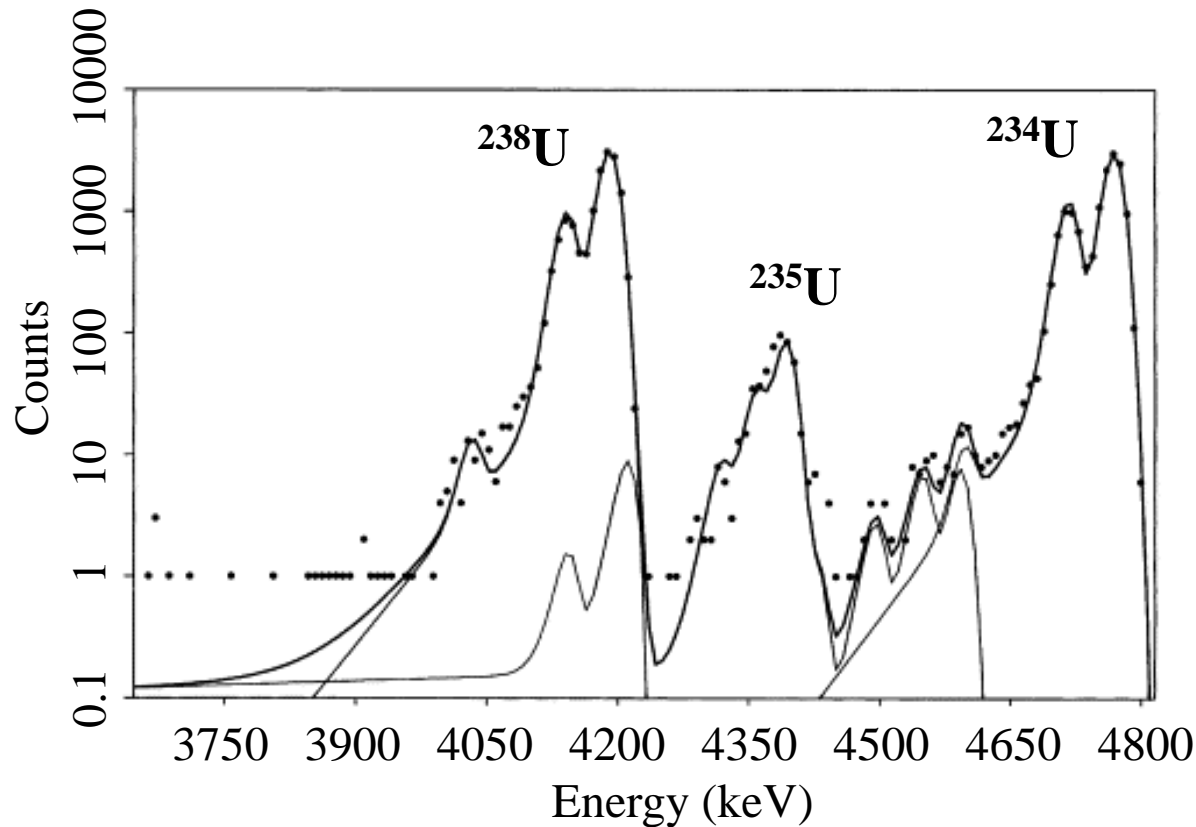
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- Alpha Spectrometry: Many overlapping peaks that require complex multi-peak fitting

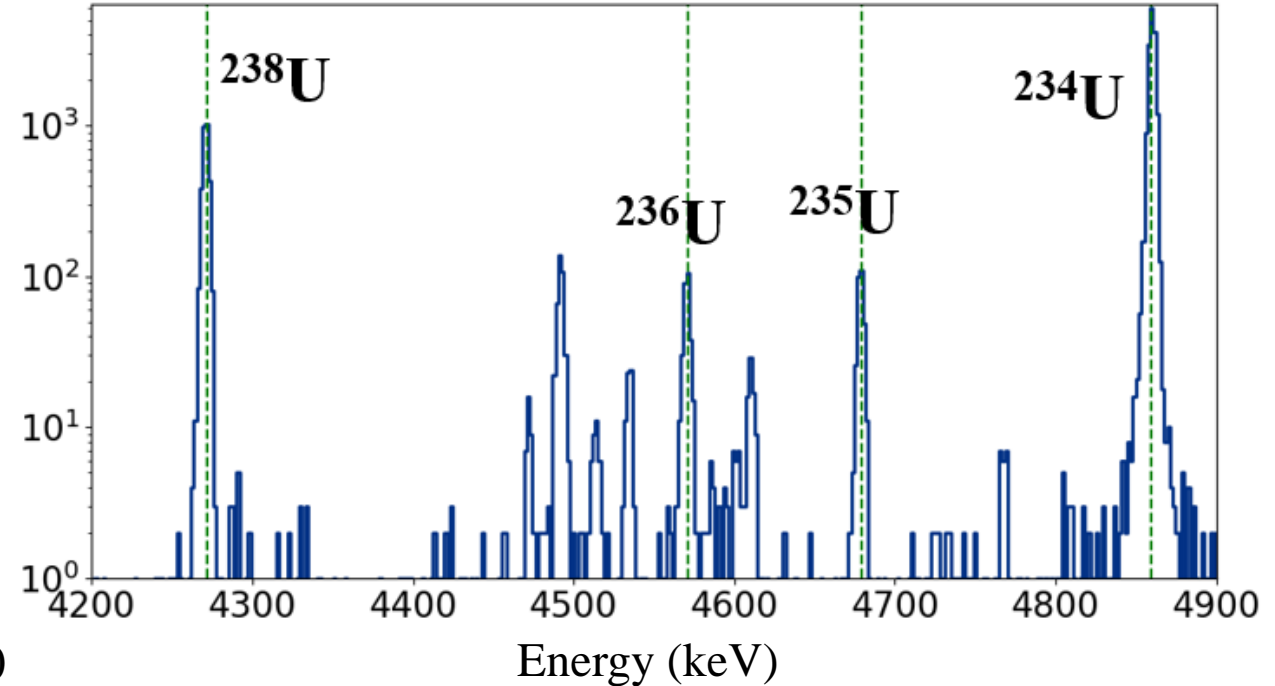
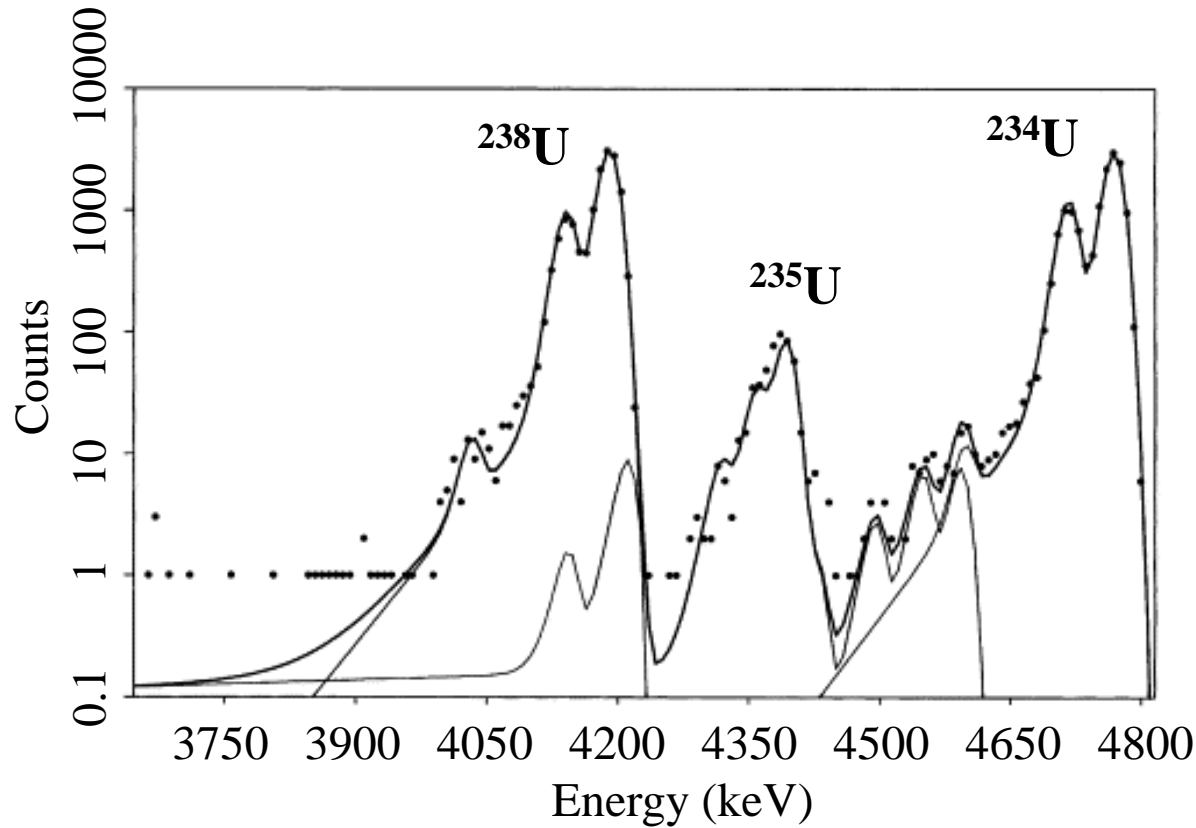
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- Decay Energy Spectroscopy: One peak* at the Q-value for each isotope

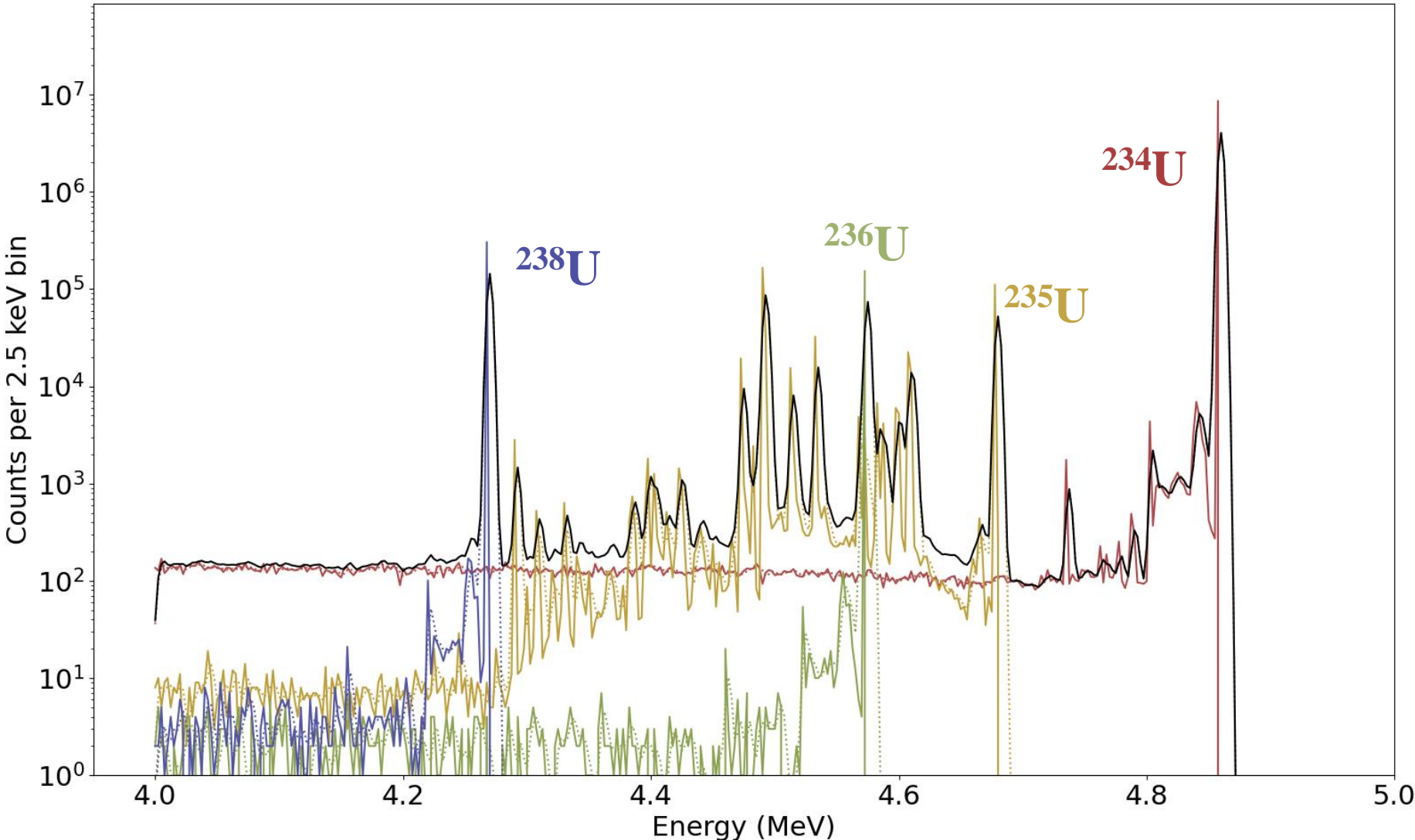
Uranium Analysis Challenges



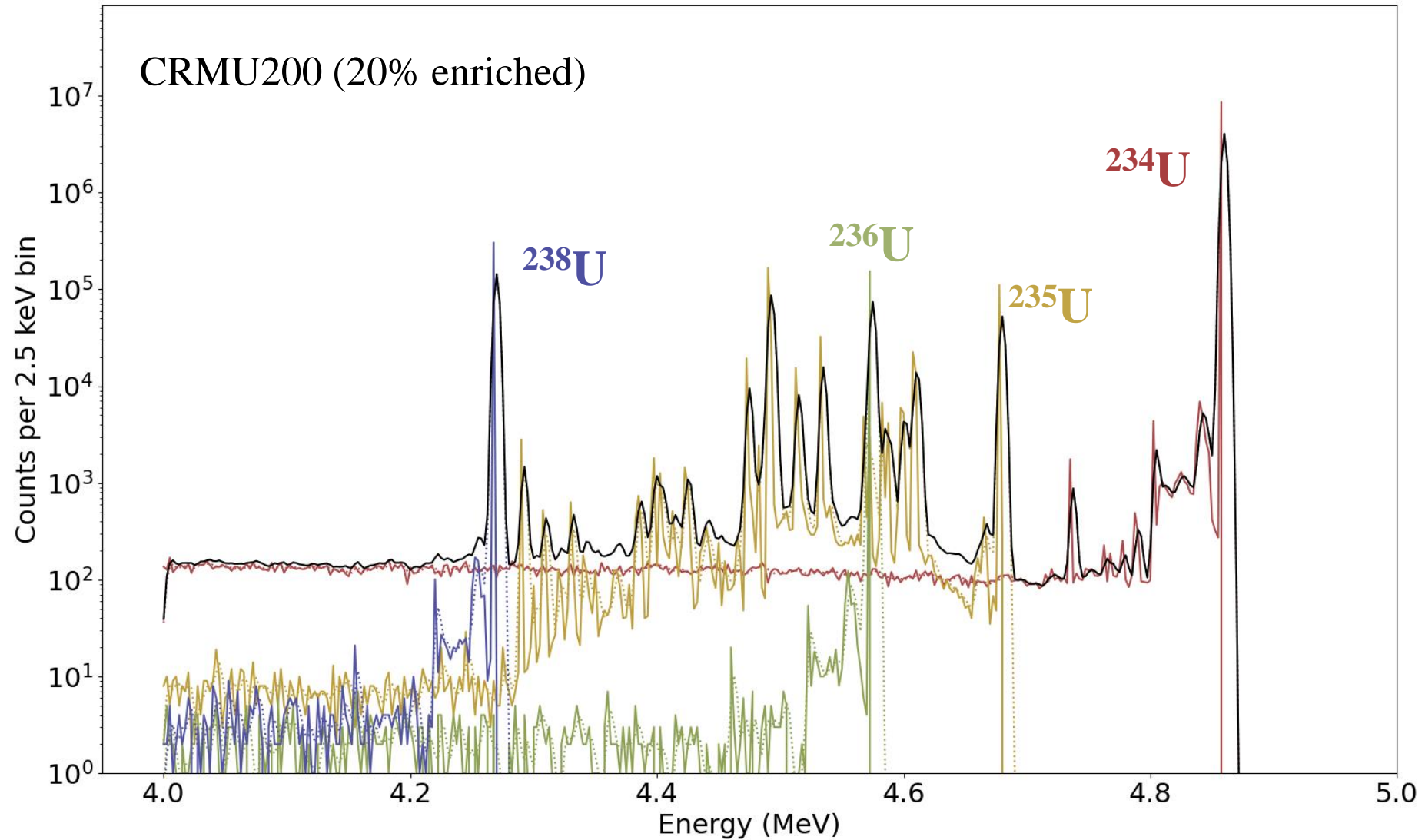
M.P. Montero, et al. "Isotopic uranium and plutonium analysis by alpha-particle spectrometry", NIM-B (2004).

- Alpha Spectrometry: Many overlapping peaks that require complex multi-peak fitting
- Decay Energy Spectroscopy: One peak* at the Q-value for each isotope
- Minimal sample prep for DES

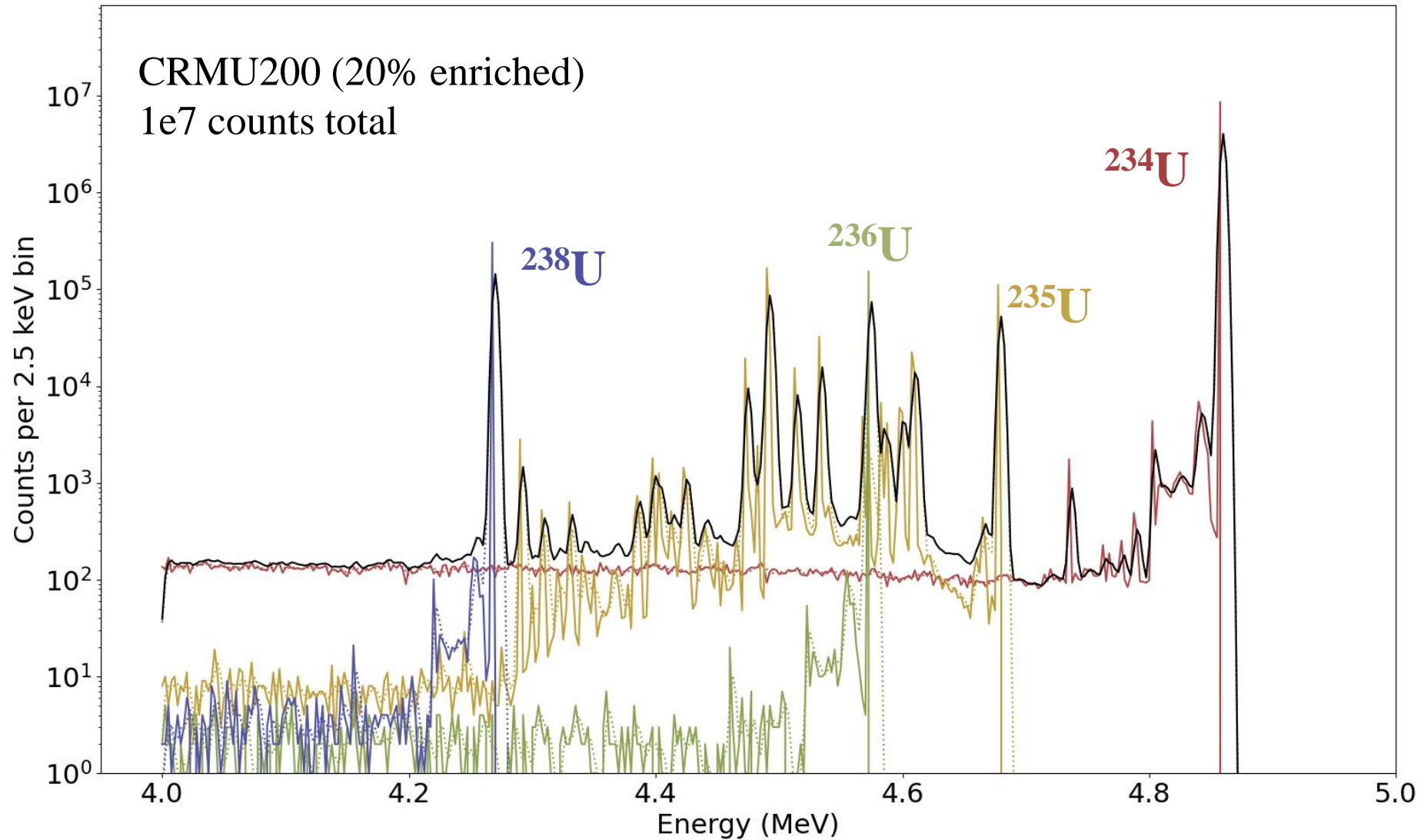
Capping Effects on Spectra



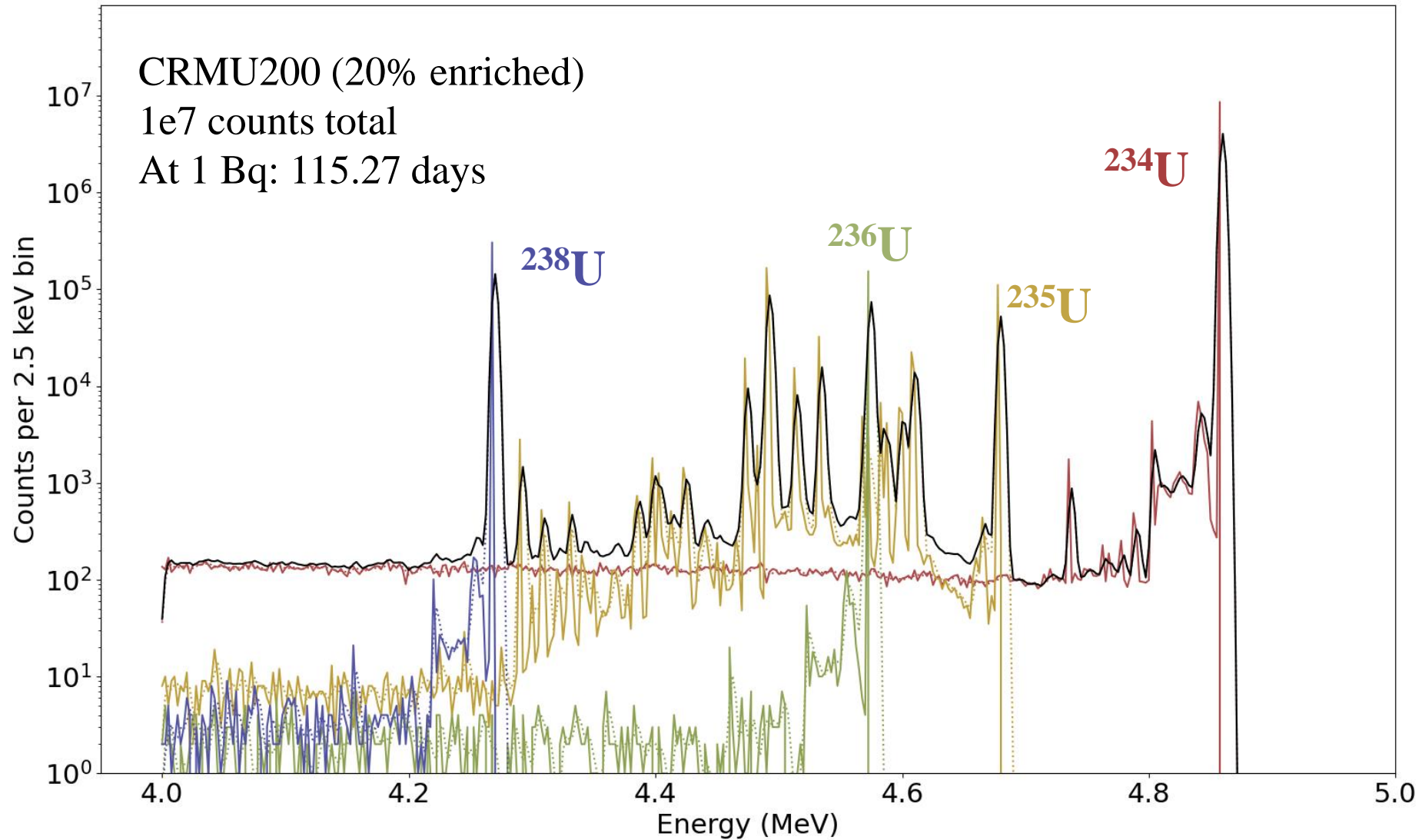
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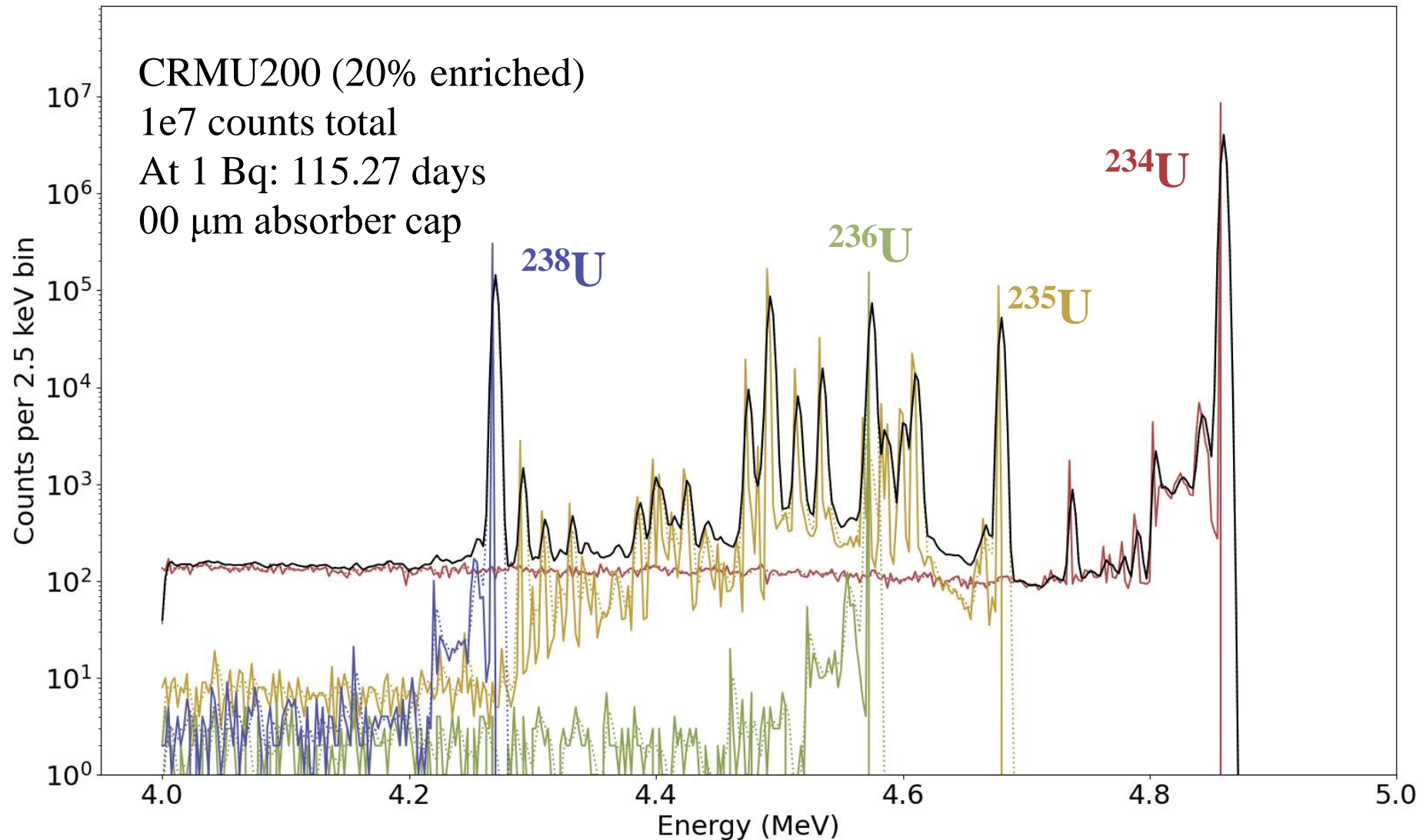
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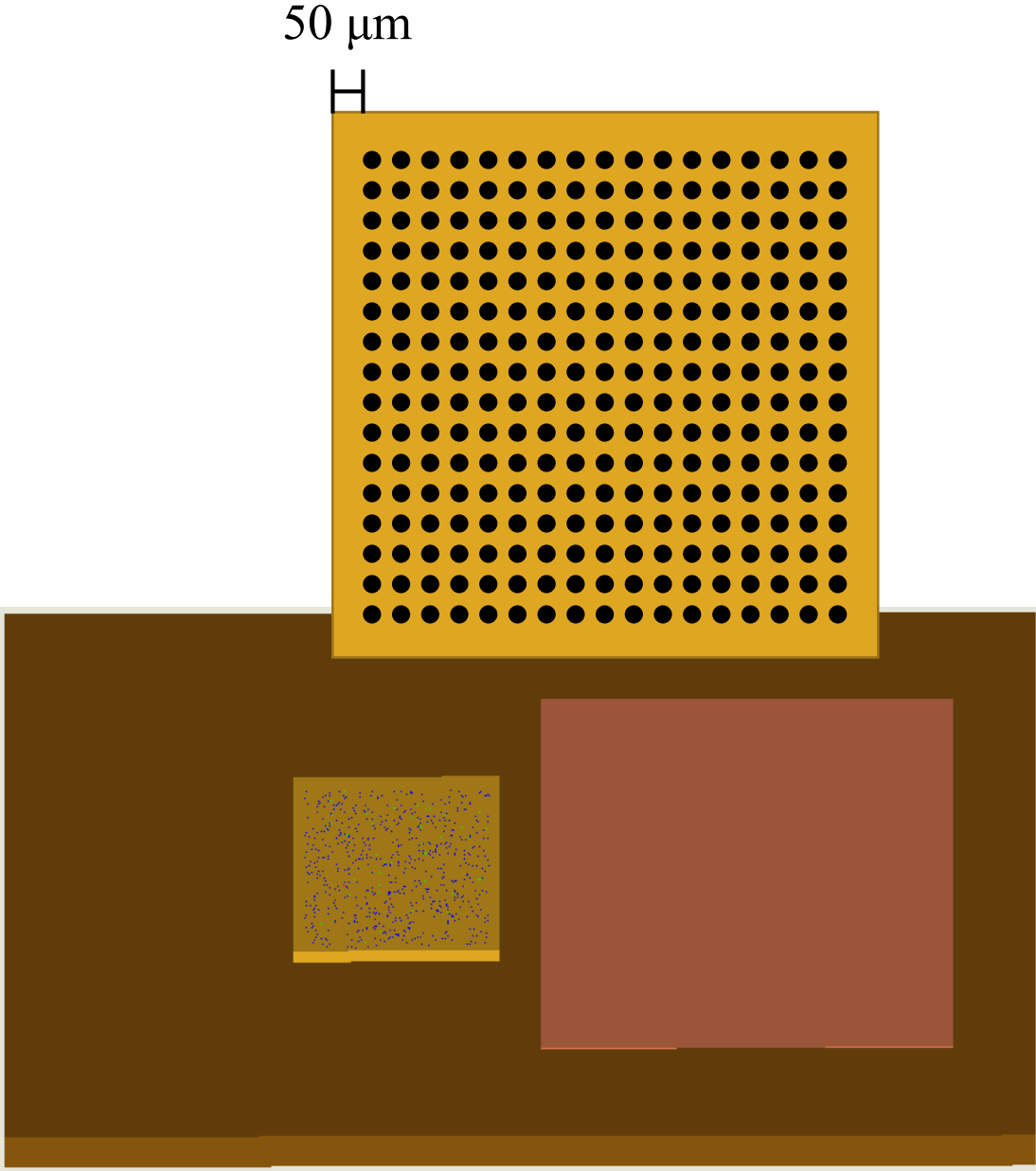
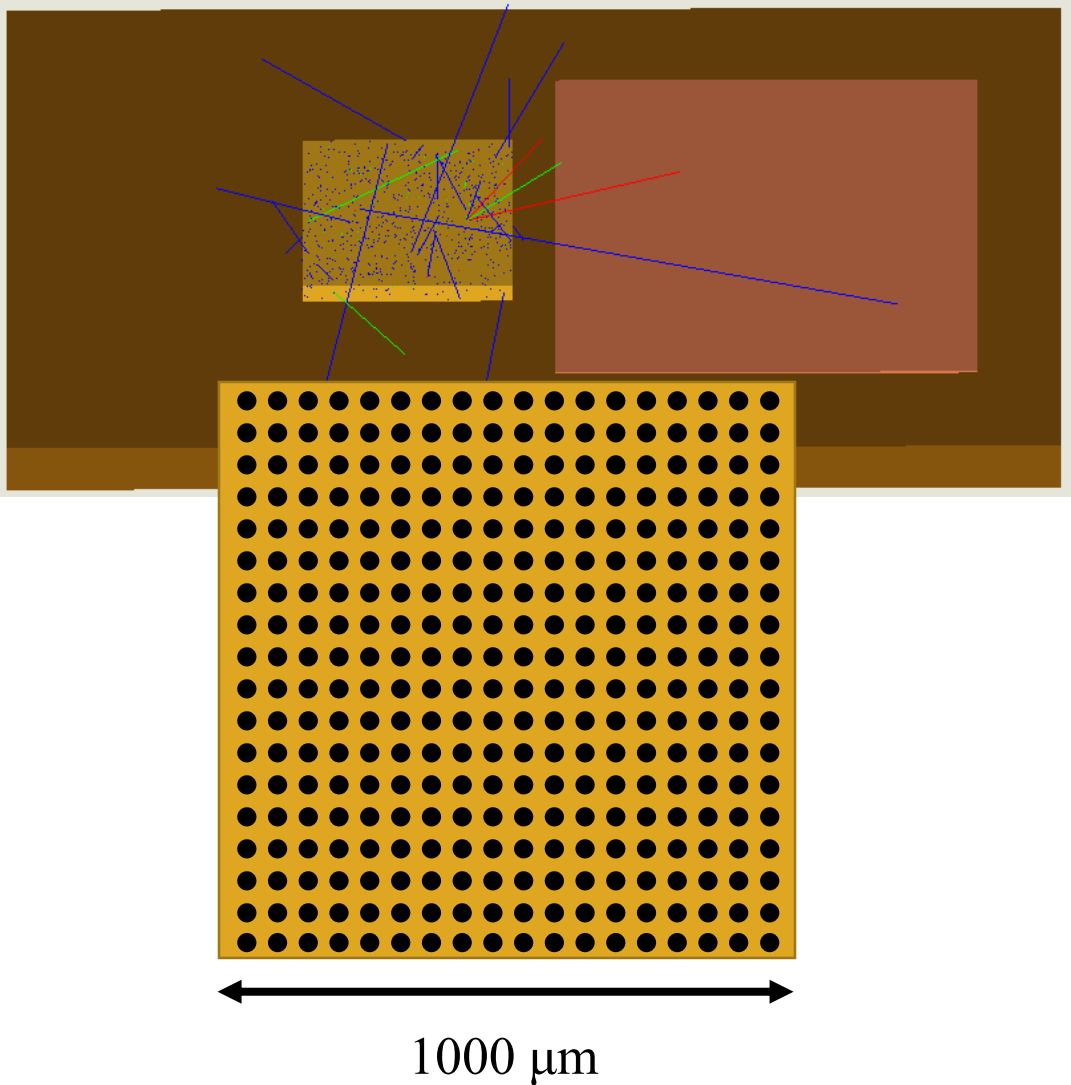
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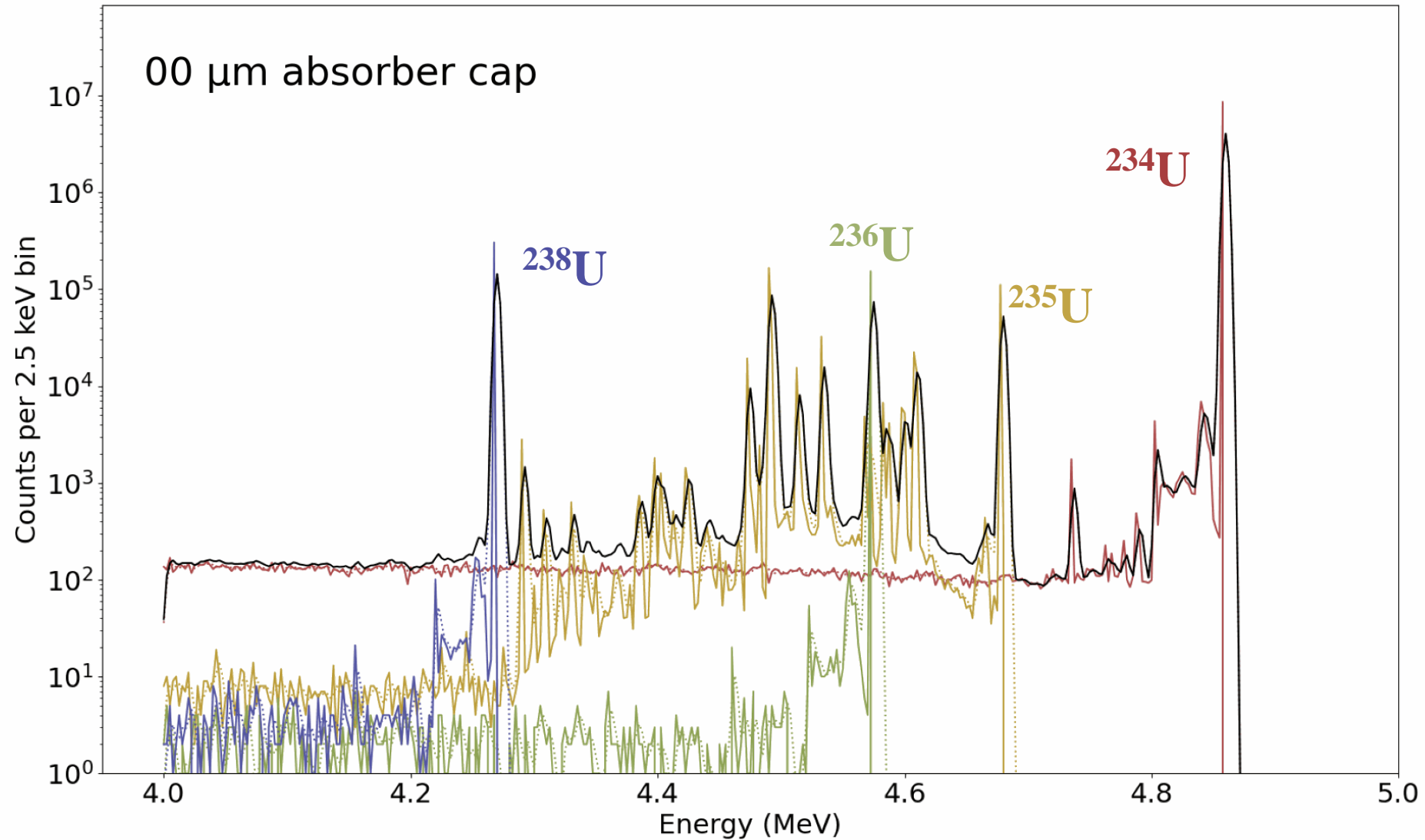
Capping Effects on Spectra



Absorber Capping



Capping Effects on Spectra



Analysis Using Geant4

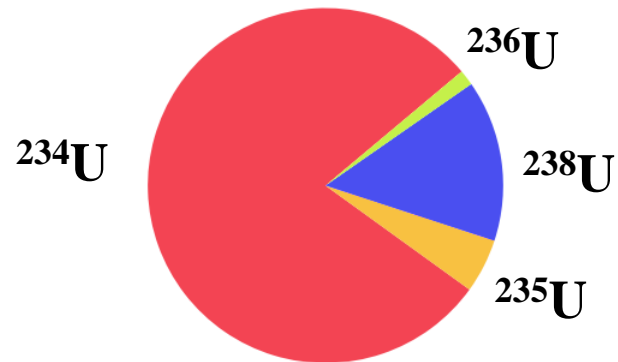
Number of Simulated Counts:

^{234}U : 7892364

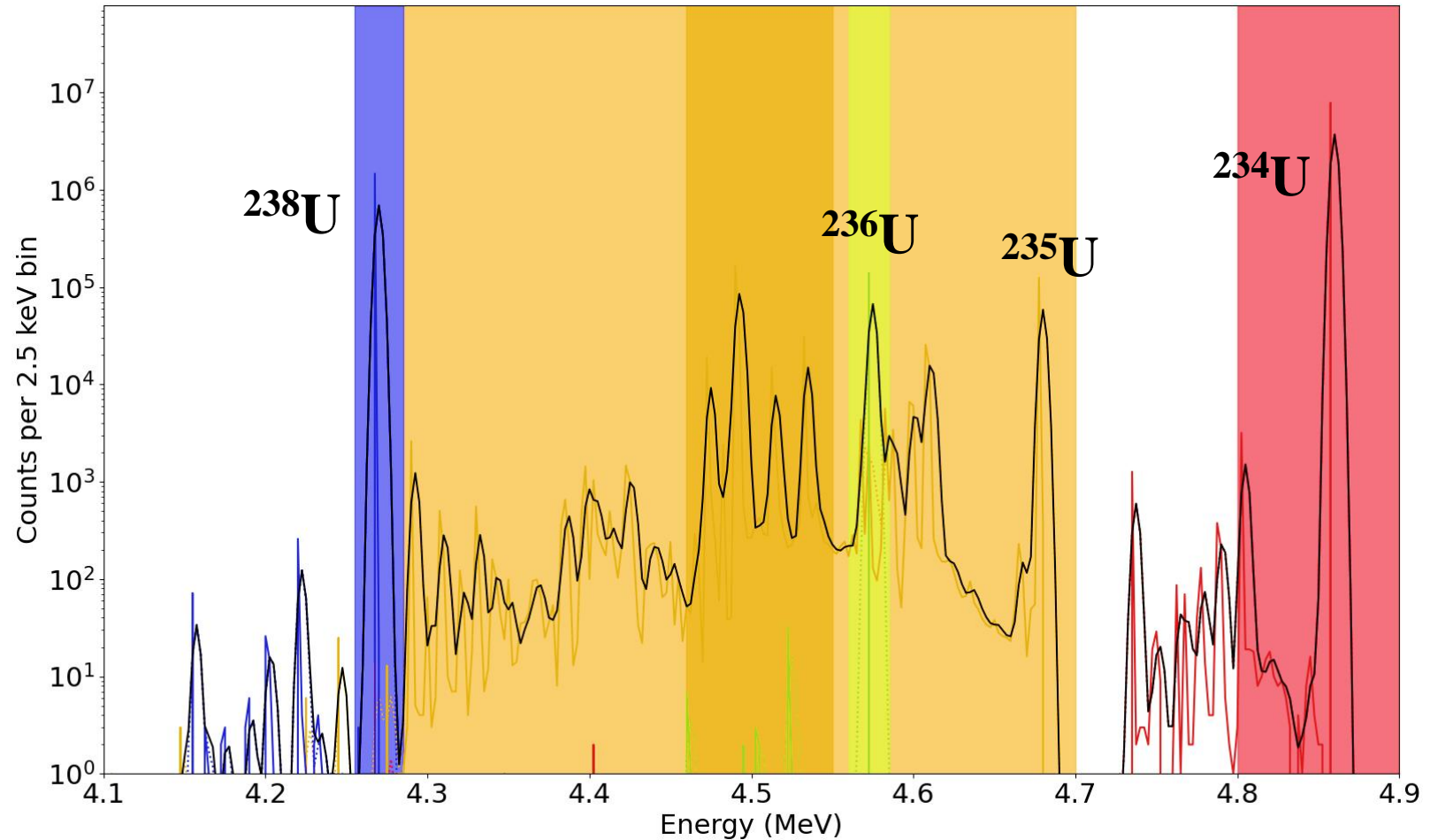
^{235}U : 493189

^{236}U : 141985

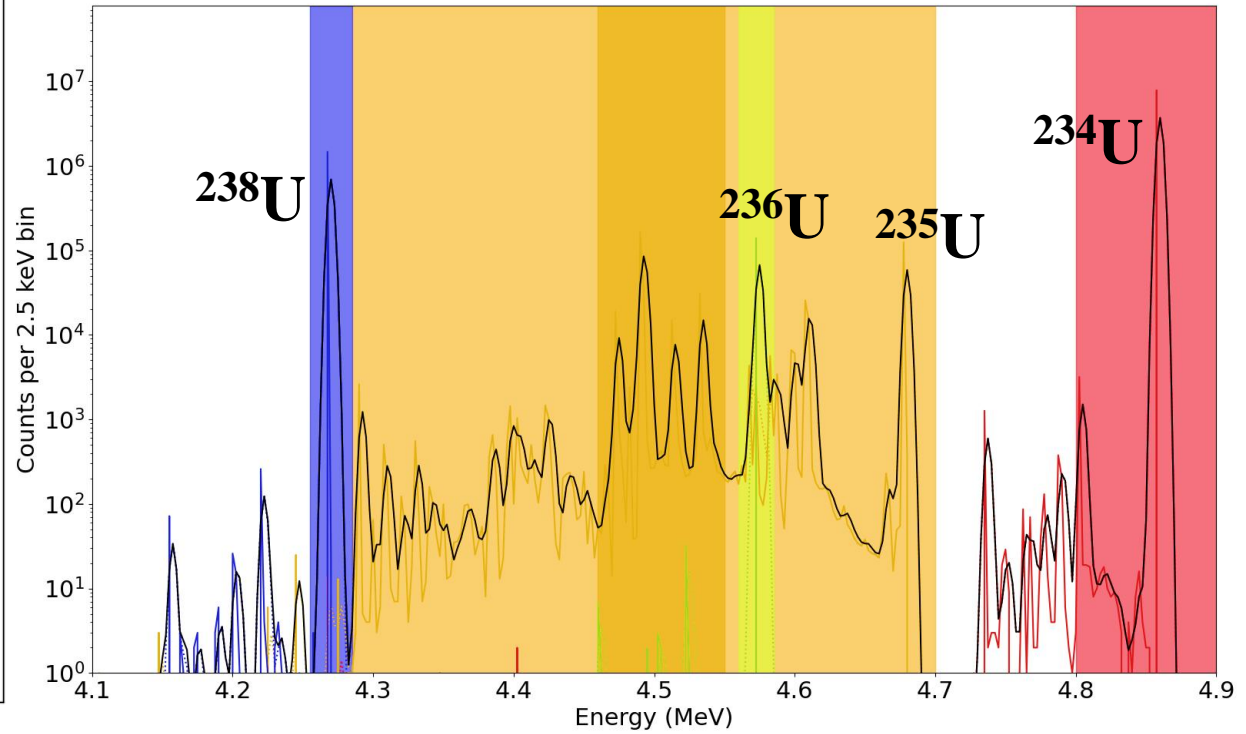
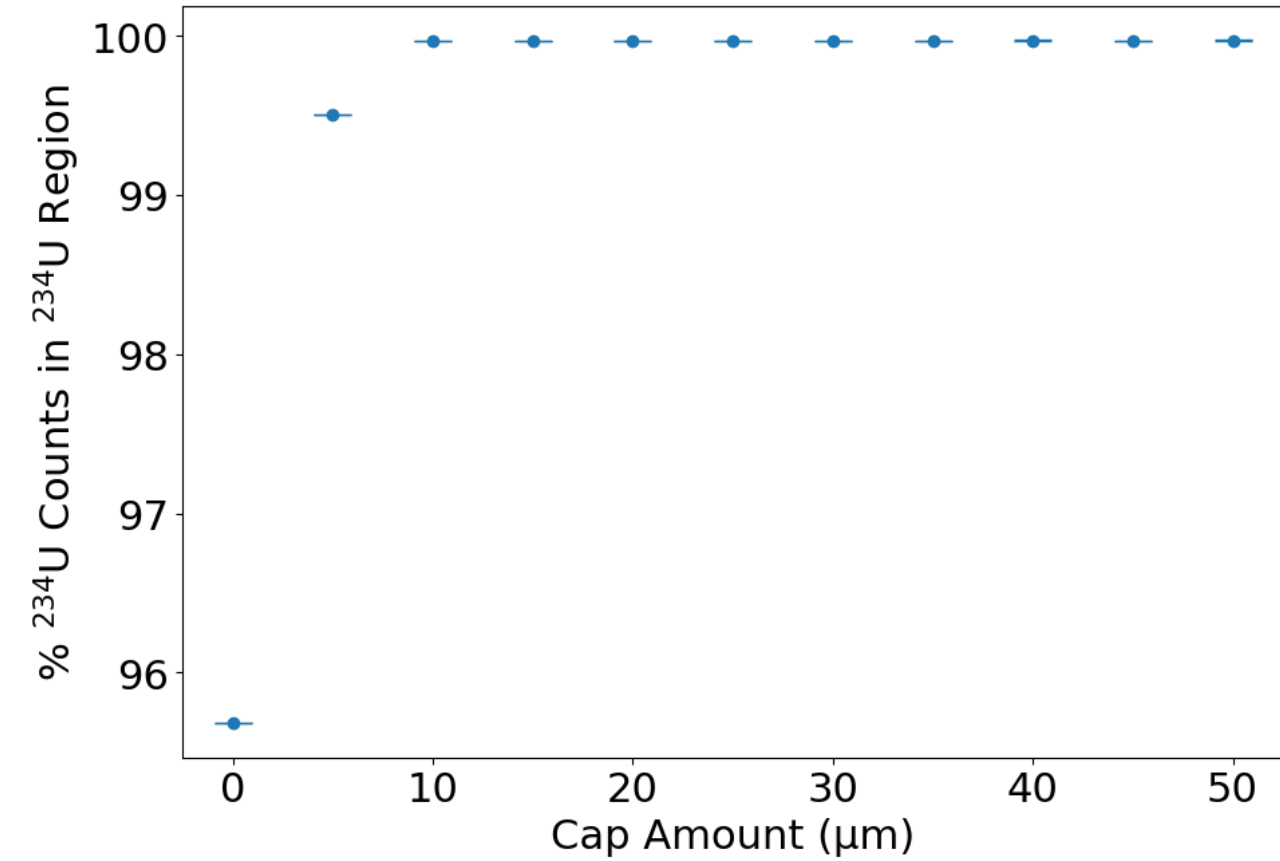
^{238}U : 1472463



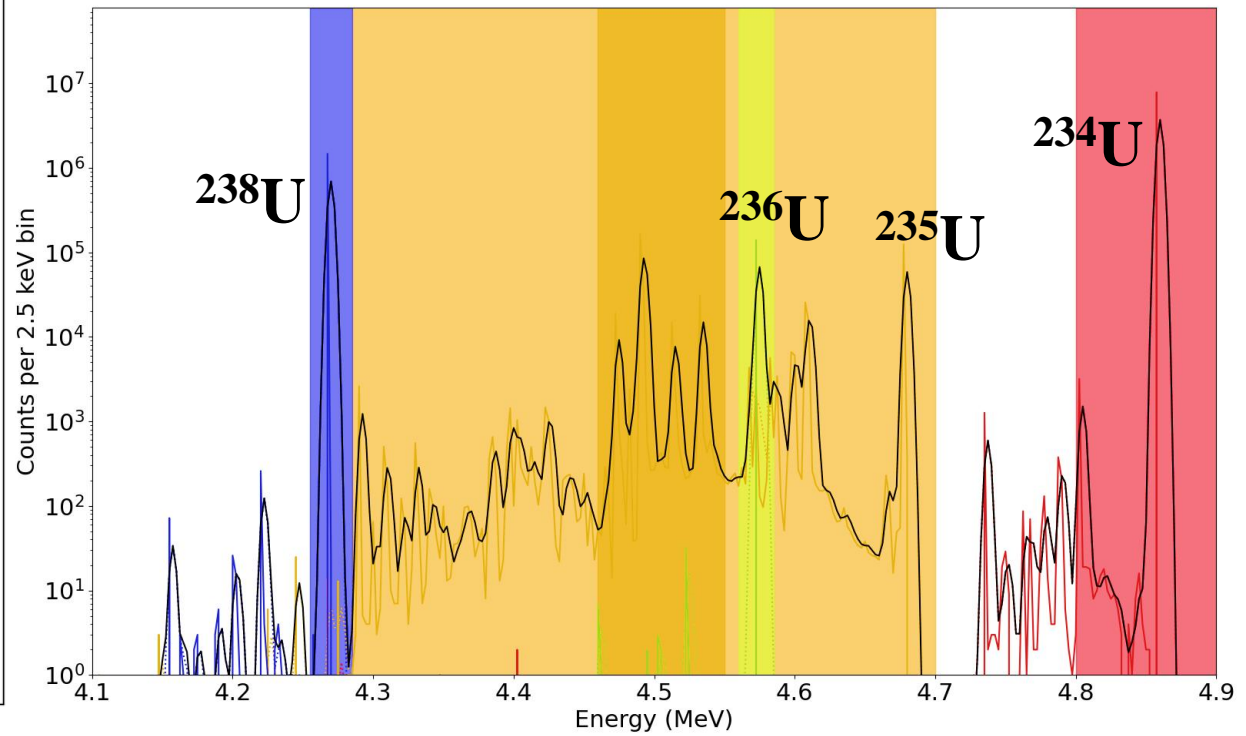
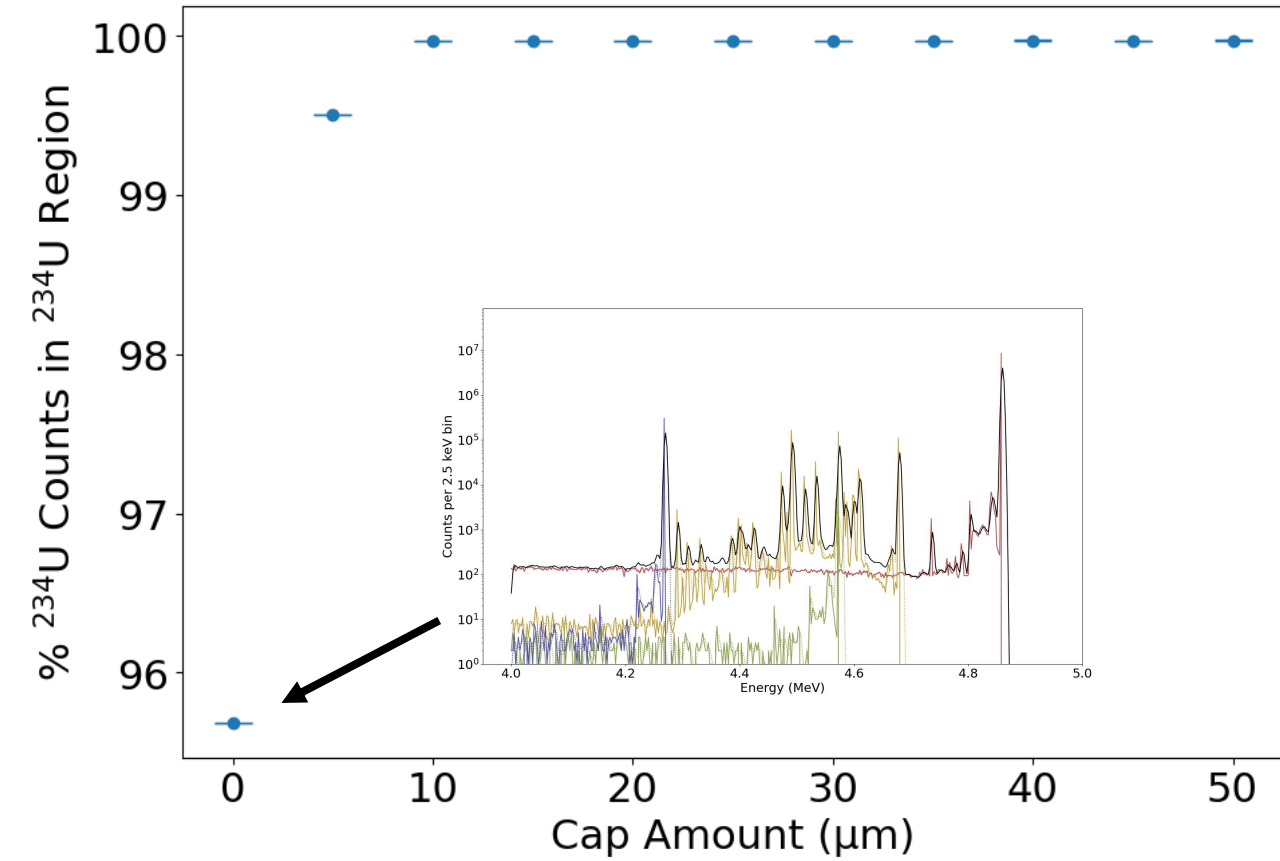
Total: $1\text{e}7$ counts (5% enrichment)



^{234}U Capping Effects



^{234}U Capping Effects



Analysis Using Geant4

How good are these “regions”?

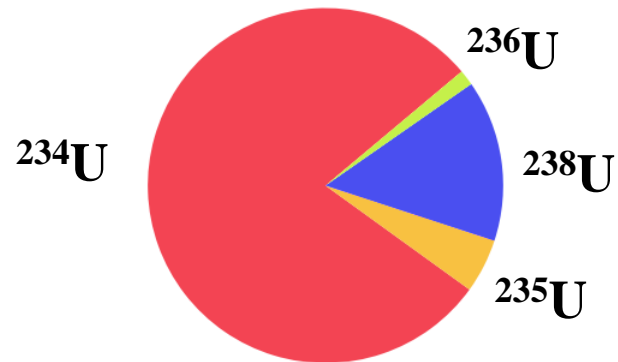
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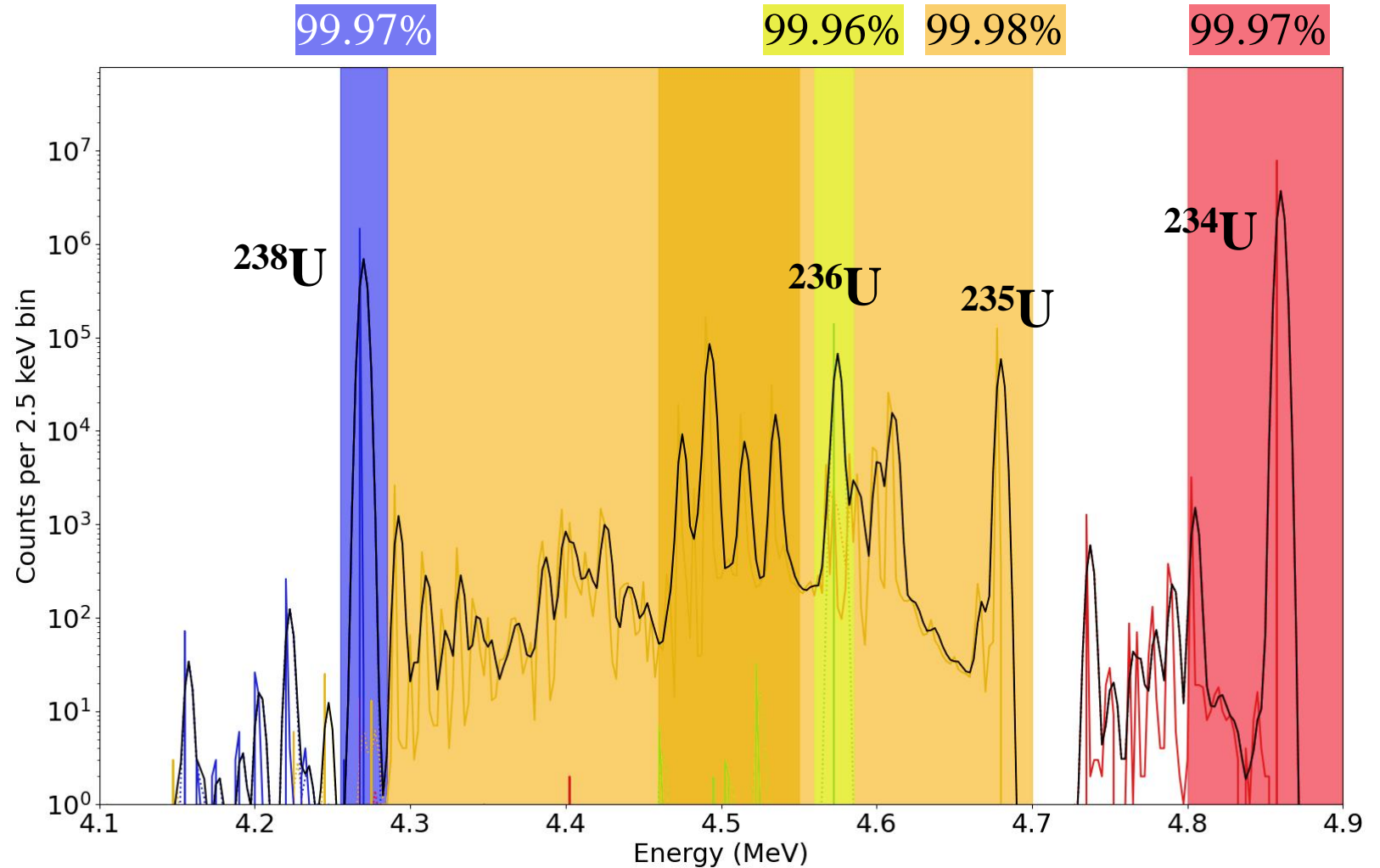
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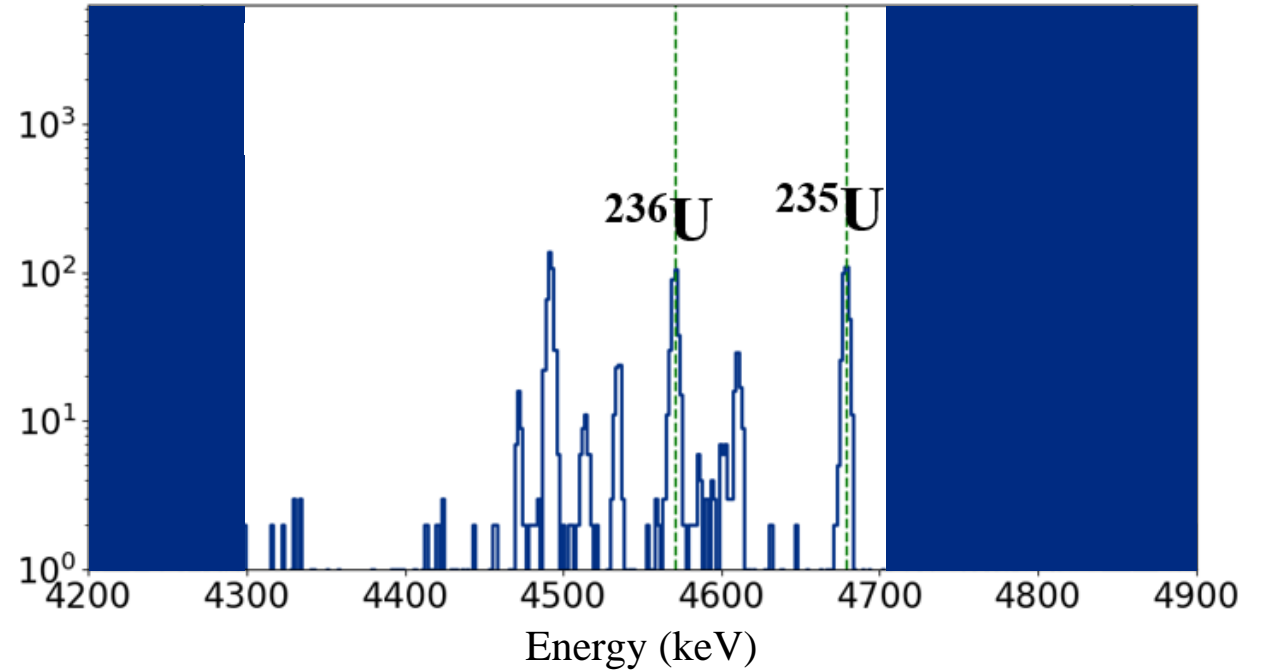
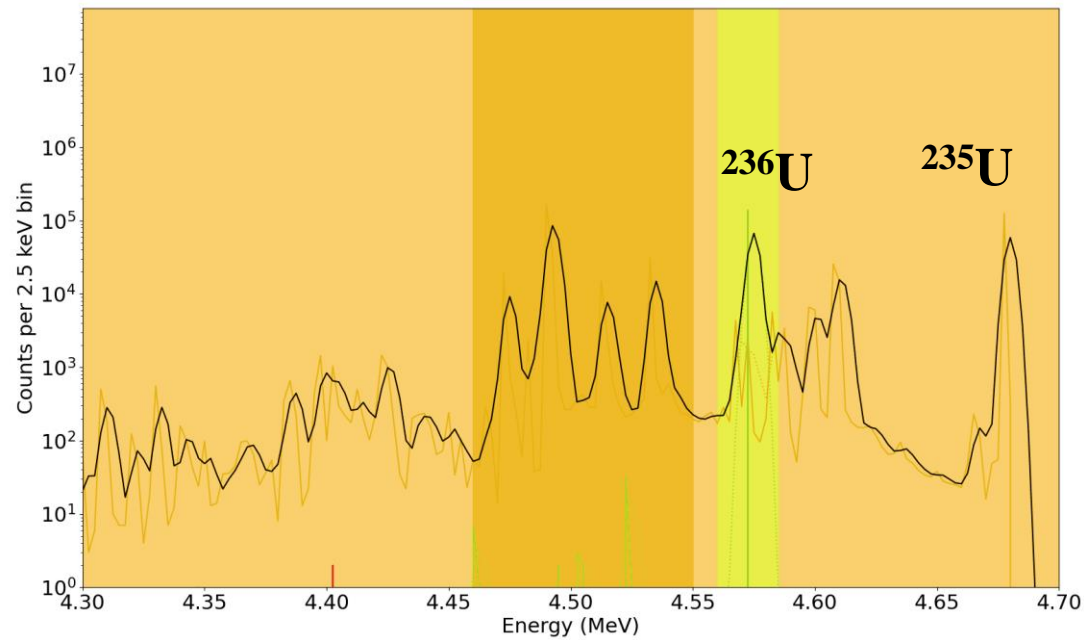
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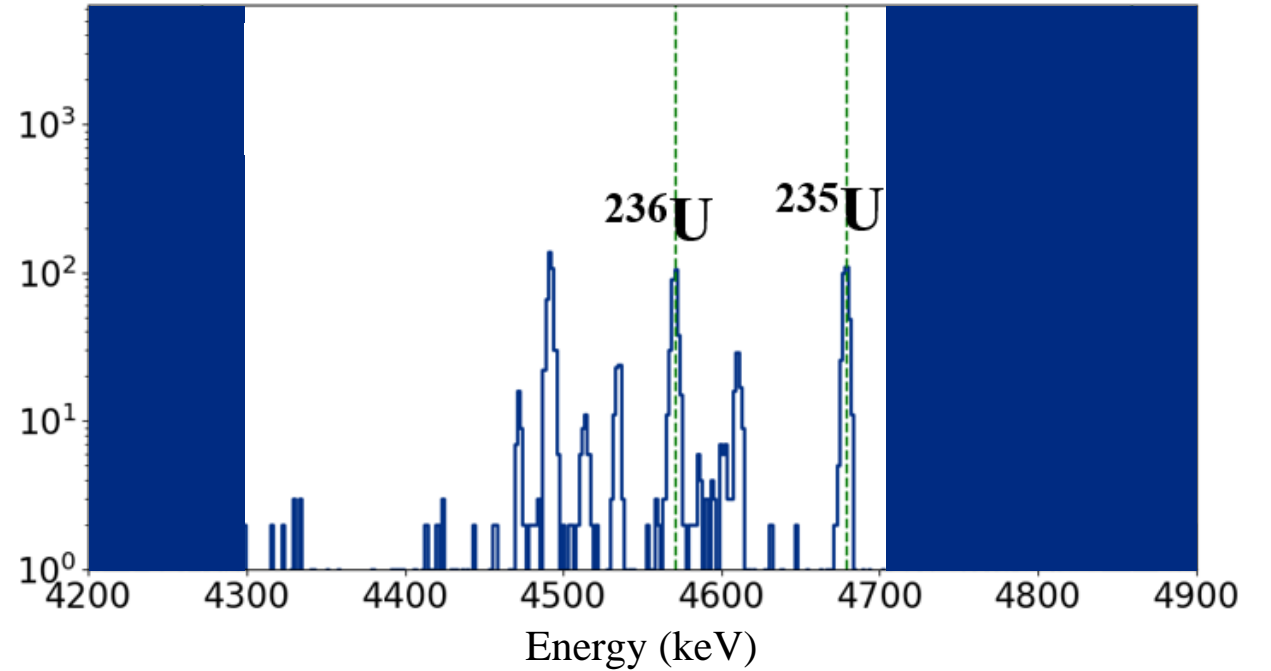
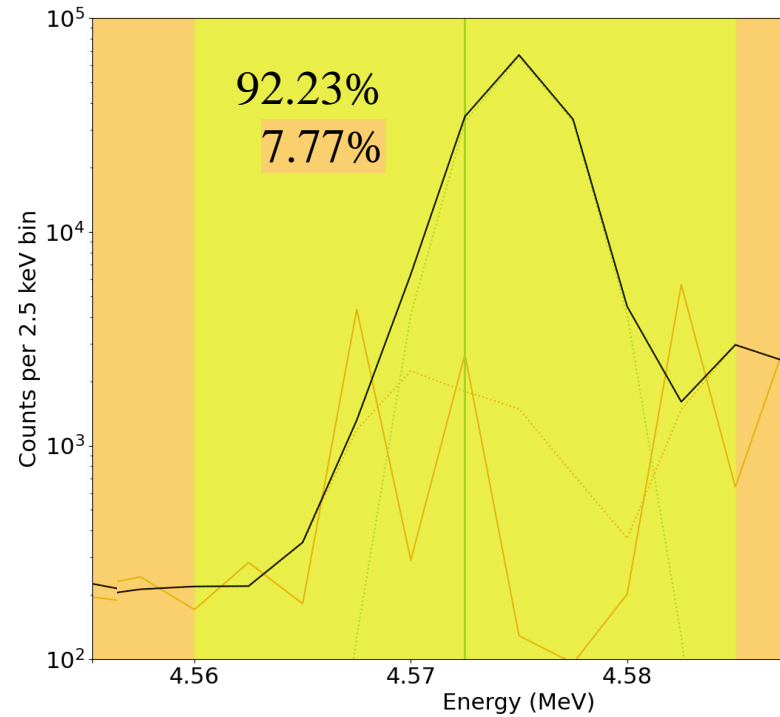


Uranium Analysis Challenges



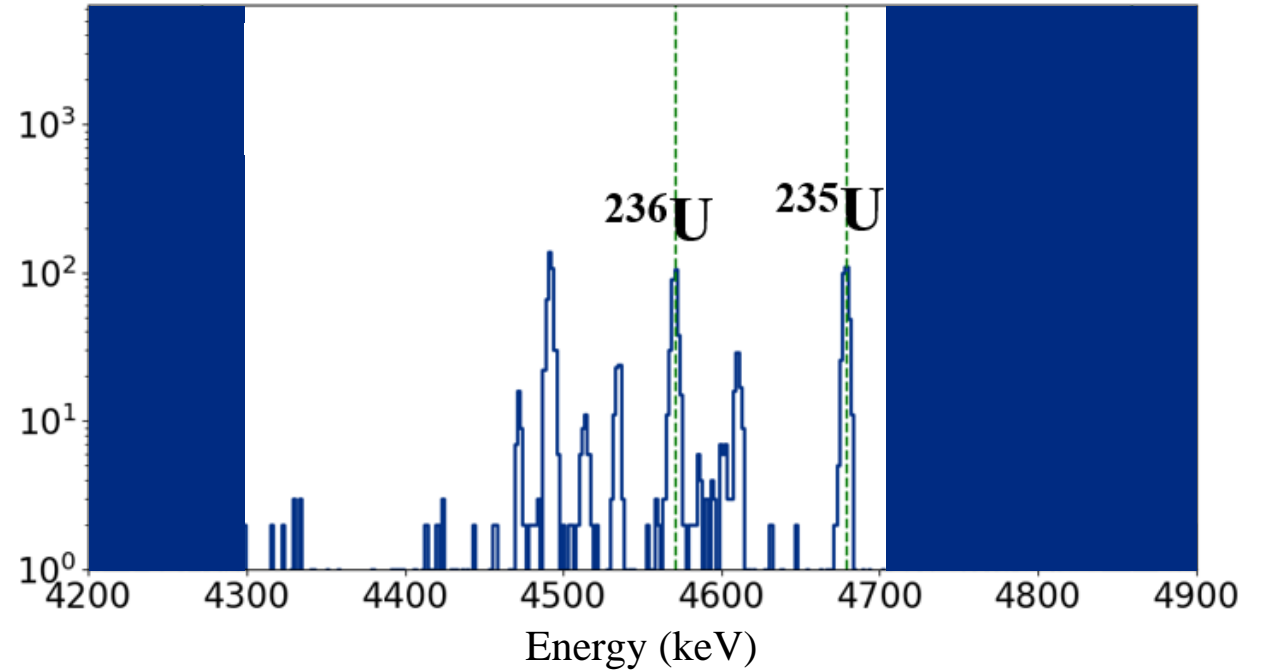
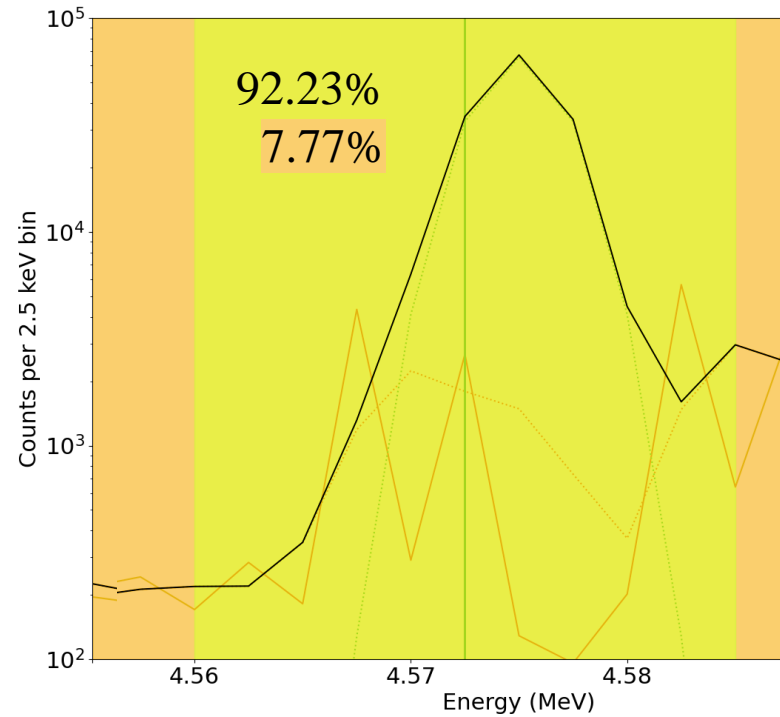
Uranium Analysis Challenges

Where do the counts come from?



Uranium Analysis Challenges

Where do the counts come from?



DES Analysis Challenge: Escape peaks can interfere with other peaks

Analysis Using Geant4

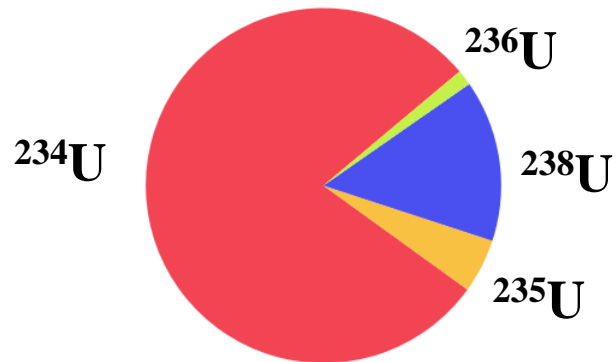
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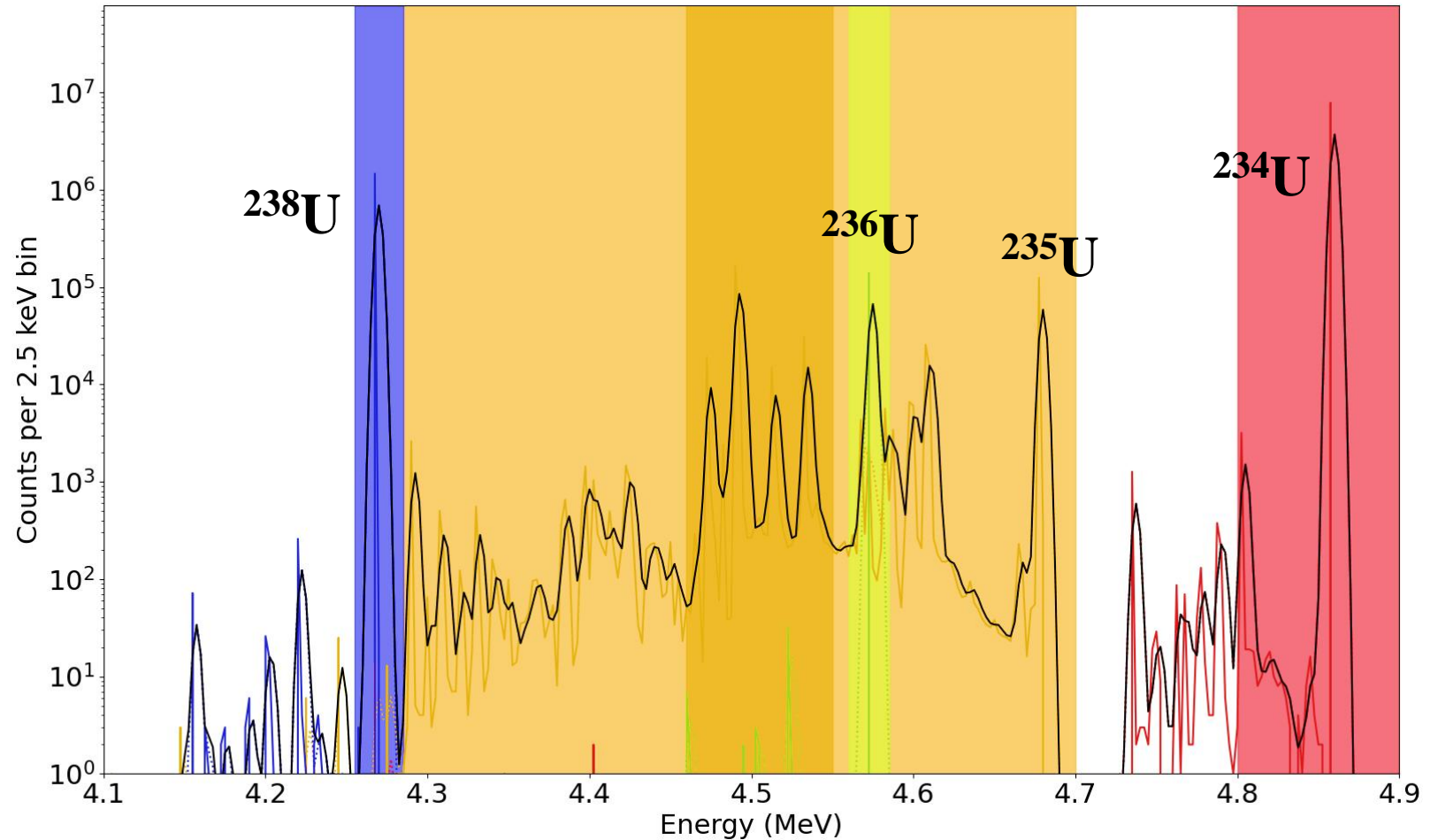
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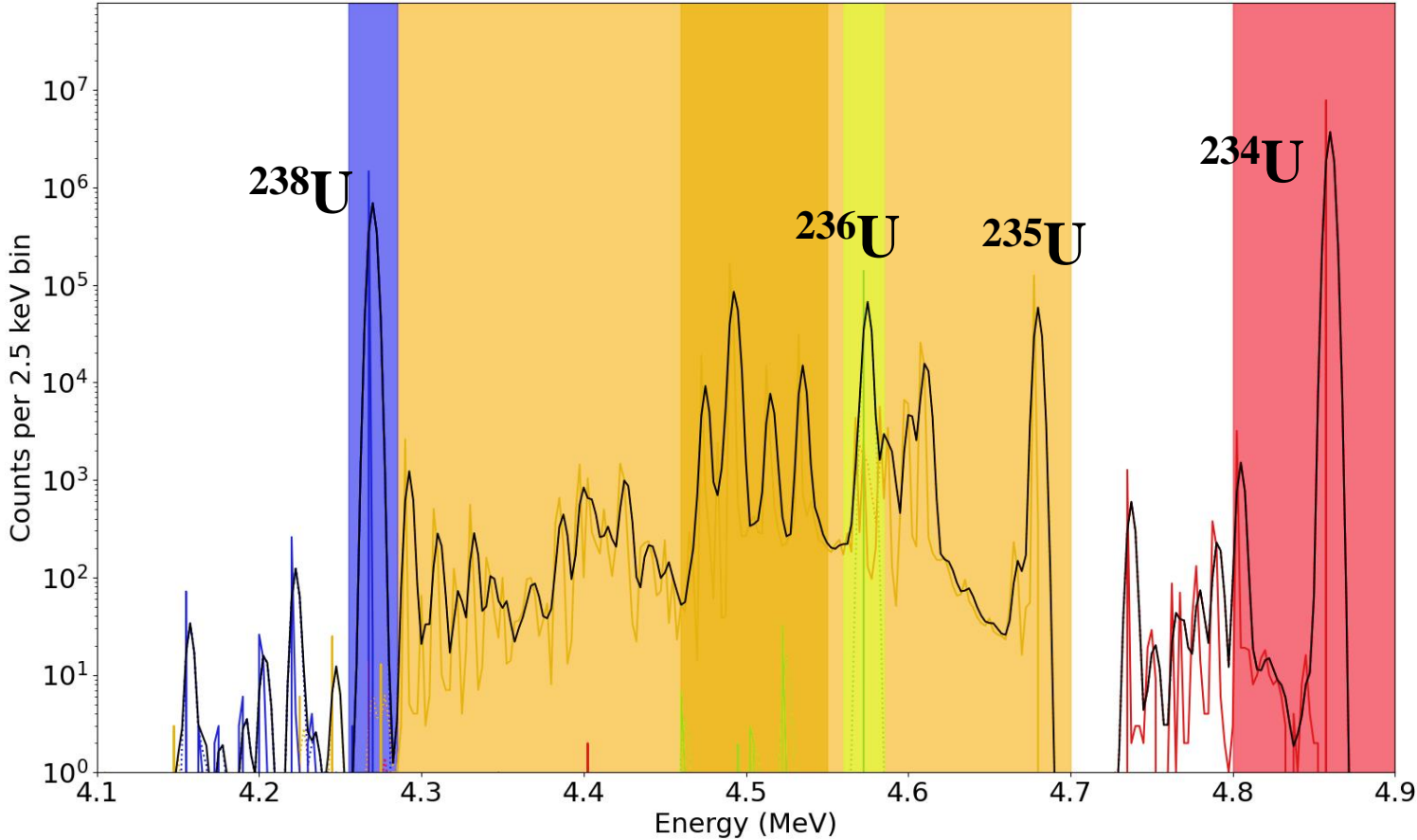
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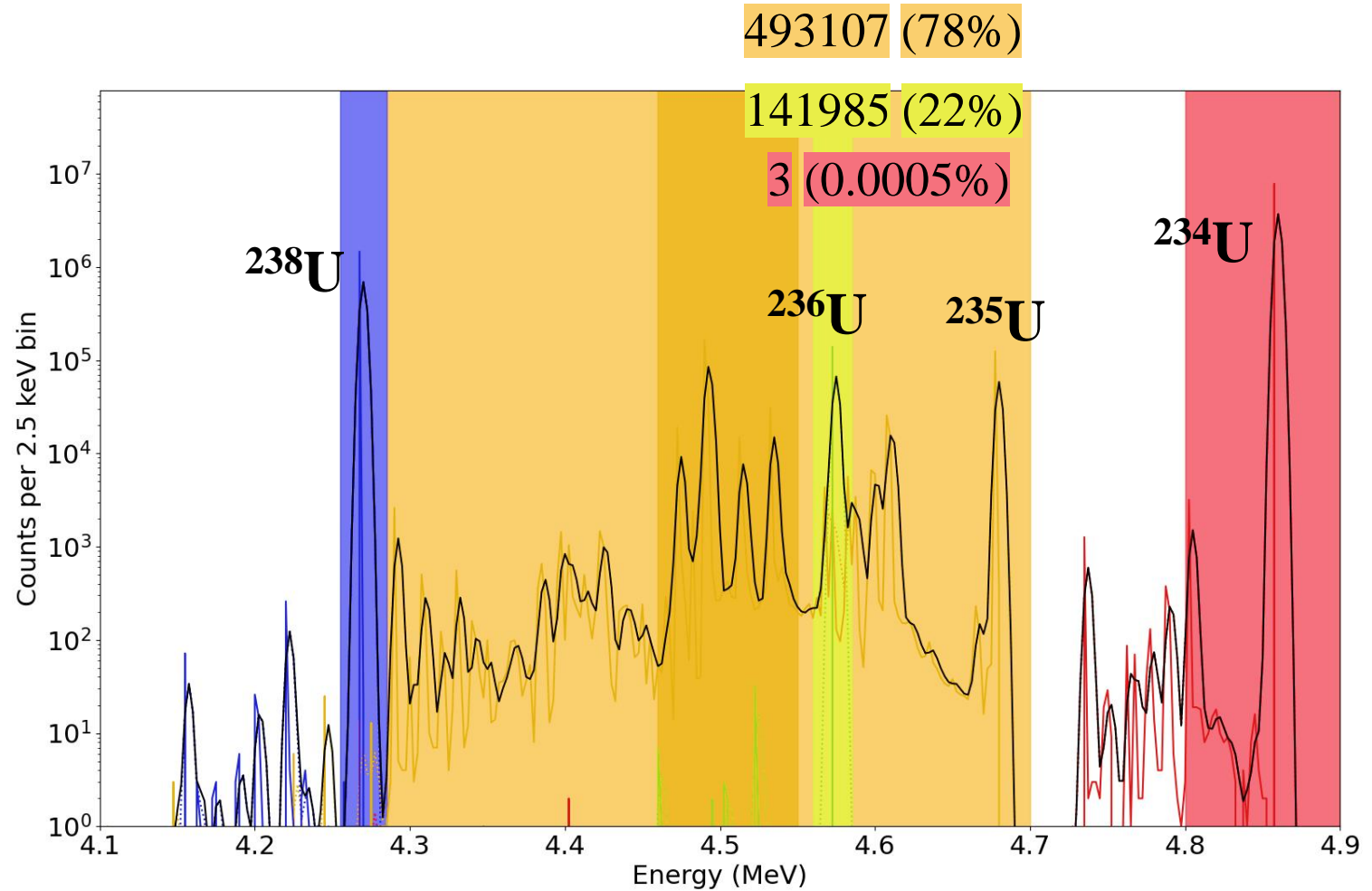
Region	Isotope	# Counts	% Counts
^{234}U	^{234}U	7890088	100%

7890088 (100%)



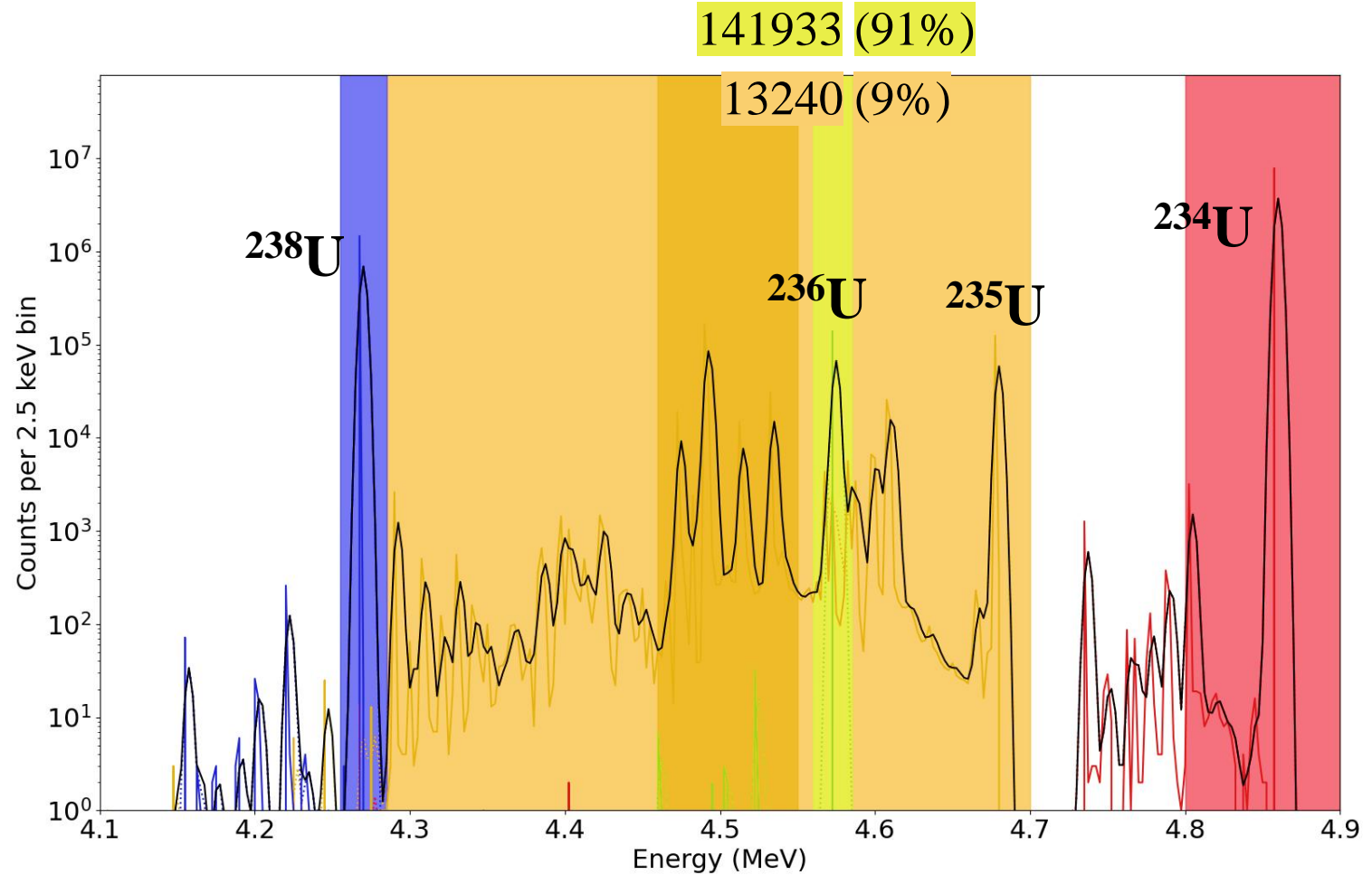
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Region	Isotope	# Counts	% Counts
^{234}U	^{234}U	7890088	100%
^{235}U	^{235}U	493107	78%
^{235}U	^{236}U	141985	22%
^{235}U	^{234}U	3	0.0005%



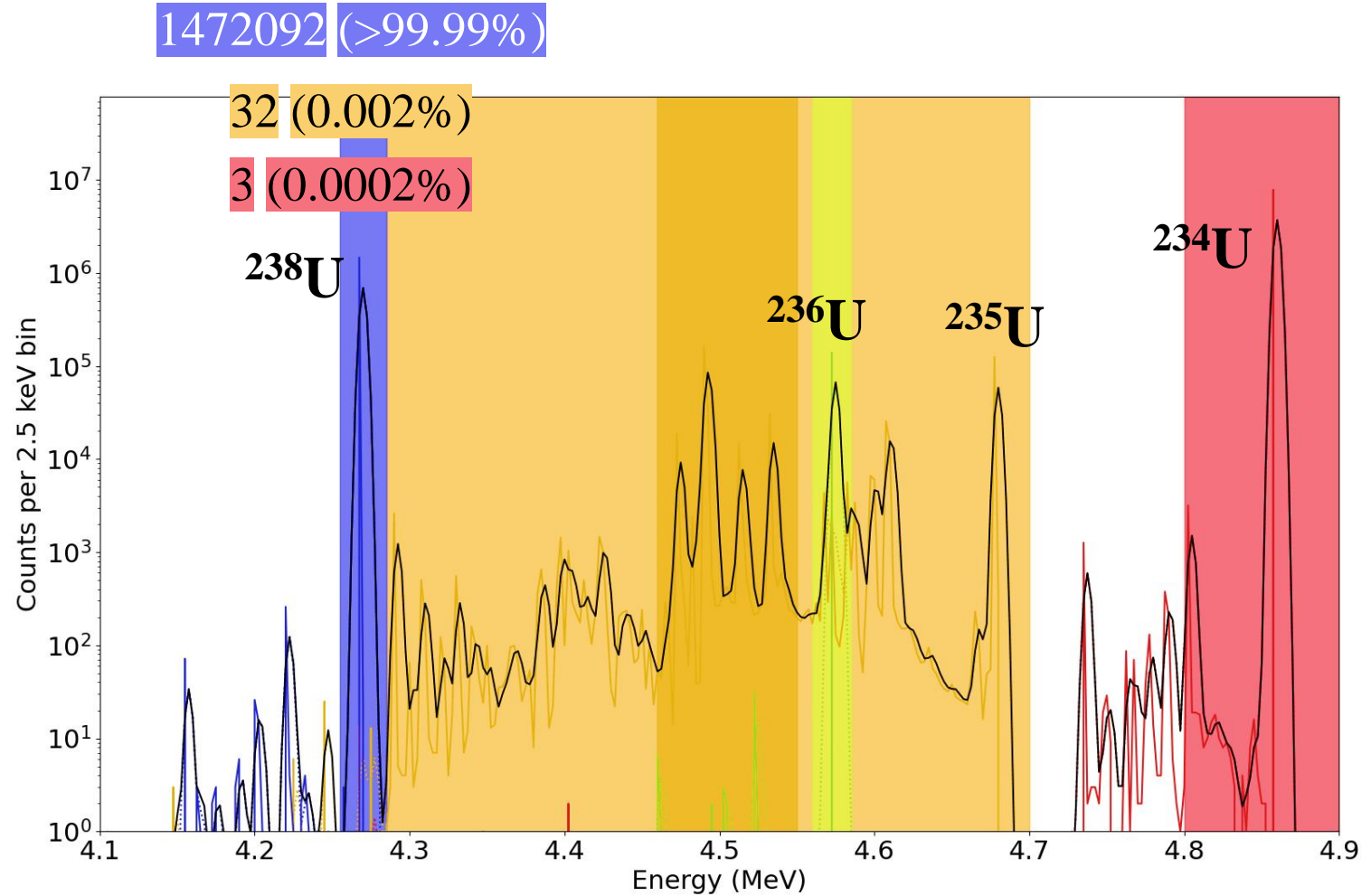
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^{235}U	^{234}U	3	0.0005%
^{236}U	^{236}U	141933	91%
^{236}U	^{235}U	13240	9%



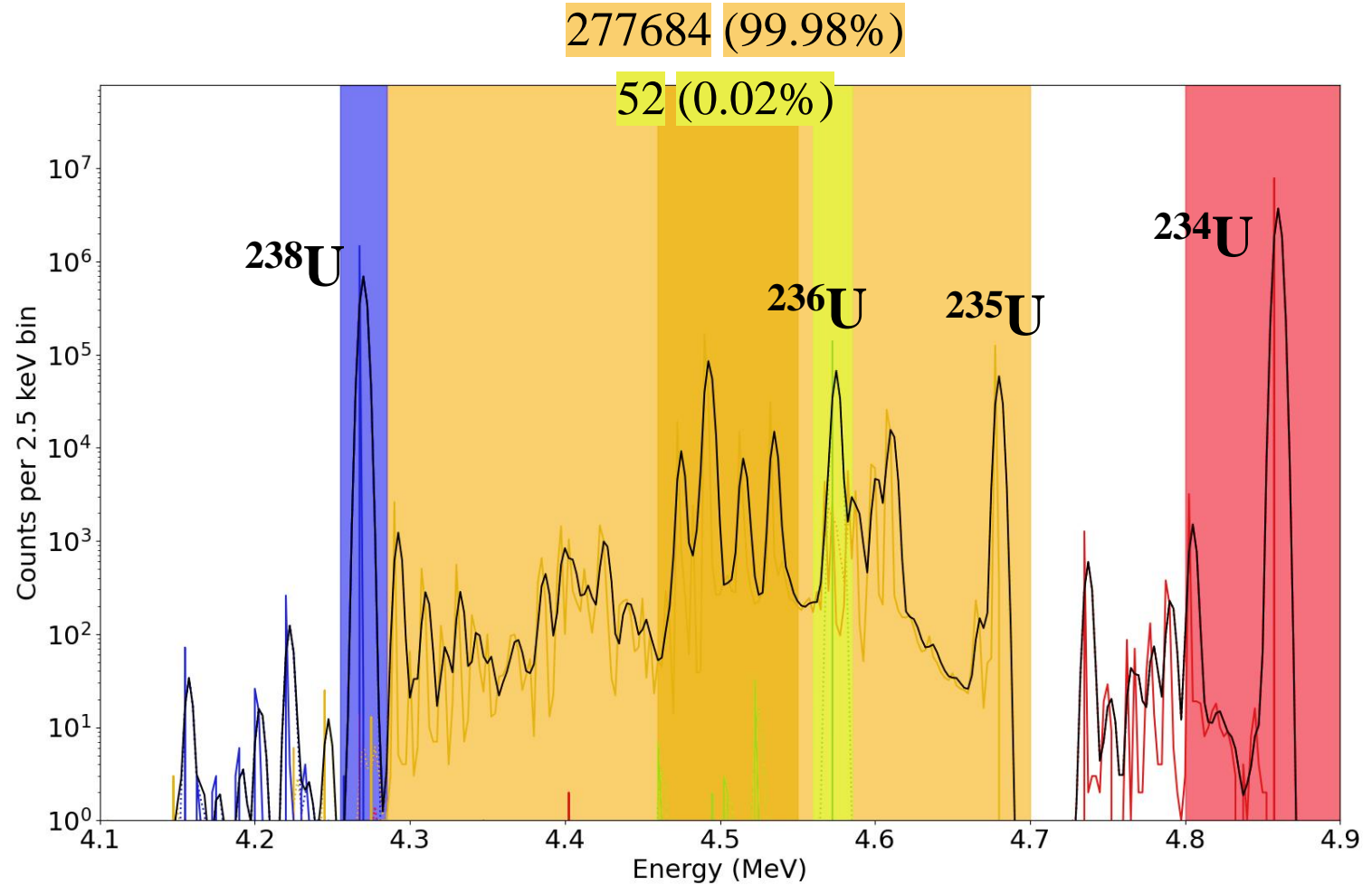
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^{238}U	^{238}U	1472092	>99.99%
^{238}U	^{235}U	32	0.002%
^{238}U	^{234}U	3	0.0002%

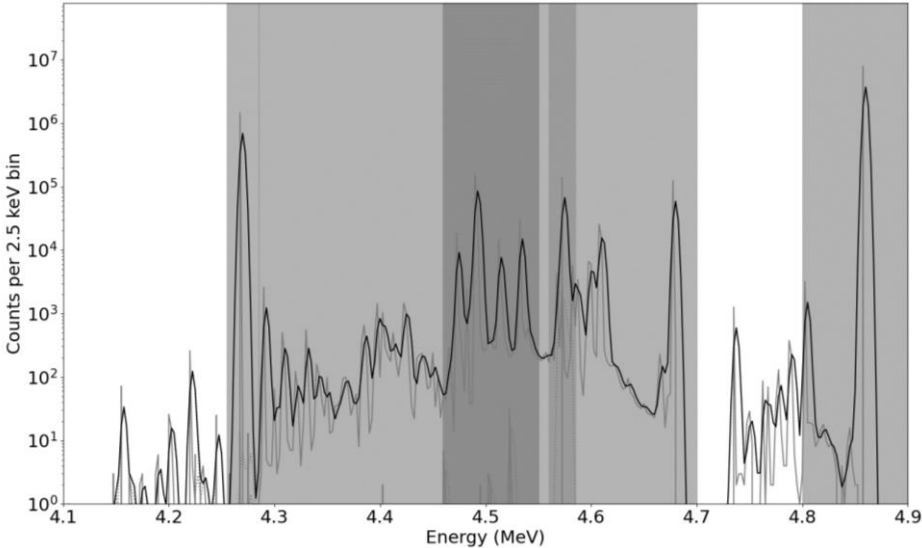
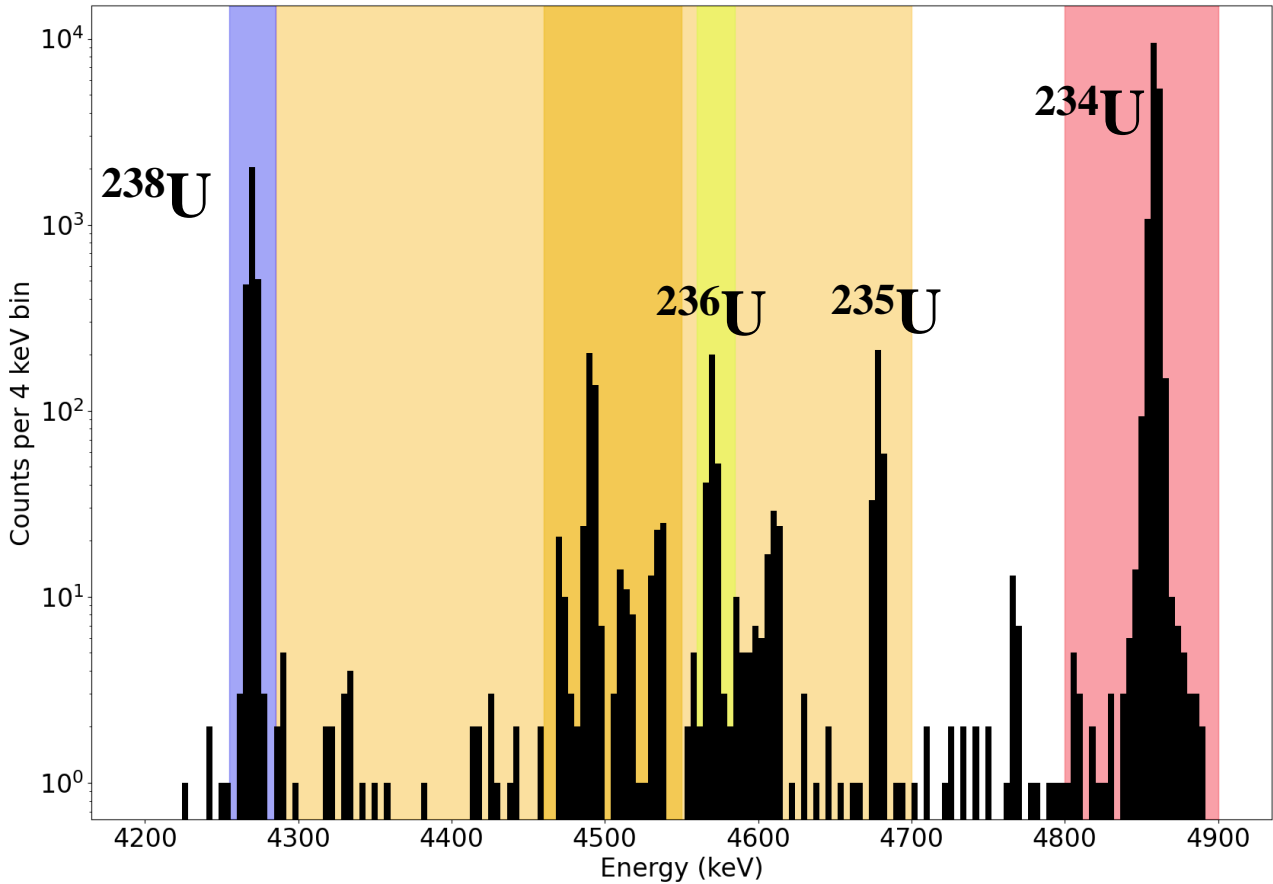


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^{236}U	^{236}U	141933	91%
^{236}U	^{235}U	13240	9%
^{238}U	^{238}U	1472092	>99.99%
^{238}U	^{235}U	32	0.002%
^{238}U	^{234}U	3	0.0002%
Inference	^{235}U	277684	99.98%
Inference	^{236}U	52	0.02%

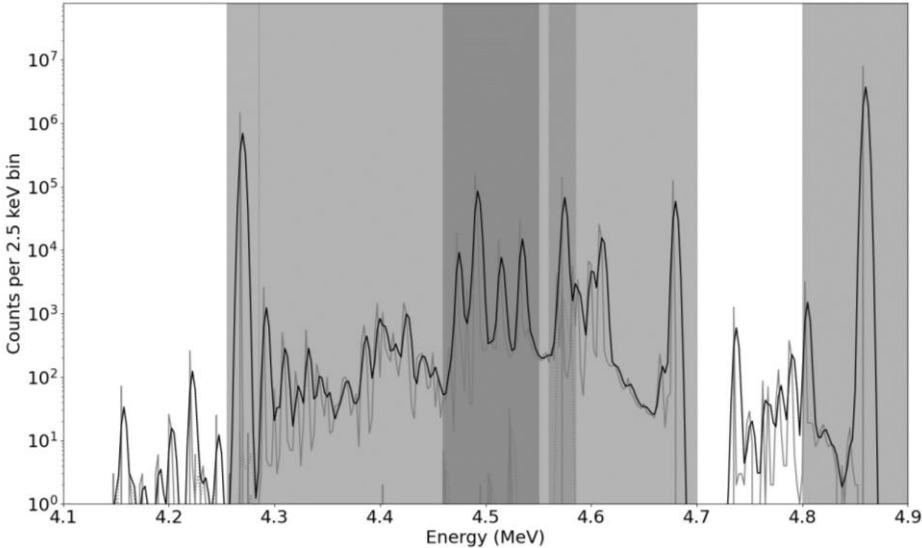
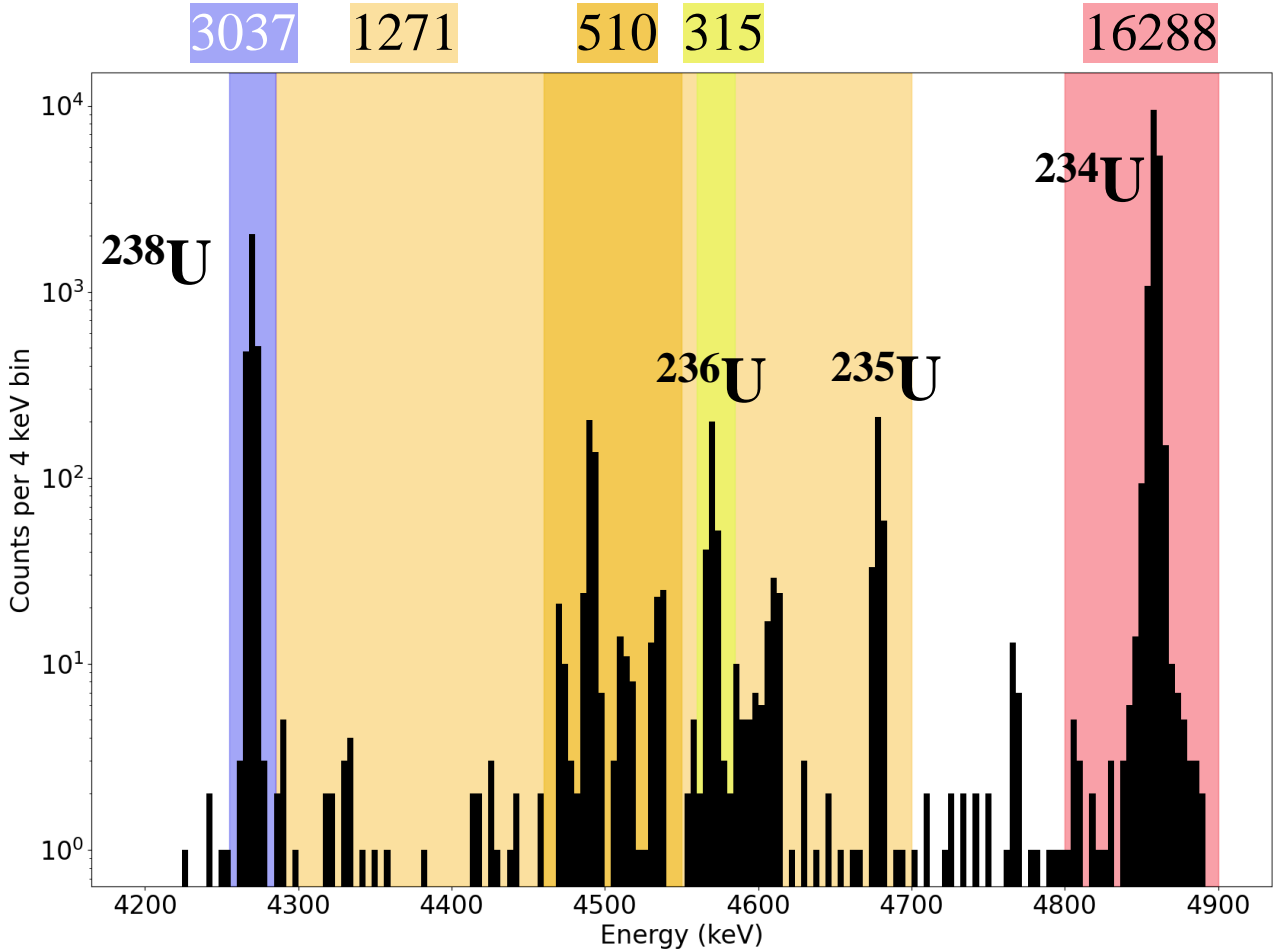


Analysis Using Geant4

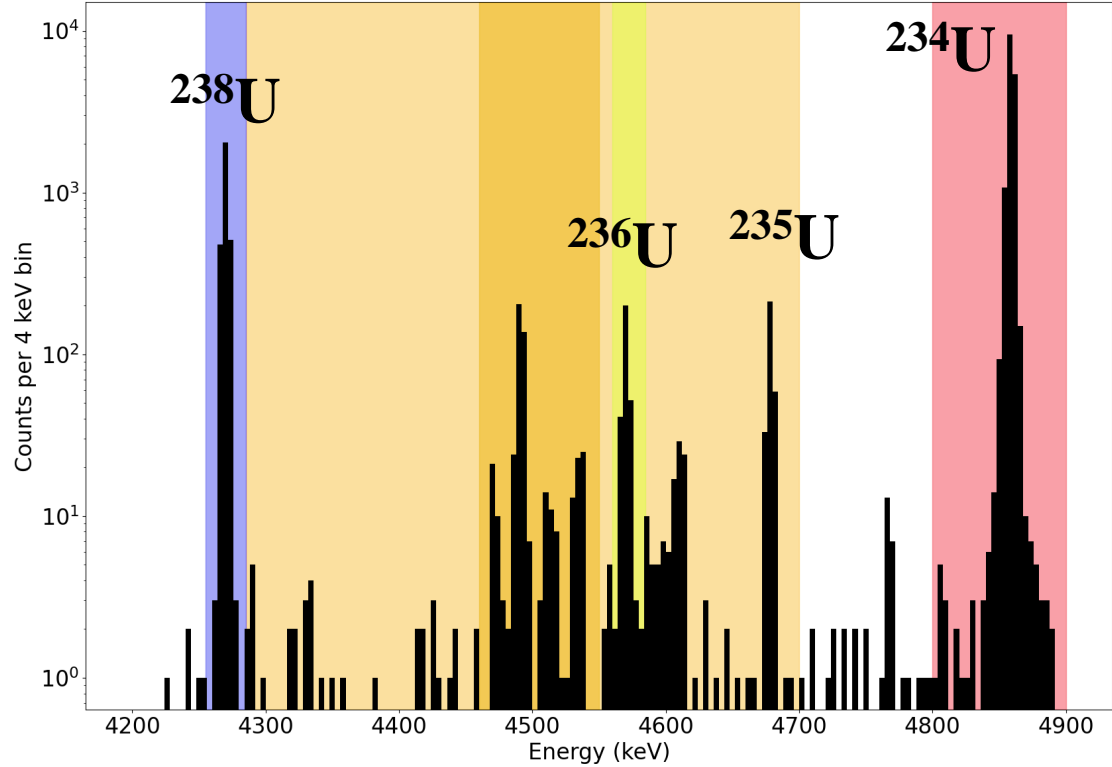


Analysis Using Geant4

For an experimental spectrum, we do not know the origin of the counts in a region.

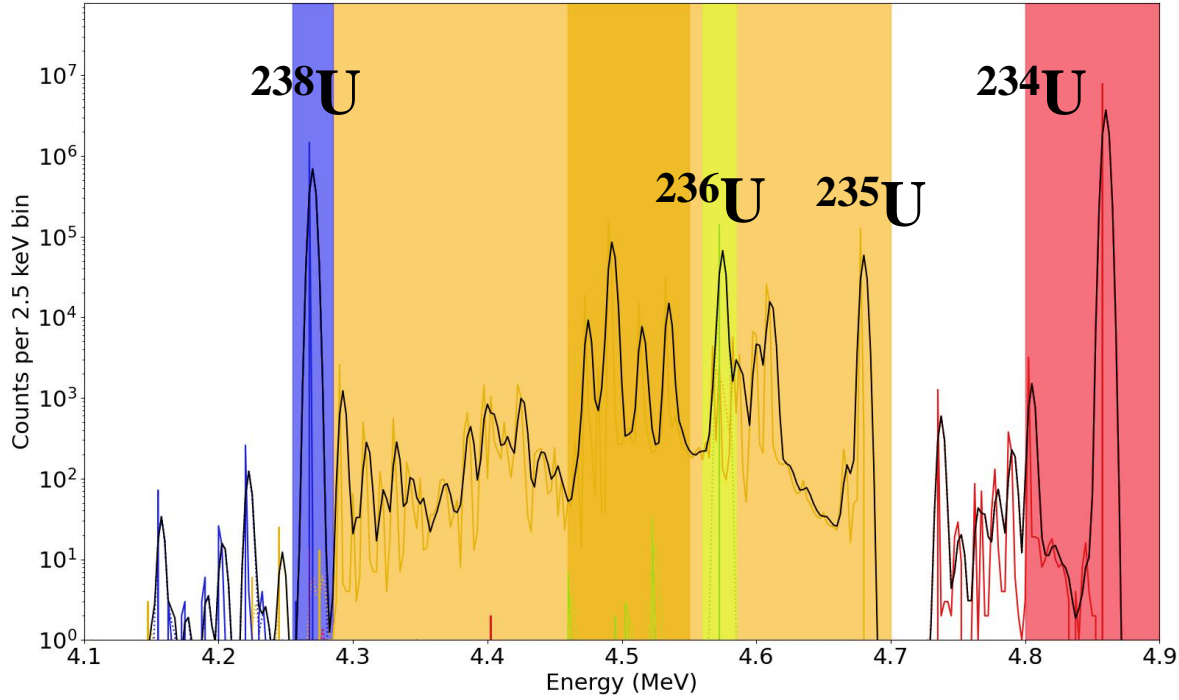


Analysis Using Geant4

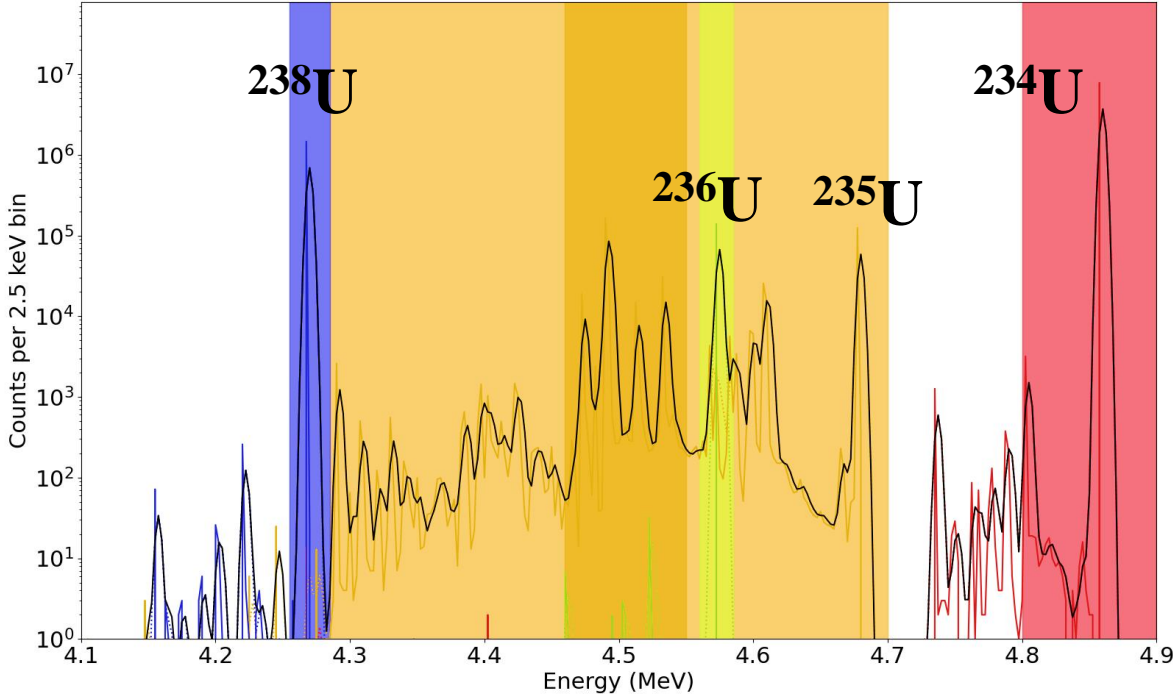
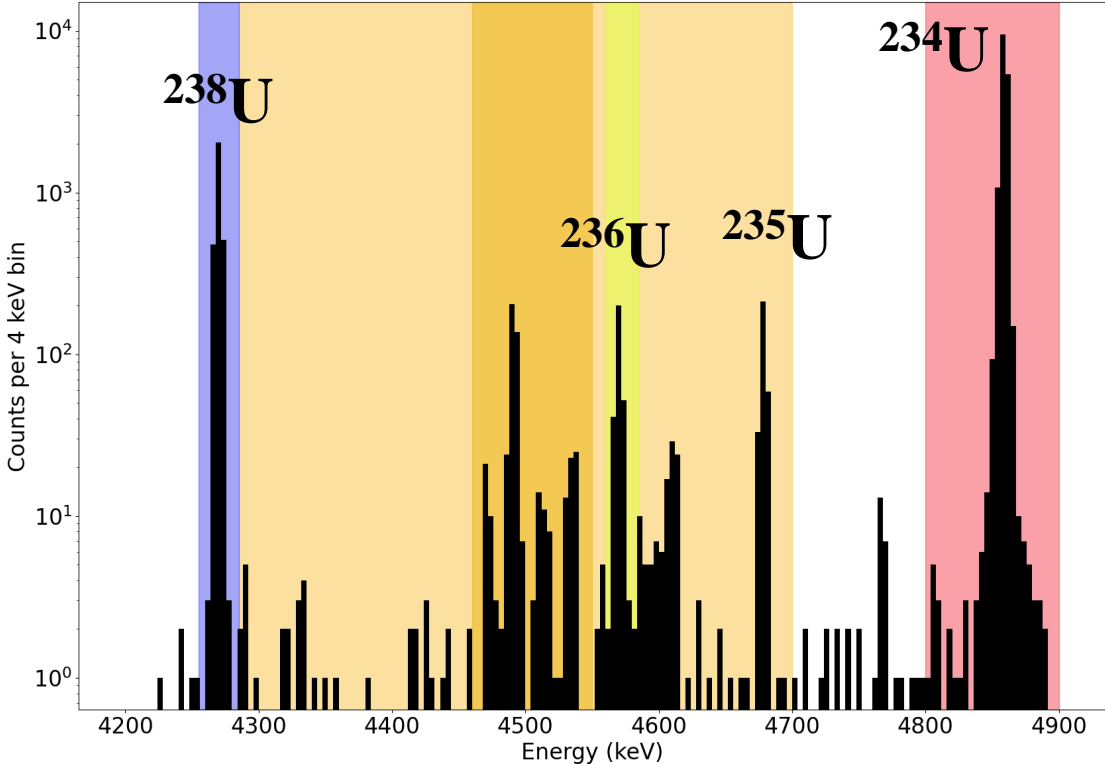


(235 in 236)

(235 in inf)

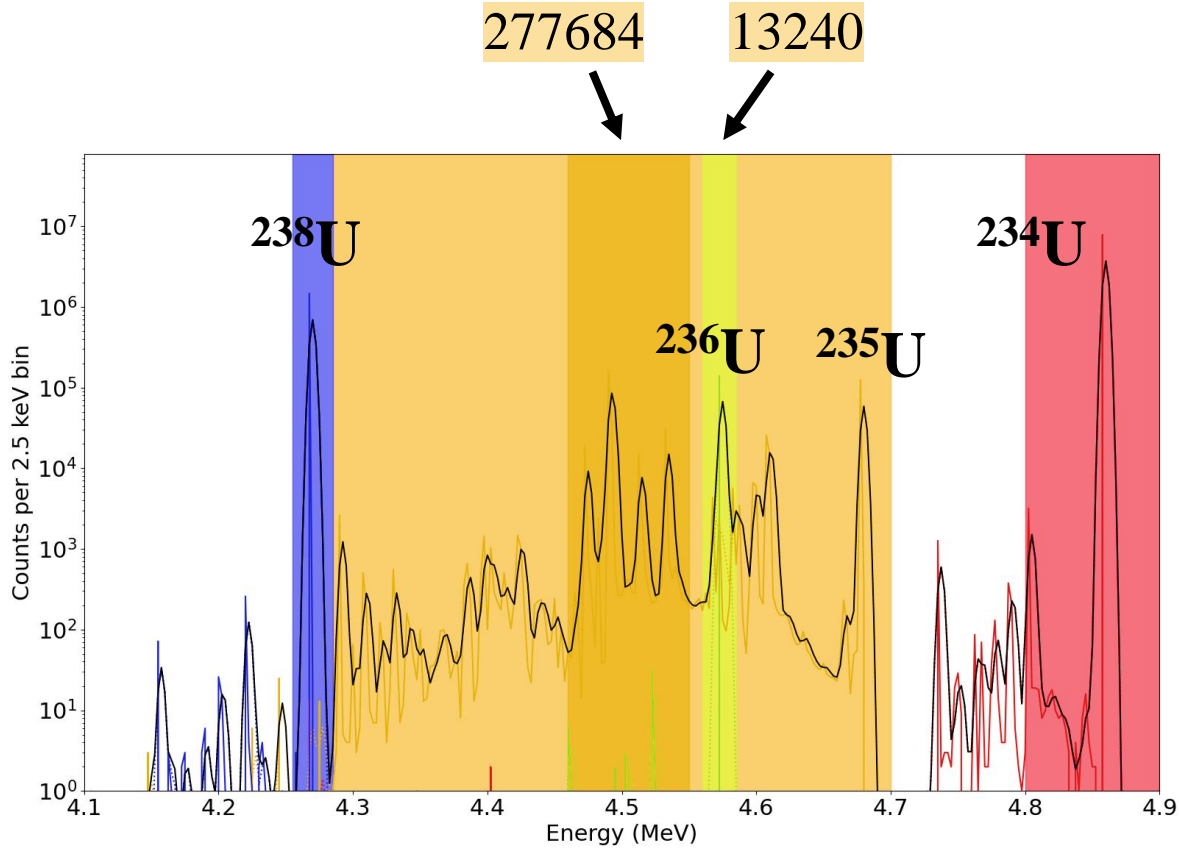
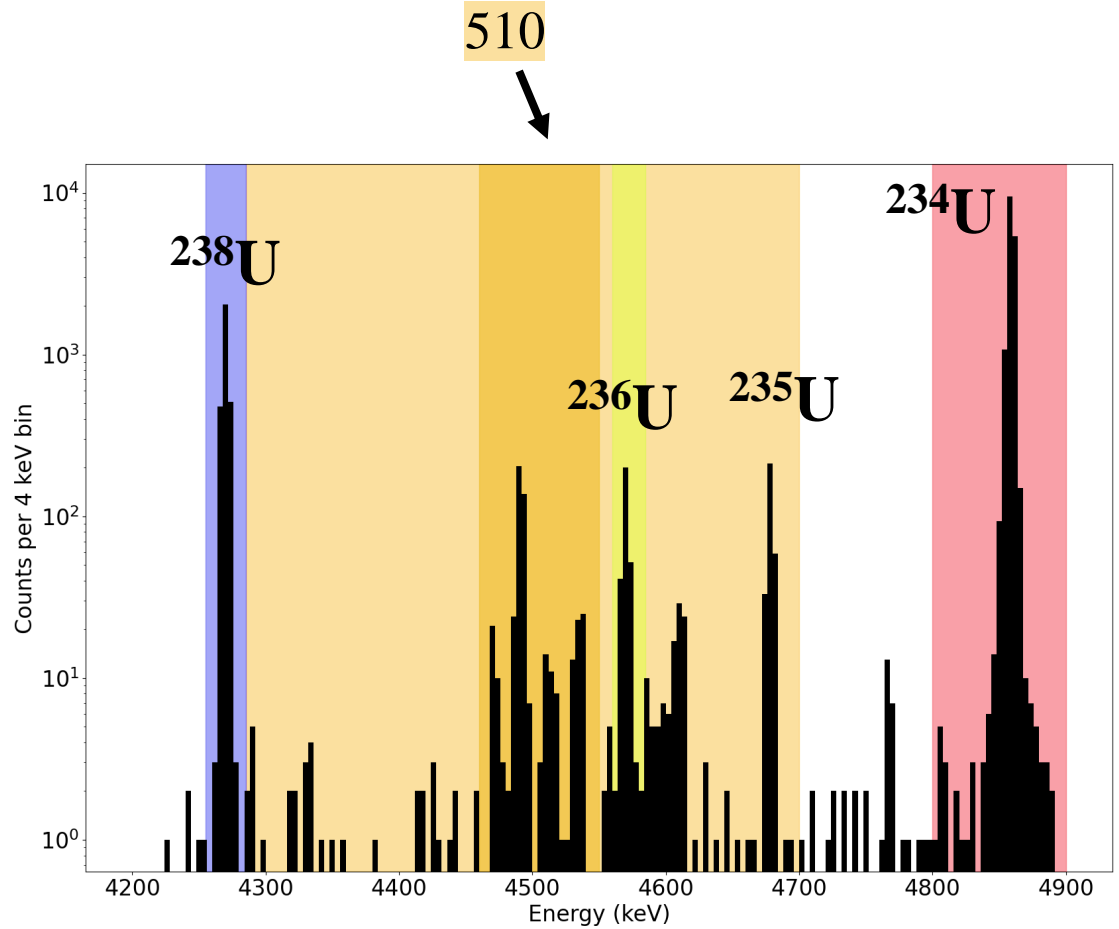


Analysis Using Geant4



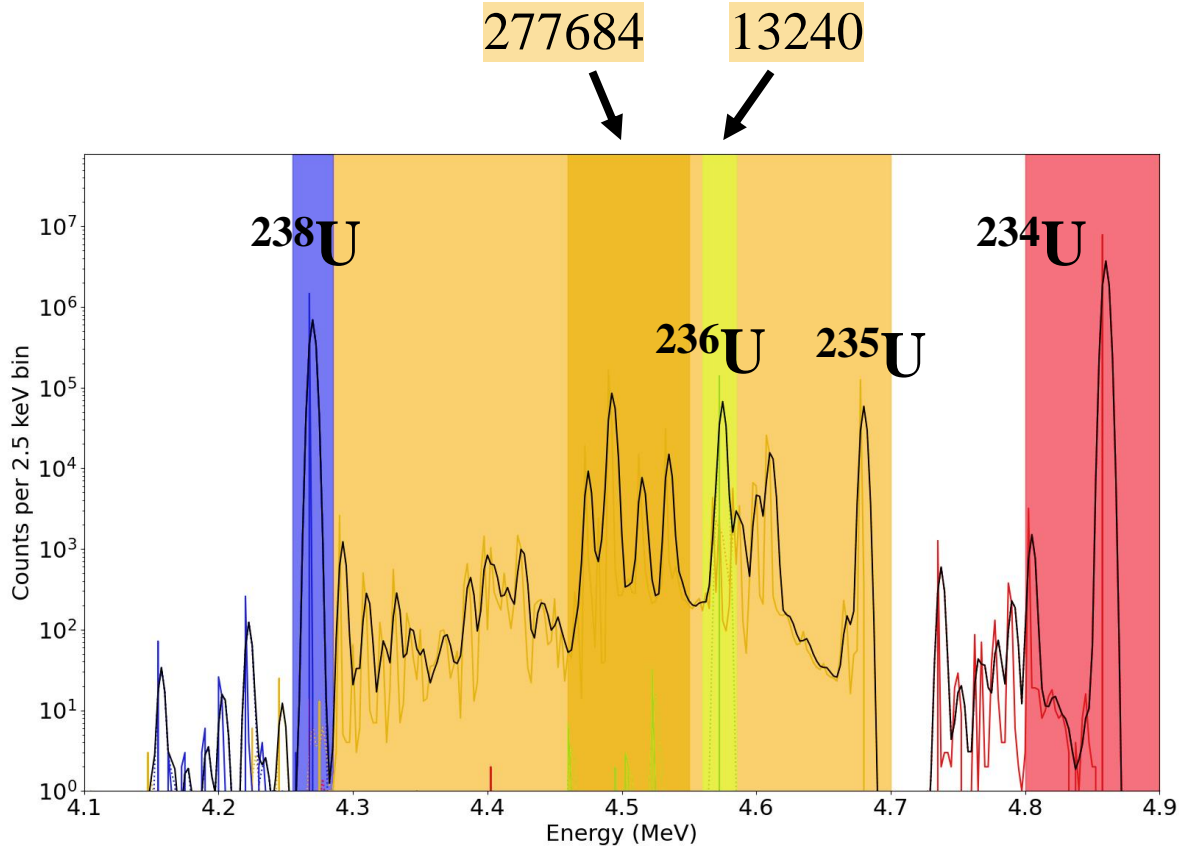
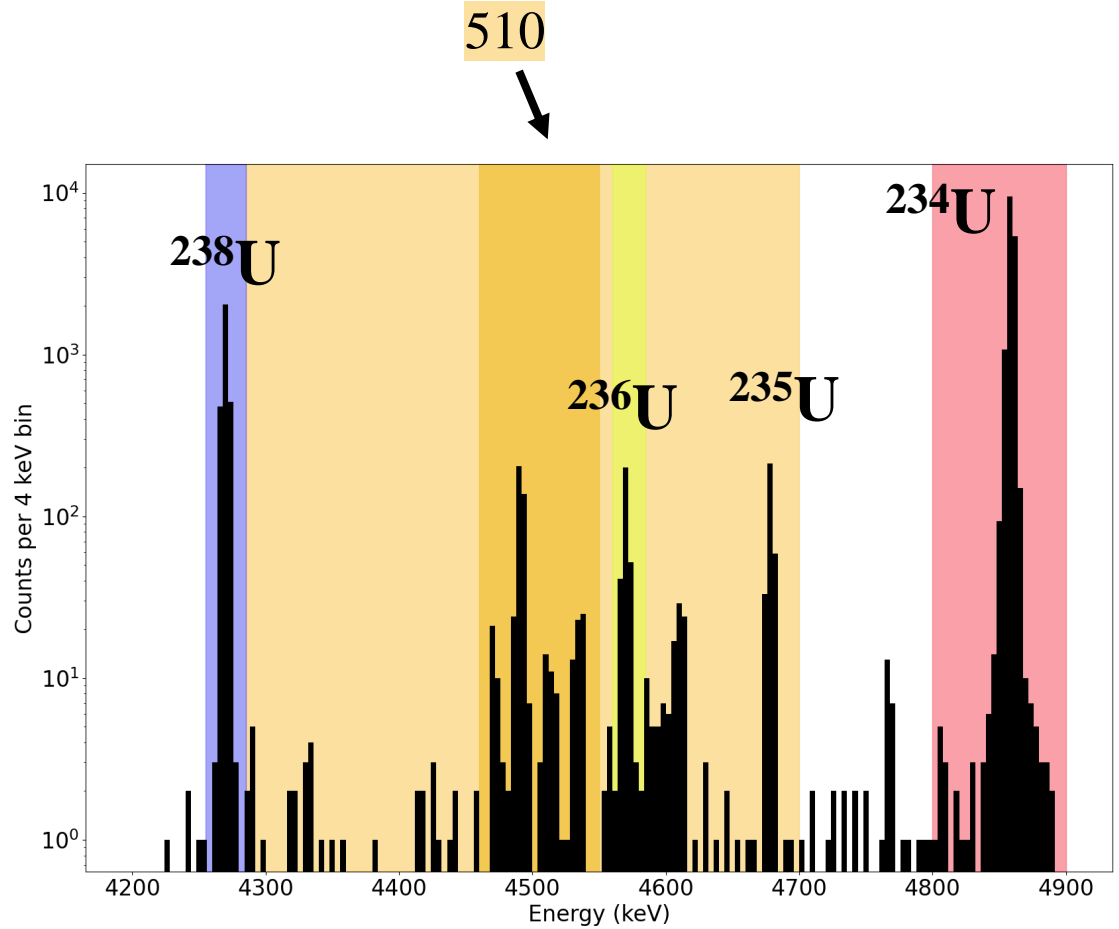
$$\frac{\text{(235 in 236)}_{\text{experimental}}}{\text{(235 in inf)}_{\text{experimental}}} = \frac{\text{(235 in 236)}_{\text{simulated}}}{\text{(235 in inf)}_{\text{simulated}}}$$

Analysis Using Geant4



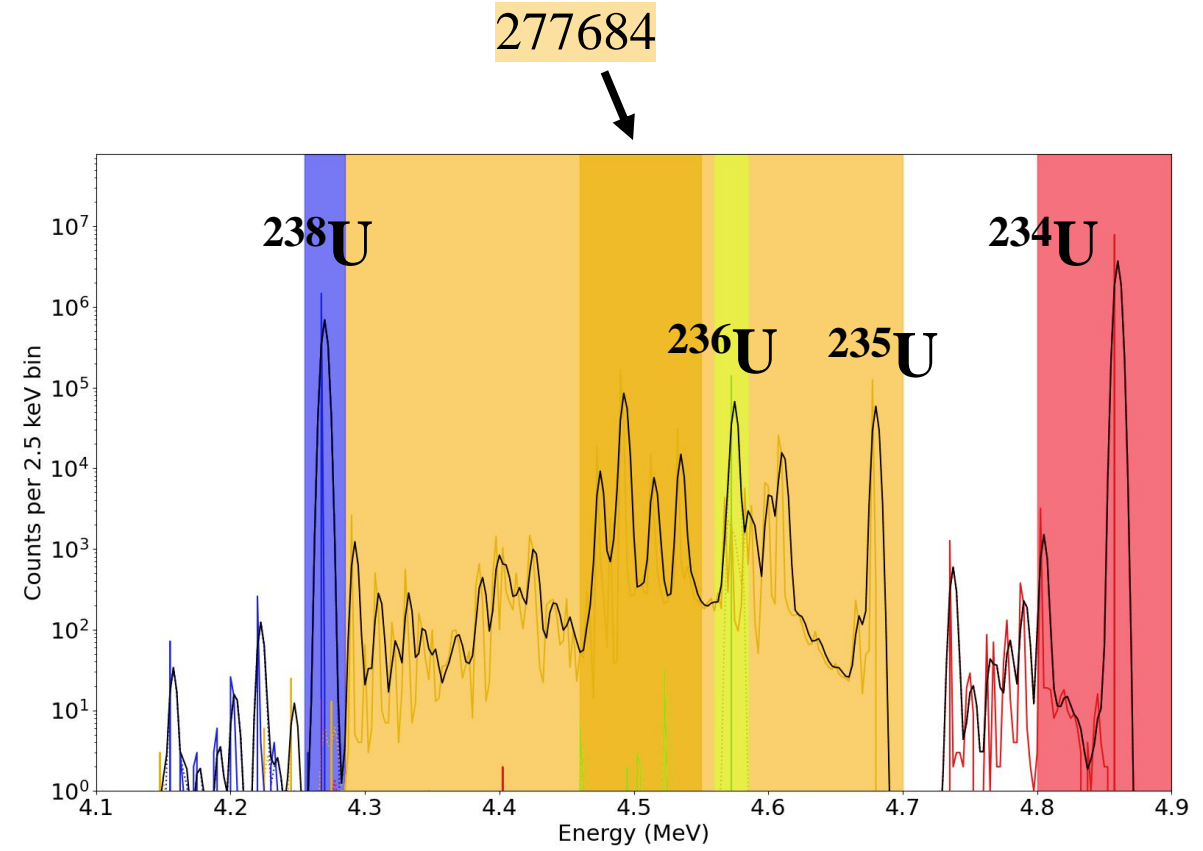
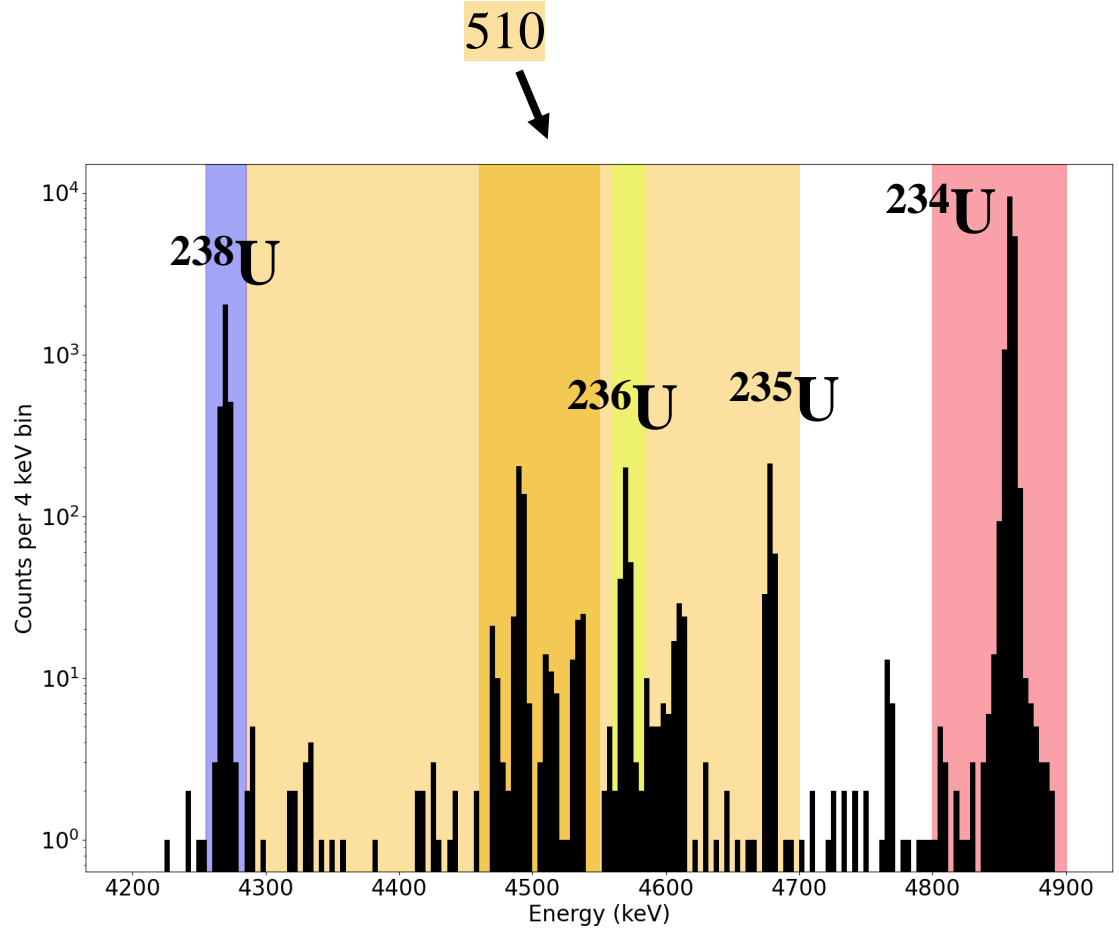
$$\frac{\text{(235 in 236)}_{\text{experimental}}}{\text{(235 in inf)}_{\text{experimental}}} = \frac{\text{(235 in 236)}_{\text{simulated}}}{\text{(235 in inf)}_{\text{simulated}}}$$

Analysis Using Geant4



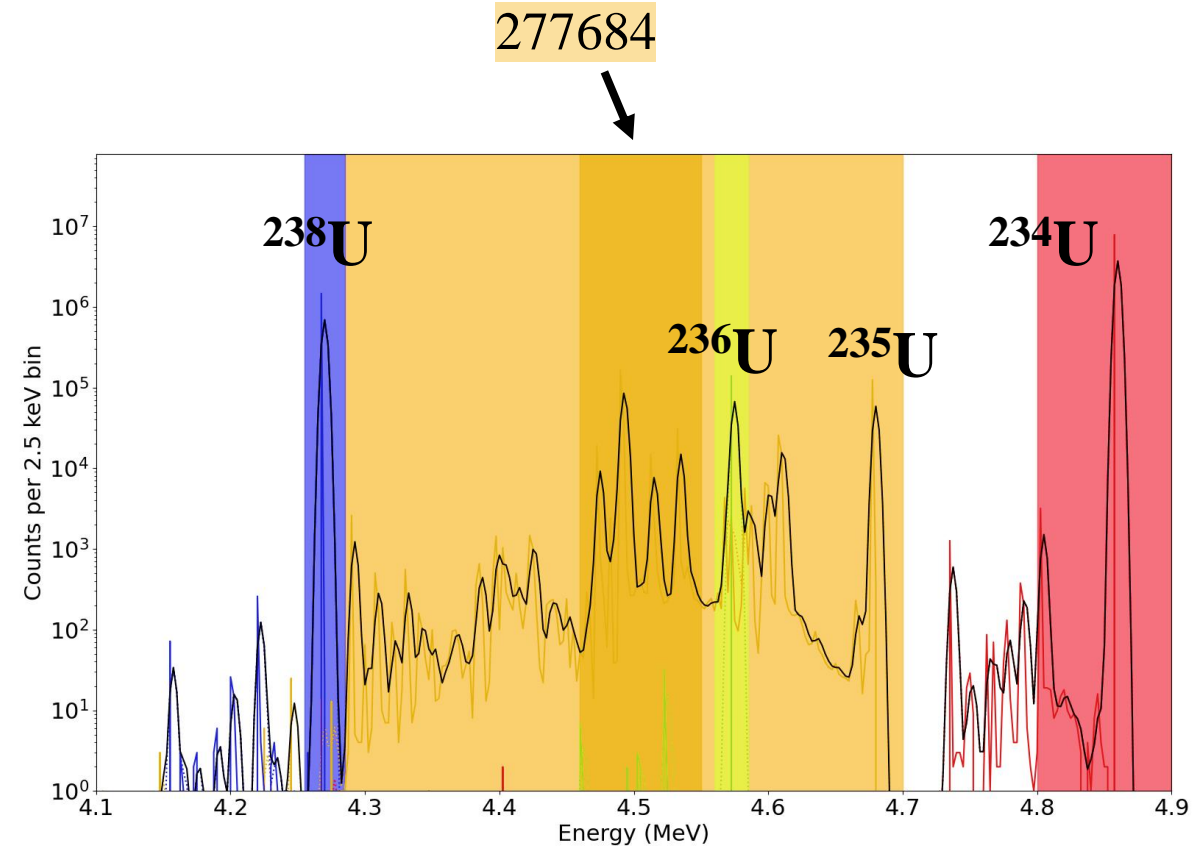
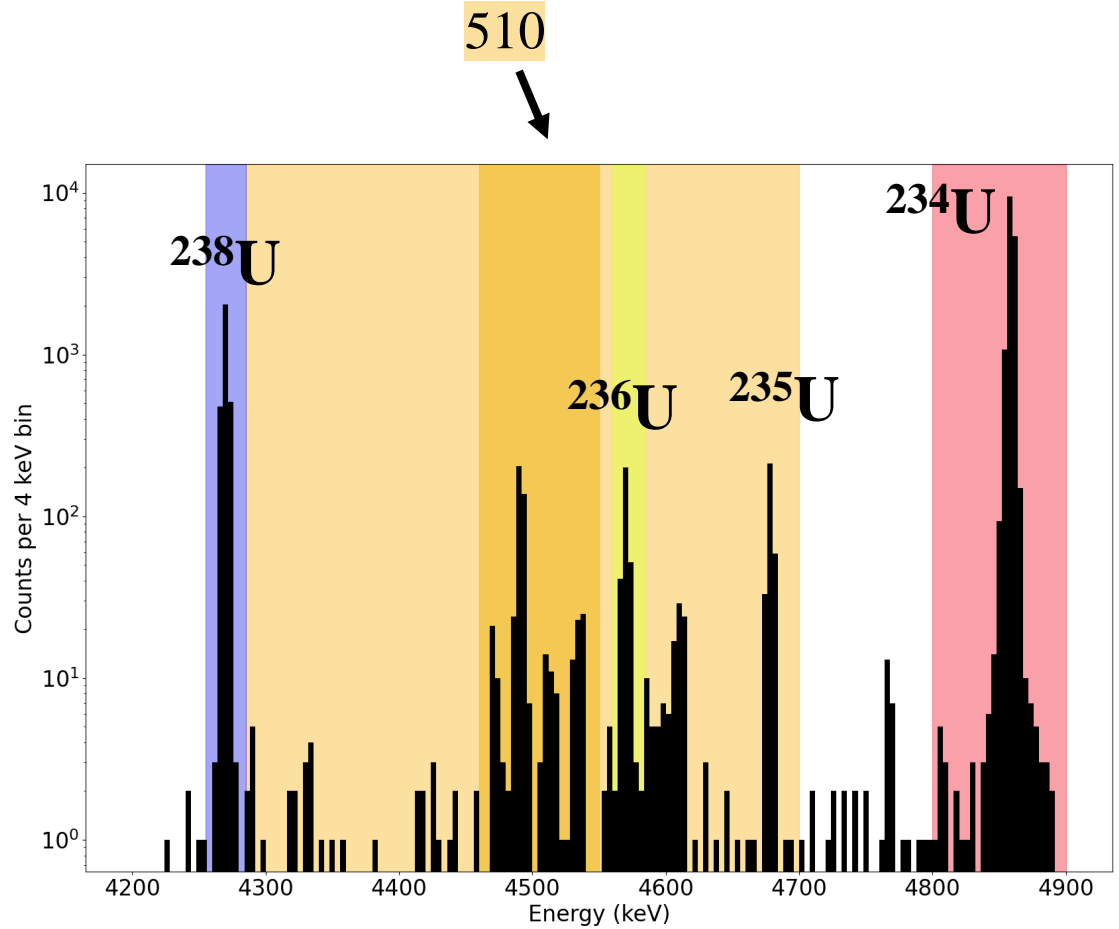
$$\frac{\text{(235 in 236)}_{\text{experimental}}}{\text{(235 in inf)}_{\text{experimental}}} = \frac{\text{(235 in 236)}_{\text{simulated}}}{\text{(235 in inf)}_{\text{simulated}}}$$

Analysis Using Geant4



$$\frac{\text{(235 in 236)}}{\text{(235 in inf)}}_{\text{experimental}} = \frac{\text{(13240)}}{\text{(235 in inf)}}_{\text{simulated}}$$

Analysis Using Geant4

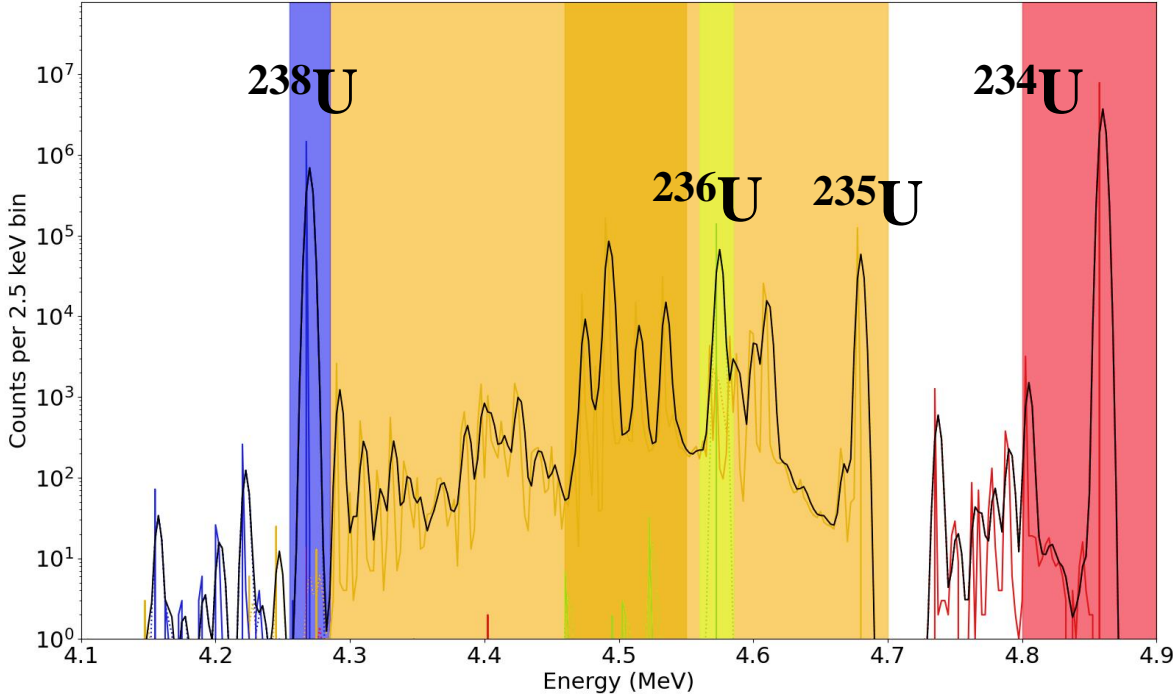
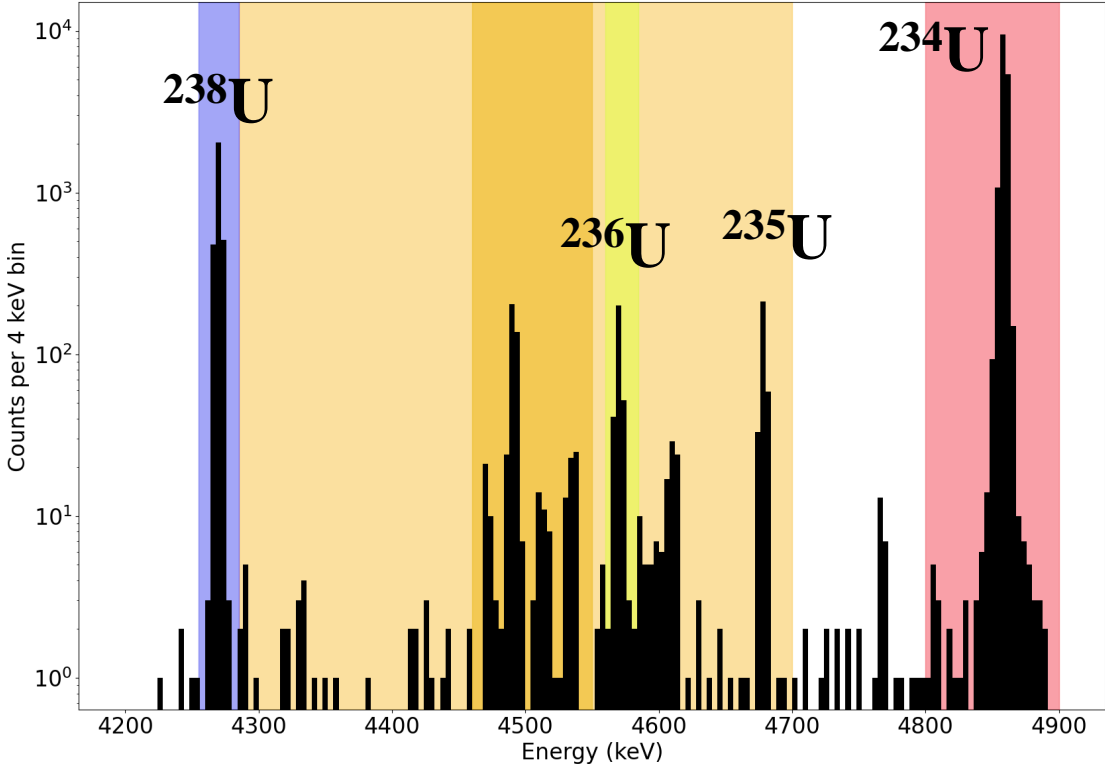


$$\frac{\text{(235 in 236)}}{\text{(235 in inf)}} = \frac{\text{(13240)}}{\text{(235 in inf)}}$$

experimental simulated

Analysis Using Geant4

510

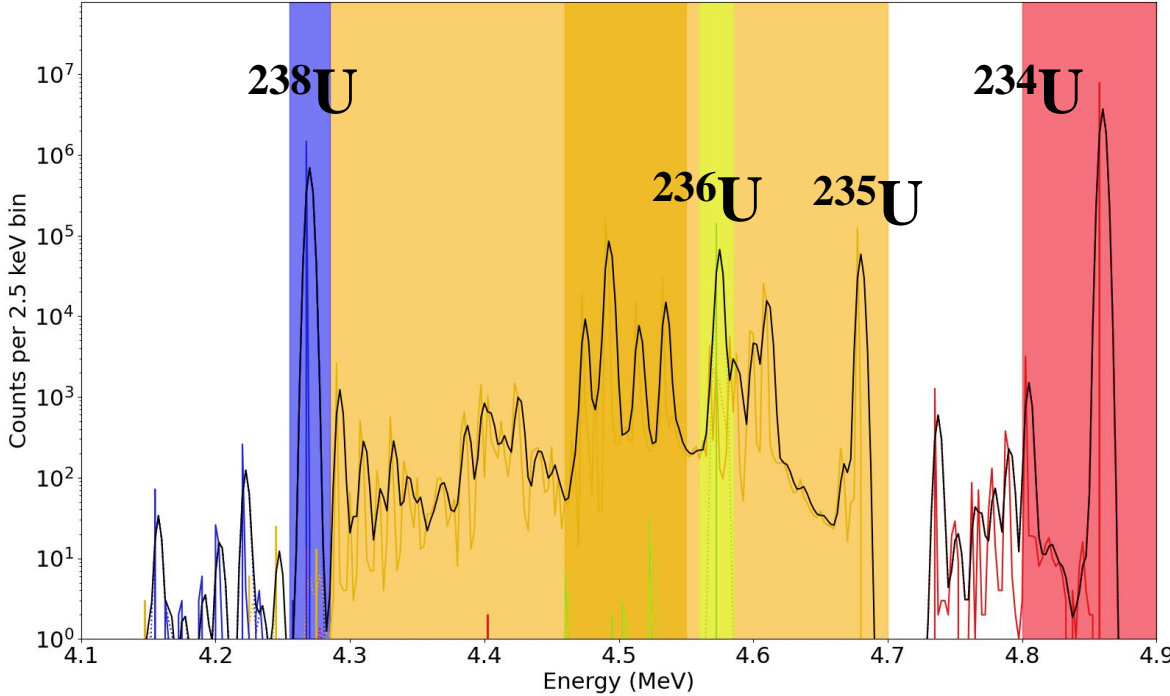
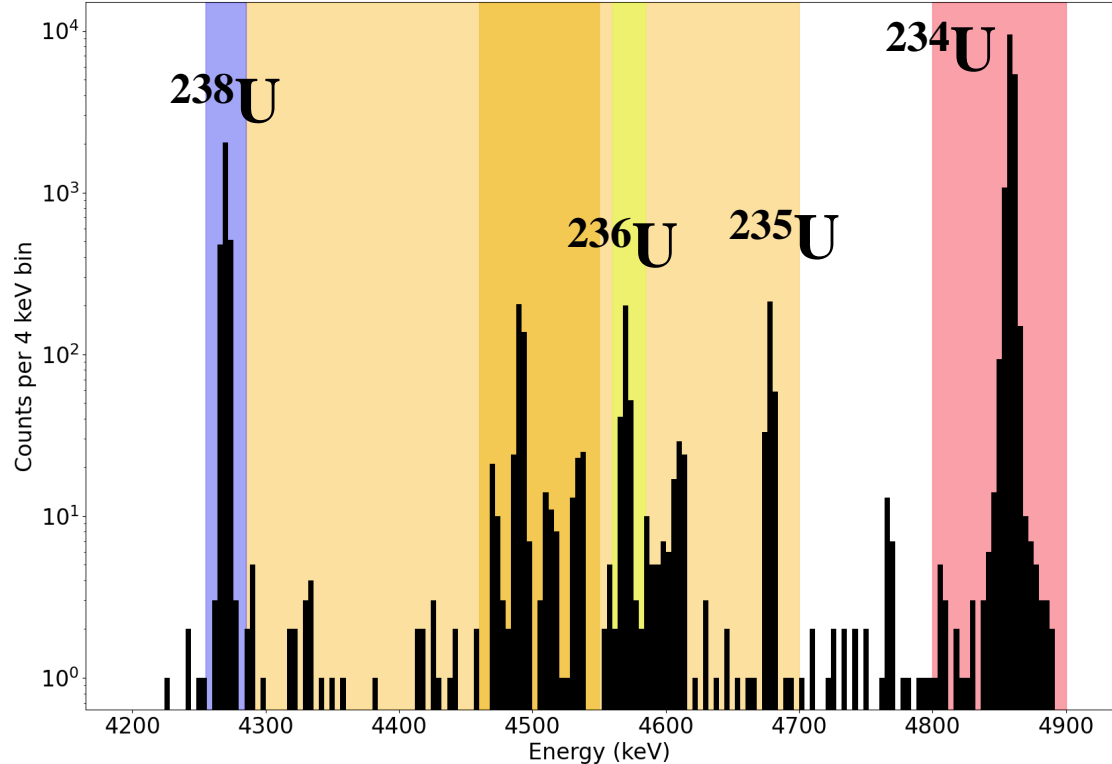


$$\frac{\text{(235 in 236)}}{\text{(235 in inf)}} = \frac{\text{(13240)}}{\text{(277684)}}$$

experimental simulated

Analysis Using Geant4

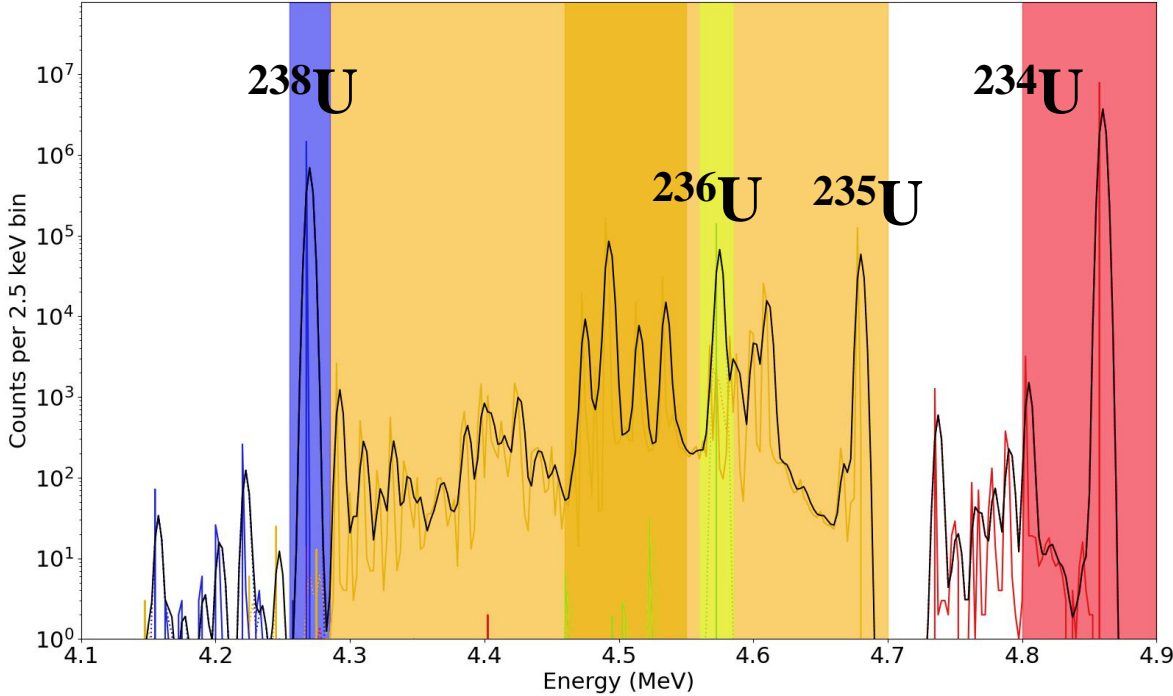
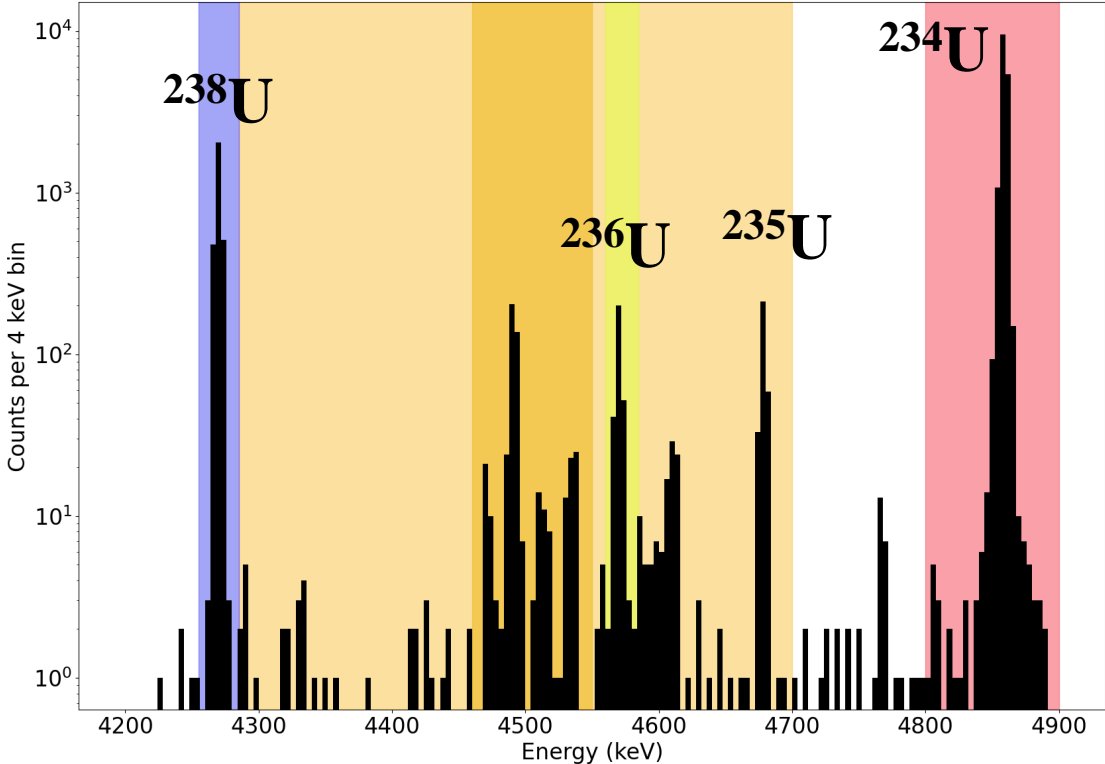
510



$$\frac{\text{(235 in 236)}}{\text{(235 in inf)}} = \frac{\text{(13240)}}{\text{(277684)}}$$

experimental simulated

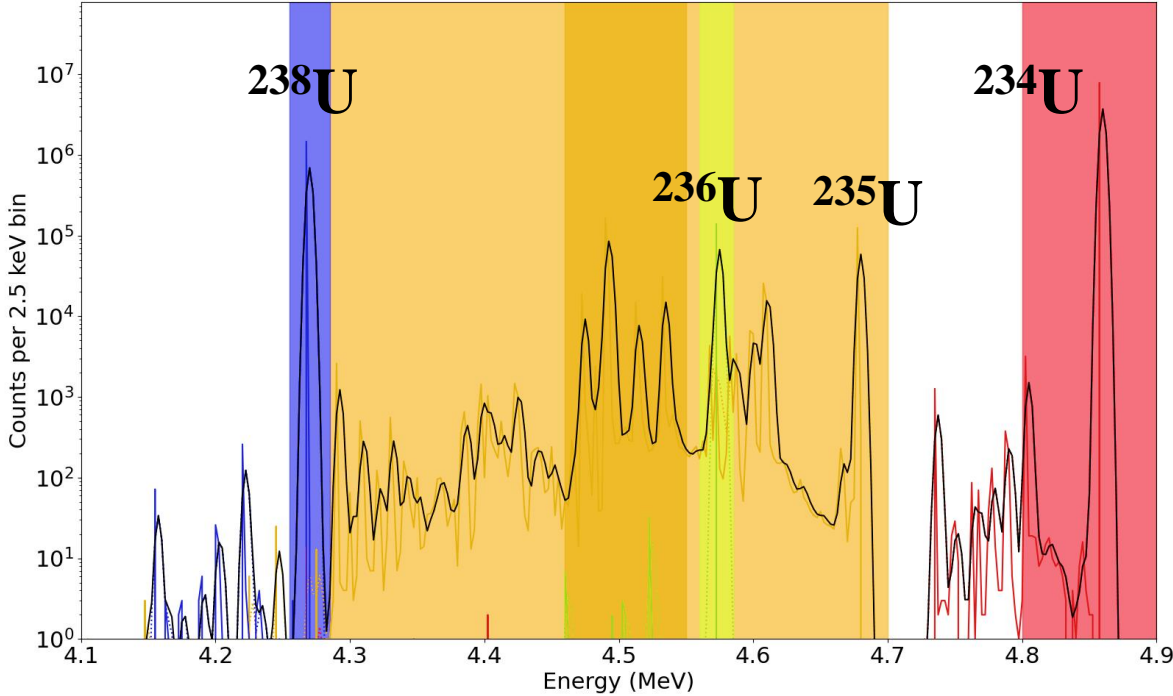
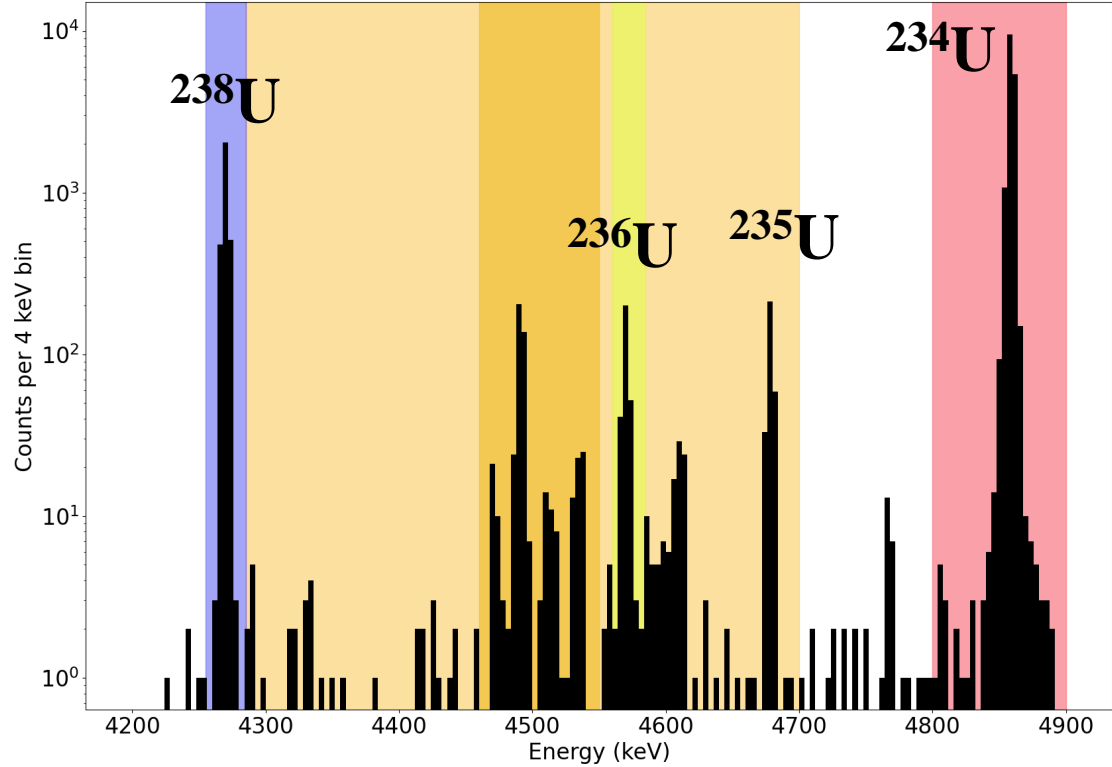
Analysis Using Geant4



$$\frac{\text{(235 in 236)}}{\text{(510)}} = \frac{\text{(13240)}}{\text{(277684)}}$$

experimental simulated

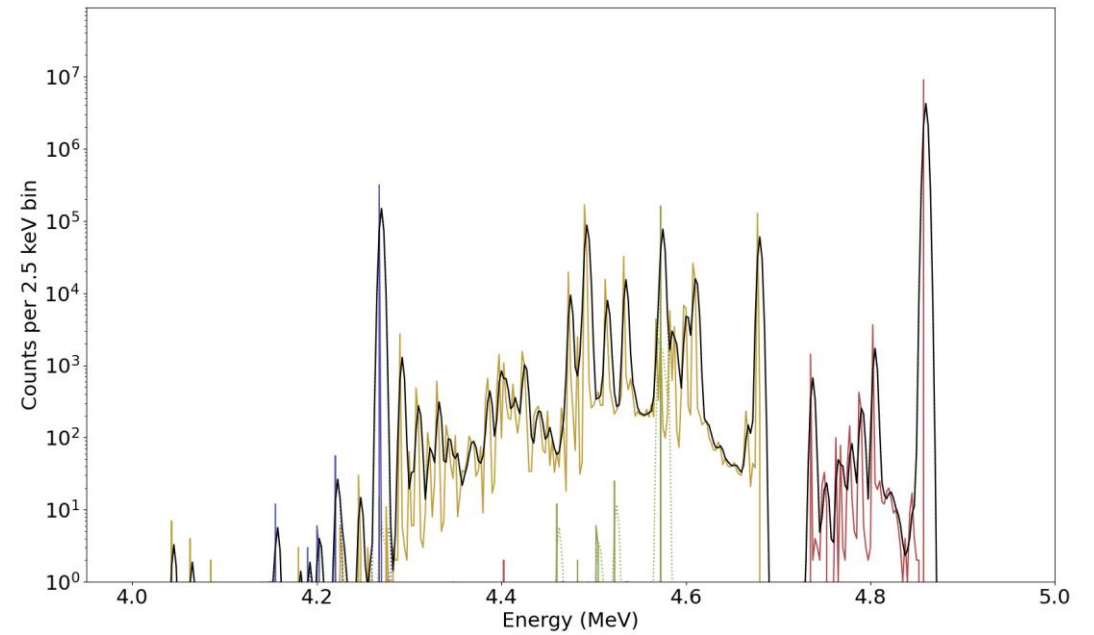
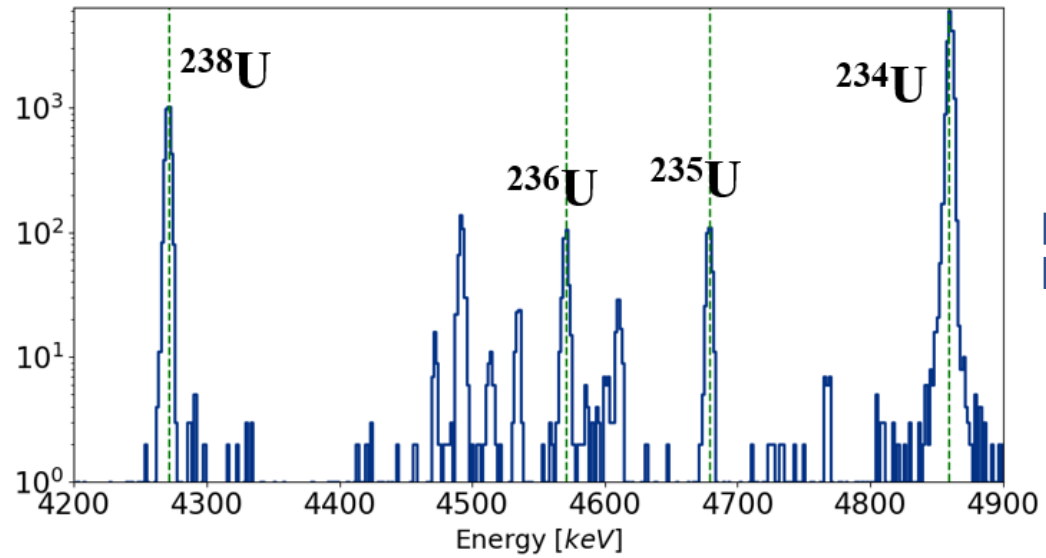
Analysis Using Geant4



(235 in 236) = 24.3

experimental

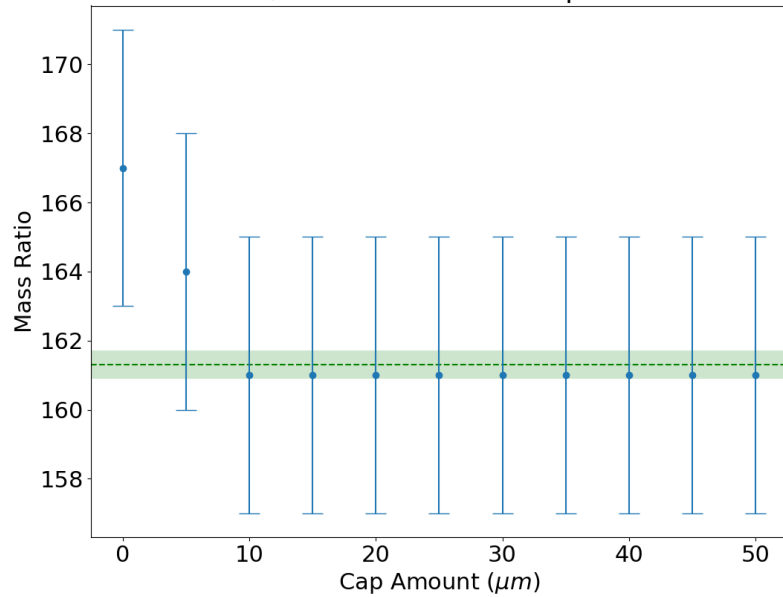
Best-Case Analysis



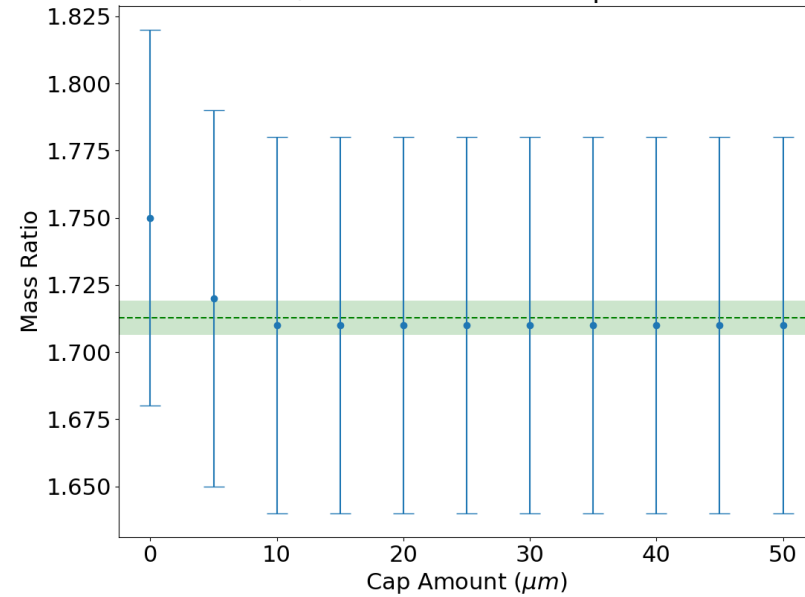
Best-Case Analysis

20% Enriched

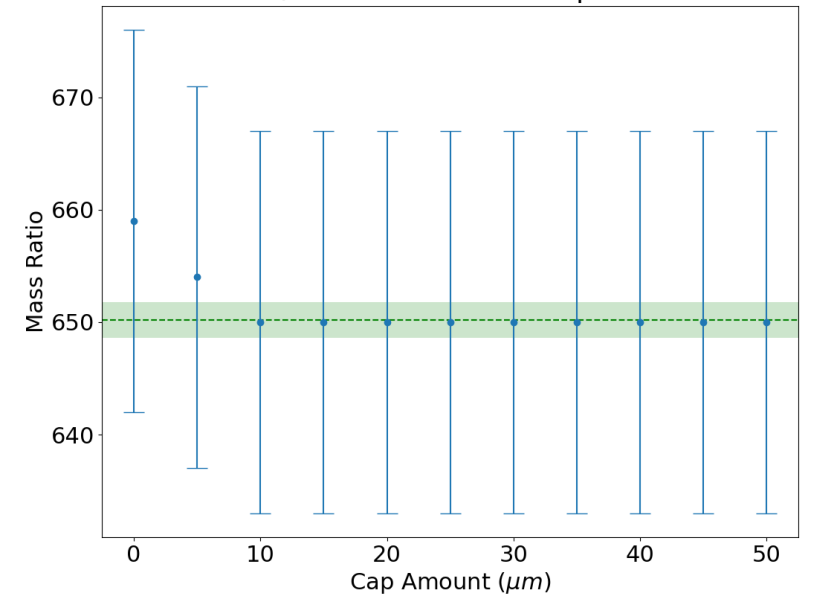
$^{235}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



$^{236}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



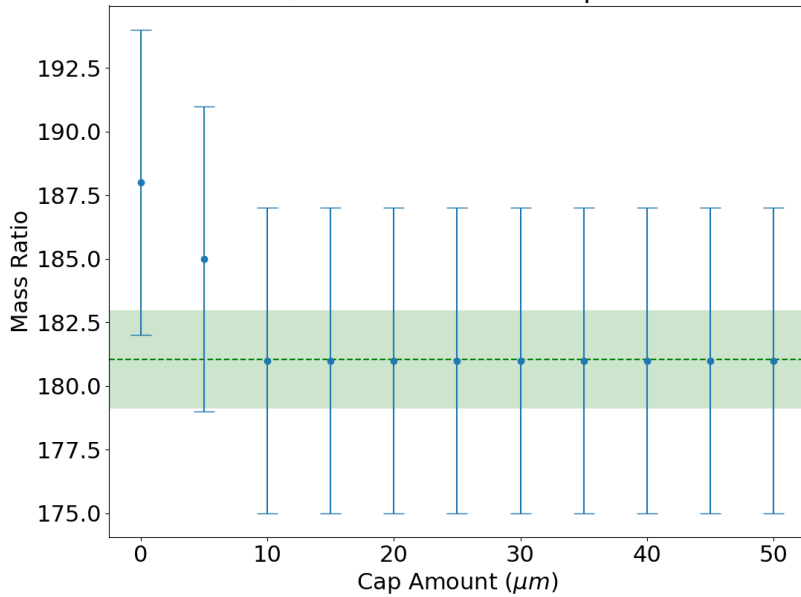
$^{238}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



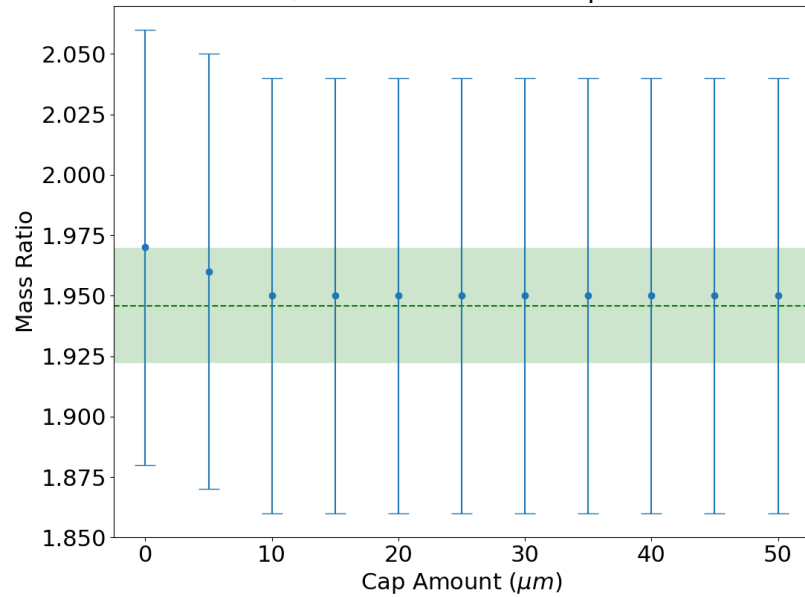
Best-Case Analysis

1.5% Enriched

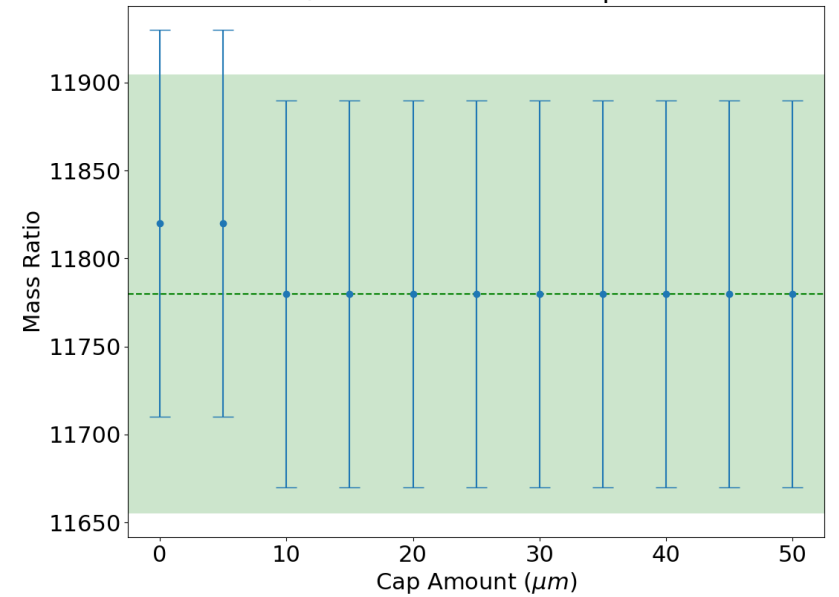
$^{235}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



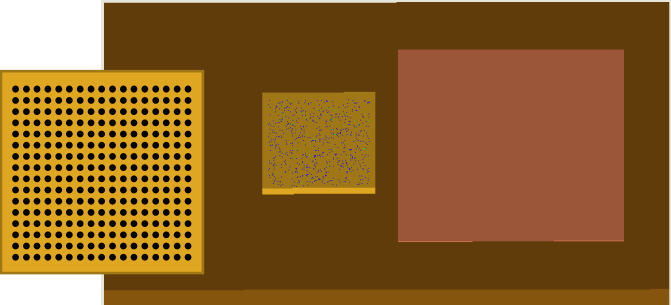
$^{236}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



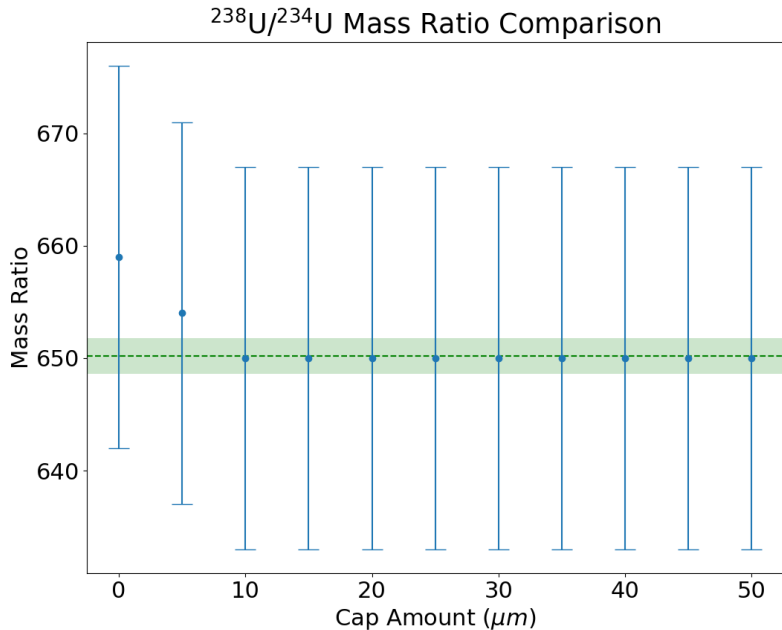
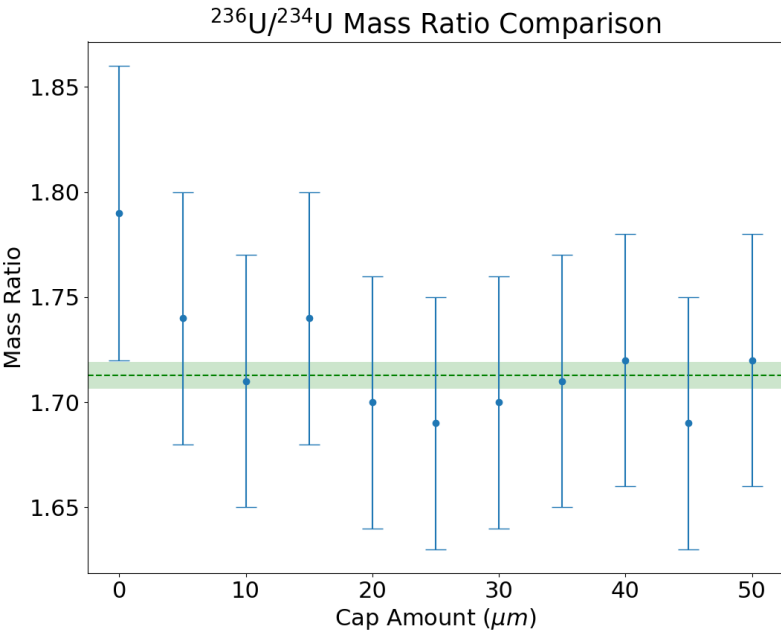
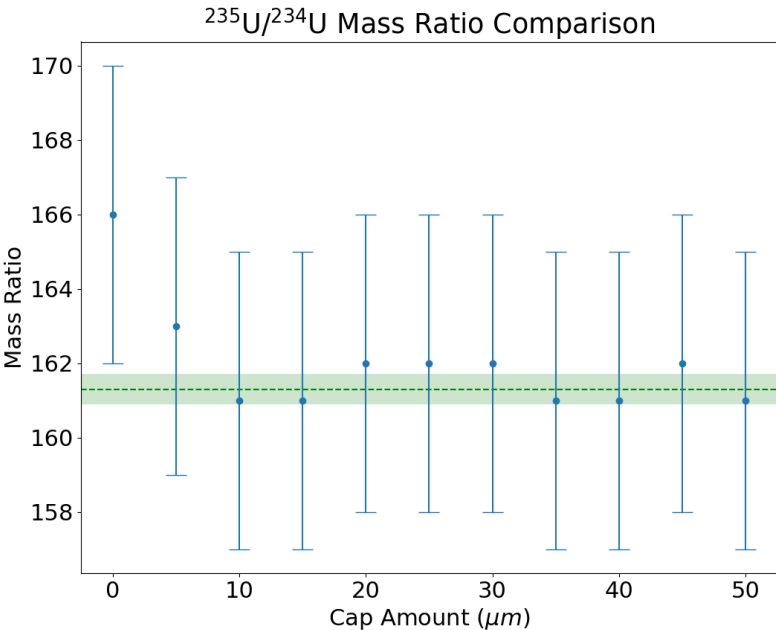
$^{238}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



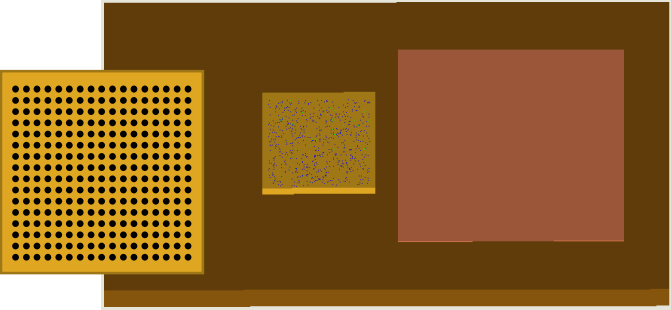
Analysis Results



20% Enriched

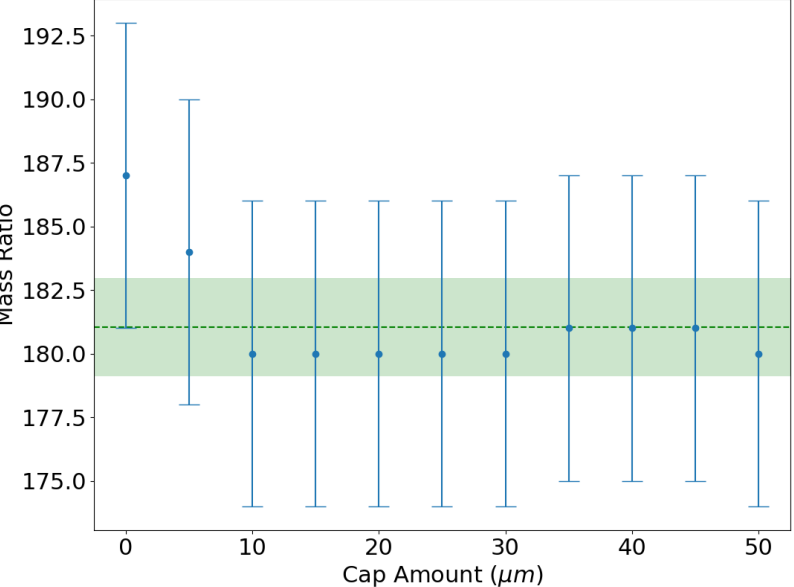


Analysis Results

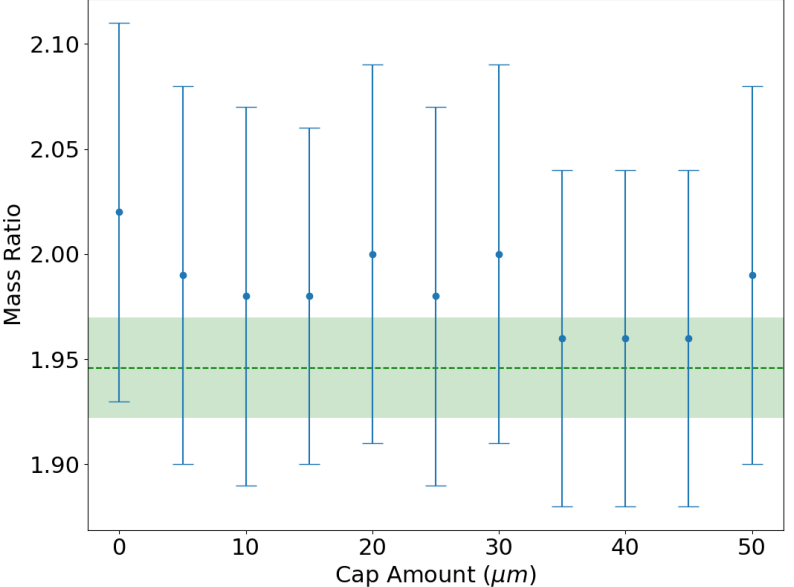


1.5% Enriched

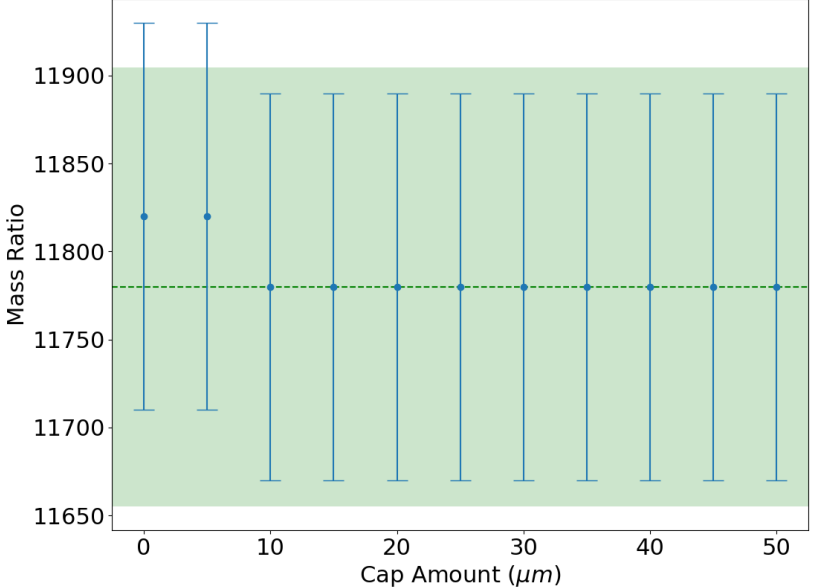
$^{235}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



$^{236}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



$^{238}\text{U}/^{234}\text{U}$ Mass Ratio Comparison



Future Plans

- Repeat simulation with plutonium sources to see if similar patterns related to capping emerge
- Develop peak fitting algorithm for alternative uranium analysis method
- Simulate aluminum absorbers as alternative to gold as a potential new method of measuring spectra with DES