LA-UR-16-26068
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Title: Special Nuclear Material Gamma-Ray Signatures for Reachback Analysts

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Intended for: LSS Spectroscopy Course, VA 2016

Issued: 2016-08-29 (rev.1)

Special Nuclear Material Gamma-Ray Signatures for Reachback Analysts

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August 2016

Introduction

- Special Nuclear Material (SNM):
  - Highly-Enriched Uranium (HEU): Key Nuclide is ^{235}U
  - Plutonium: Key Nuclide is ^{239}Pu
  - ^{237}Np
  - ^{233}U
  - Others but they are less common

- All of above listed nuclides are ‘fissionable’. Of these, all <u>except</u> ^{237}Np are *fissile*.
- SNM is very dense in metal form and is composed of elements with high atomic numbers.
- Gram for gram SNM is not very radioactive compared to common sources like ^{137}Cs (Note: except ^{233}U items with high (ppm) ^{232}U concentrations).

General Signatures of SNM

- HEU: gammas mainly at lower energies
- Plutonium:
  - Gammas from low to medium/high energy
  - Neutrons (60,000 n/s/kg for WGPu)
  - It can be warm or hot to the touch in sufficient quantity
- ^{237}Np: gammas mainly at medium energies
- ^{233}U: gammas
  - Direct gammas at medium energies
  - Most intense gammas from ^{232}U (at ppm concentrations) cover a wide range up to high energies

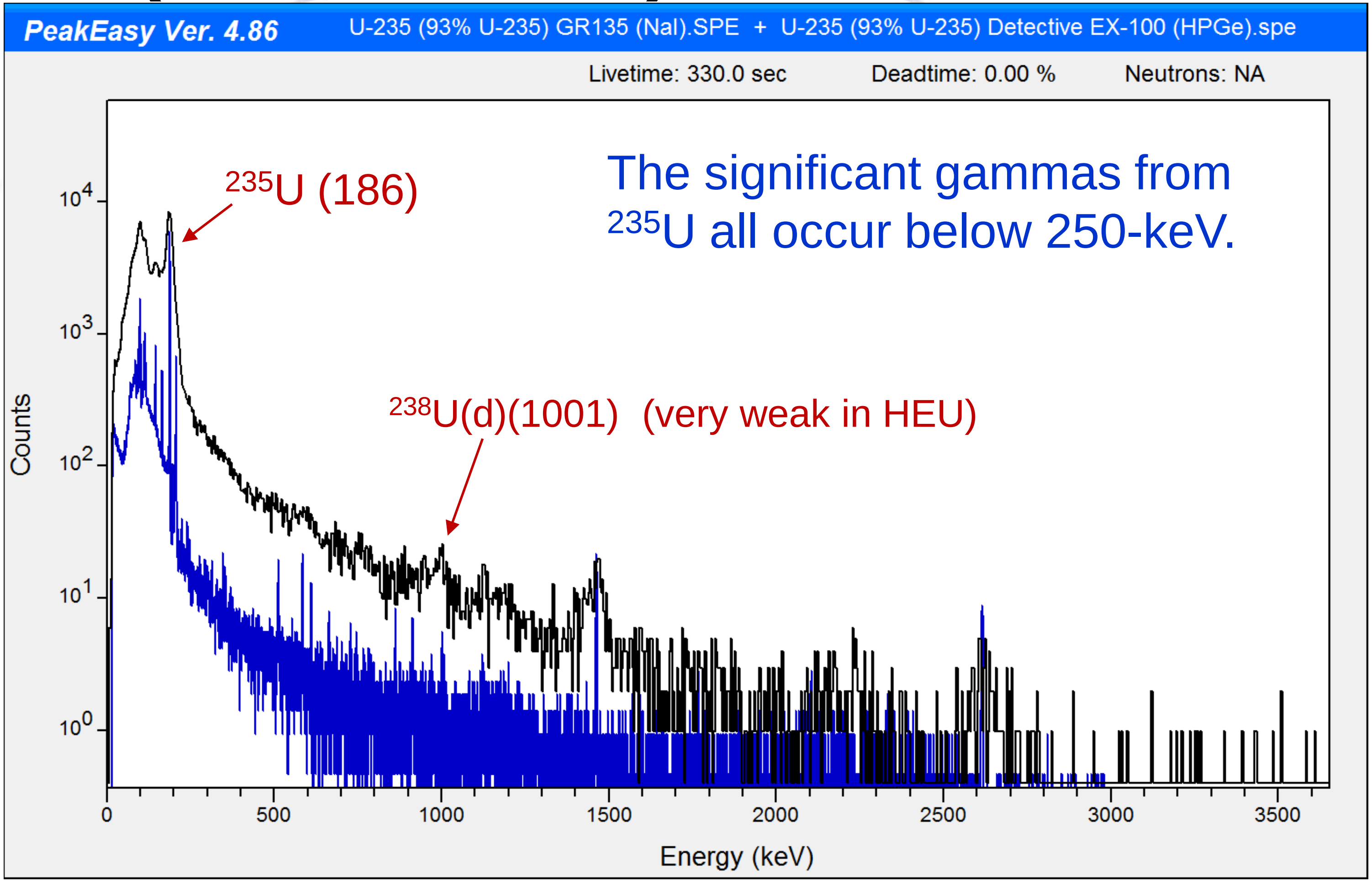
Low Energy: < ~250 keV
Medium Energy: ~250 – 1000 keV
High Energy: > ~1000 keV

Isotopes and Enrichments of Uranium

- Only three isotopes of uranium are found in nature
  - ^{238}U (99.2745(60) at%) (99.2836 wt%)
  - ^{235}U (0.7200(12) at%) (0.7110 wt%)
  - ^{234}U (0.0055(5) at%) (0.0054 wt%)

- Categories of Enrichment (E = % of ^{235}U)
  - Depleted Uranium (DU) E < 0.72 %
  - Natural Uranium (NU) E = 0.72 %
  - Enriched Uranium E > 0.72%
    - Low Enriched Uranium (LEU) 0.72% < E < 20.0 %
    - High Enriched Uranium (HEU) E ≥ 20.0 %
- Other important uranium isotopes include ^{232}U and ^{233}U

HEU (NaI vs. HPGe)



Why is estimating enrichment by simply comparing the 186 and 1001-keV peaks not recommended?

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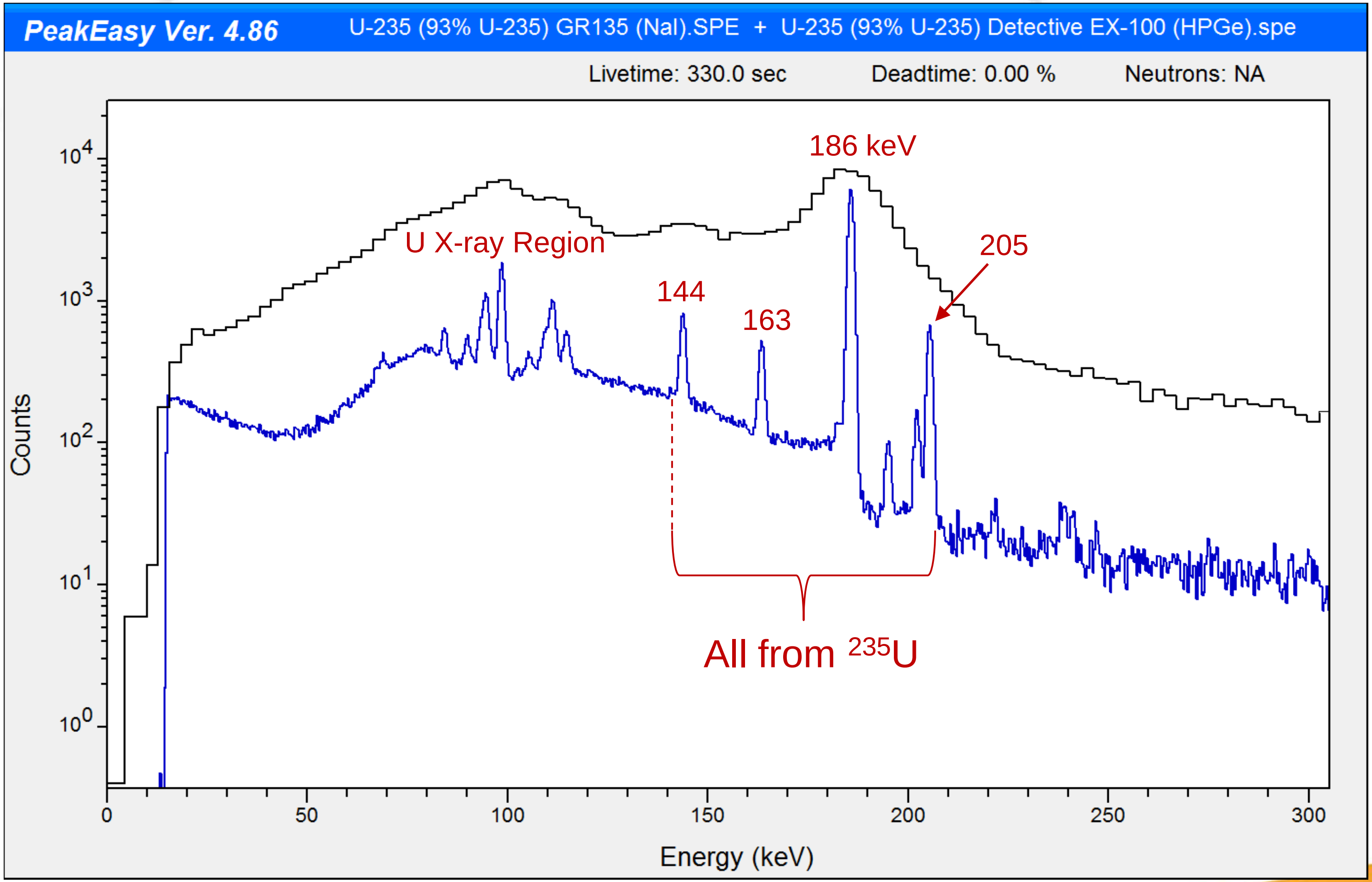
HEU (Low-Energy, Log Scale)



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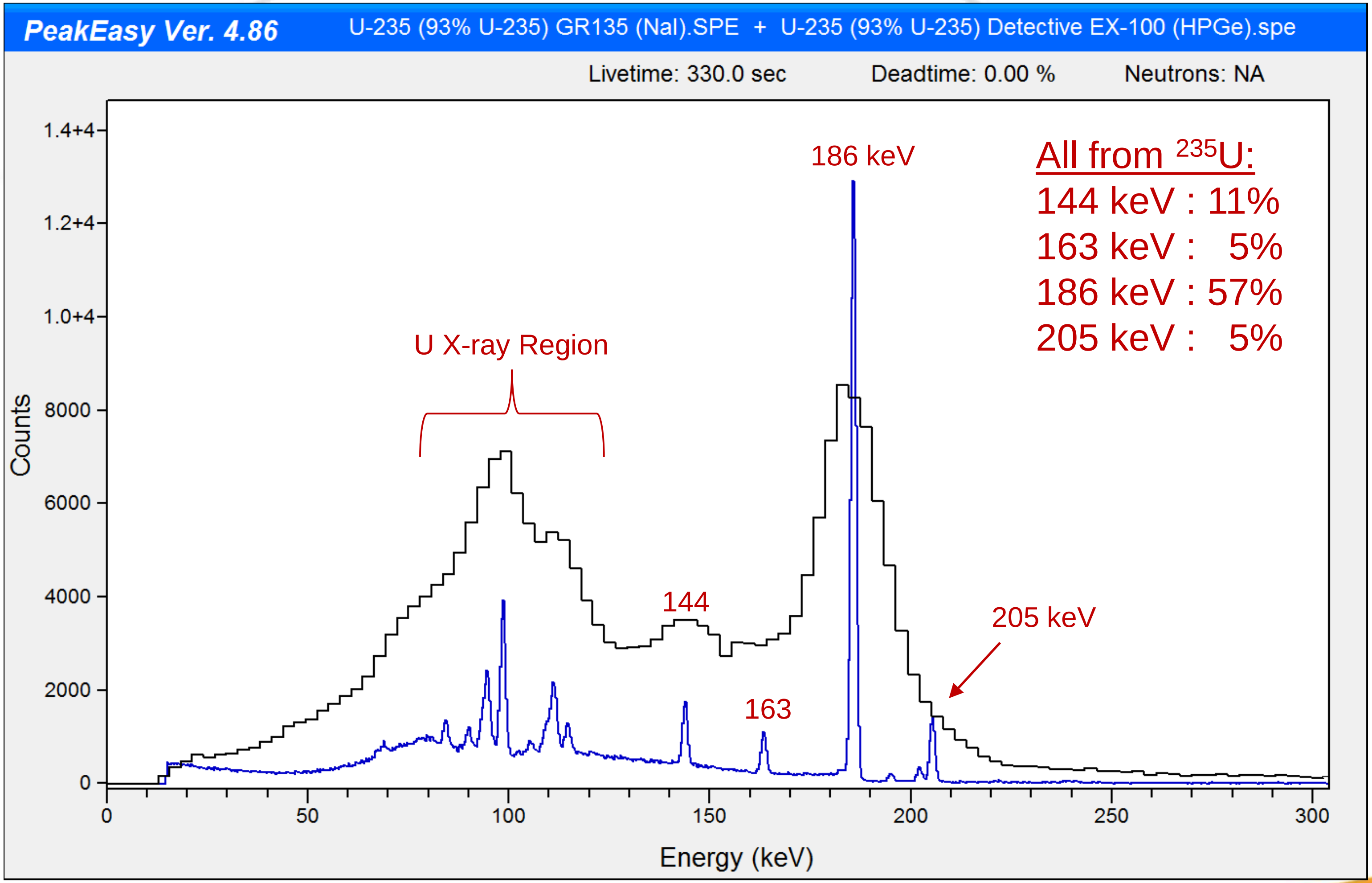
HEU (Low-Energy, Linear Scale)



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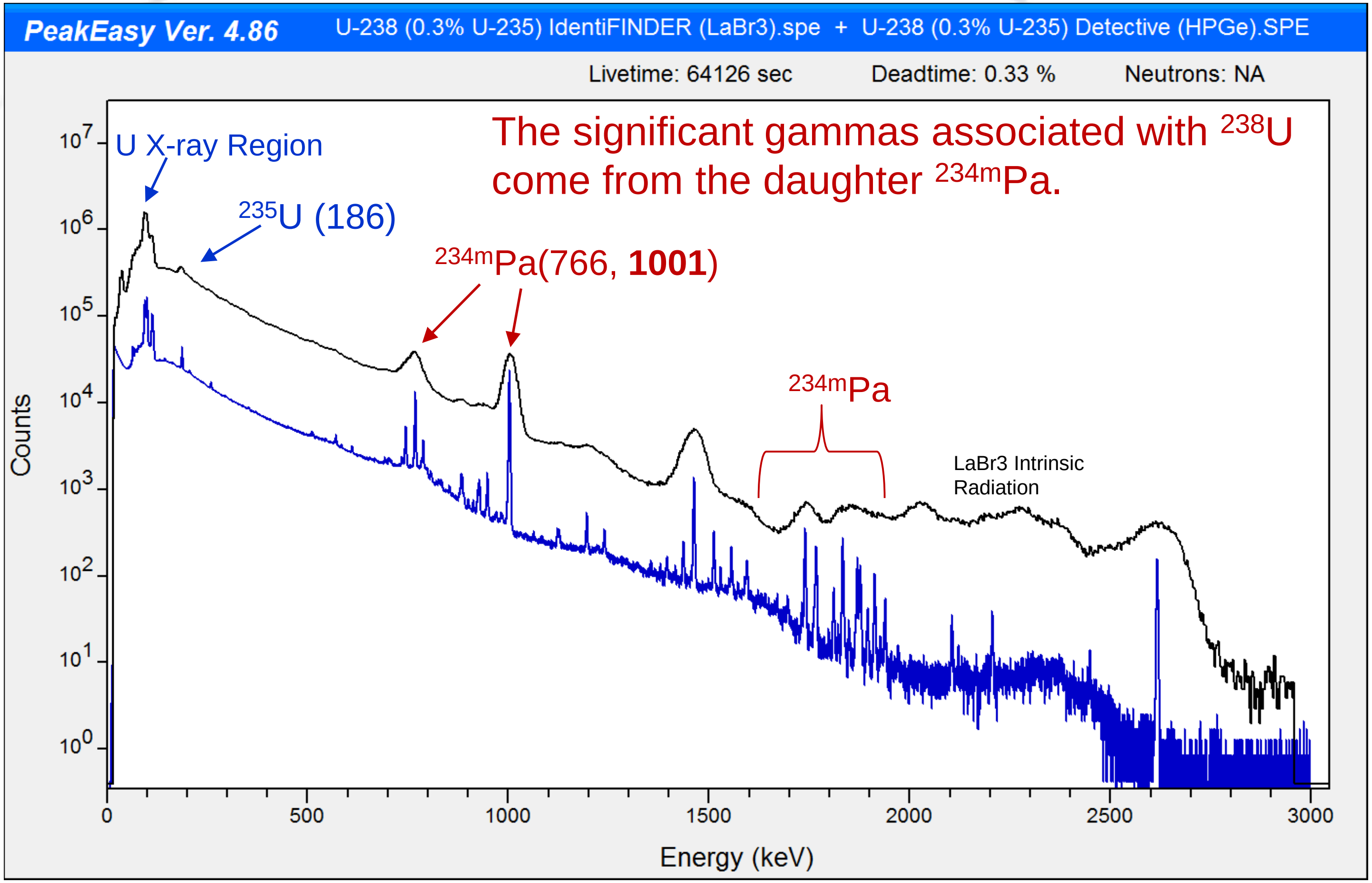
Depleted Uranium (DU)

Note: DU is not SNM by definition.



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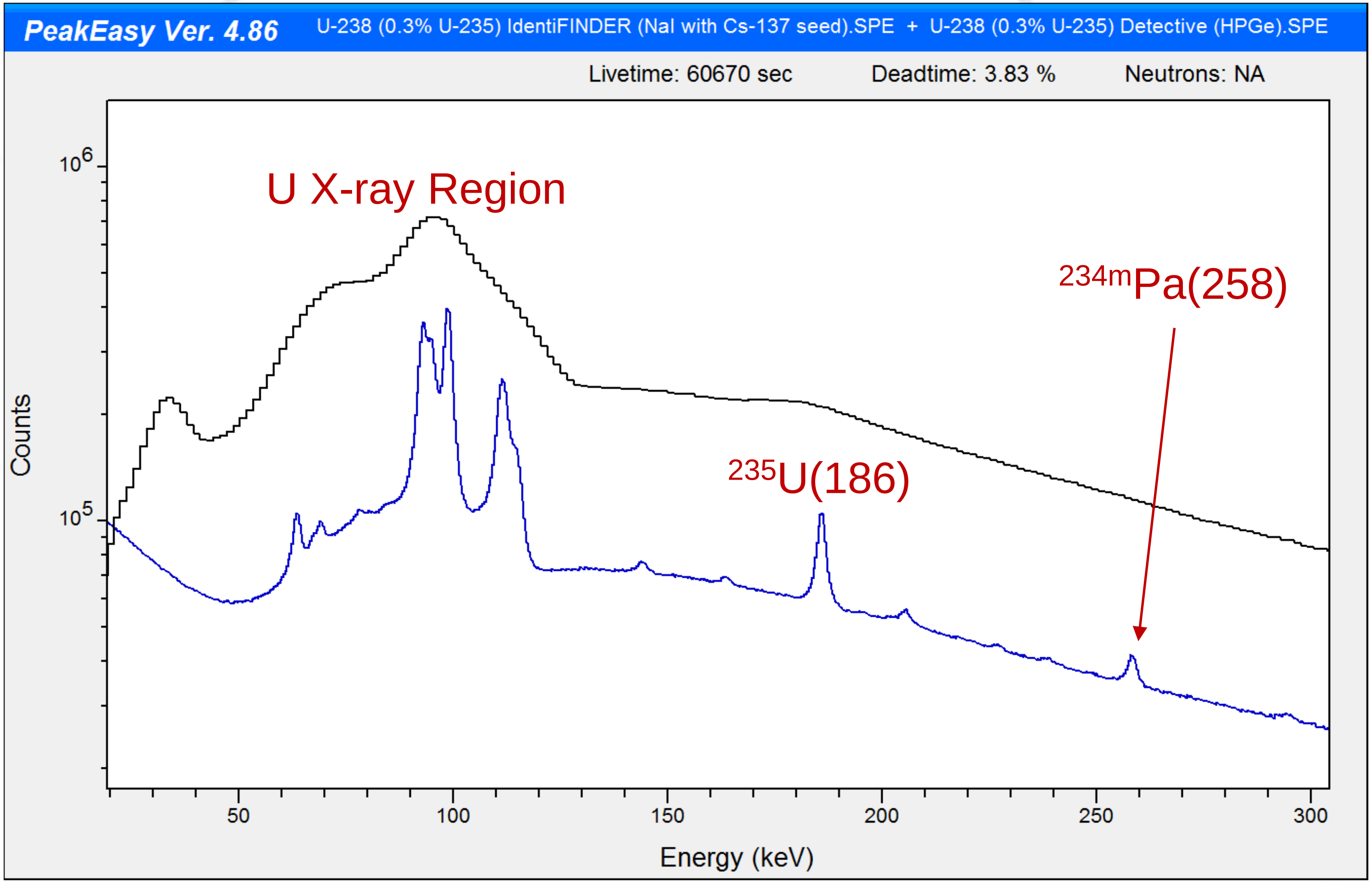
DU (Low–Energy, Log Scale)



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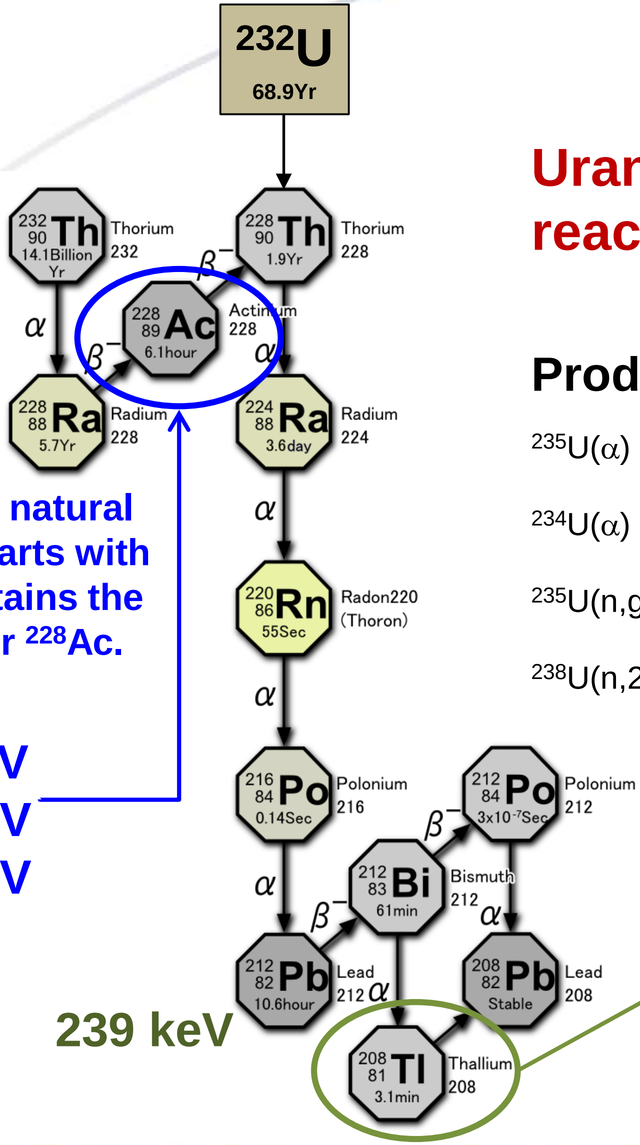
^{232}U in Recycled U

Los Alamos NATIONAL LABORATORY EST. 1943



Uranium that has been through a reactor (recycled) contains ^{232}U .

Thorium in the natural background starts with ^{232}Th, and contains the decay daughter ^{228}Ac.

911 keV
969 keV
338 keV

Production of ^{232}U in a Reactor:

$^{235}U(\alpha) \rightarrow {}^{231}Th(\beta-) \rightarrow {}^{231}Pa(n,\gamma) \rightarrow {}^{232}Pa(\beta-) \rightarrow {}^{232}U$

$^{234}U(\alpha) \rightarrow {}^{230}Th(n,\gamma) \rightarrow {}^{231}Th(\beta-) \rightarrow {}^{231}Pa(n,\gamma) \rightarrow {}^{232}Pa(\beta-) \rightarrow {}^{232}U$

$^{235}U(n,g) \rightarrow {}^{236}U(n,\gamma) \rightarrow {}^{237}U(\beta-) \rightarrow {}^{237}Np(n,2n)\ {}^{236m}Np(\beta-) \rightarrow {}^{236}Pu(\alpha) \rightarrow {}^{232}U$

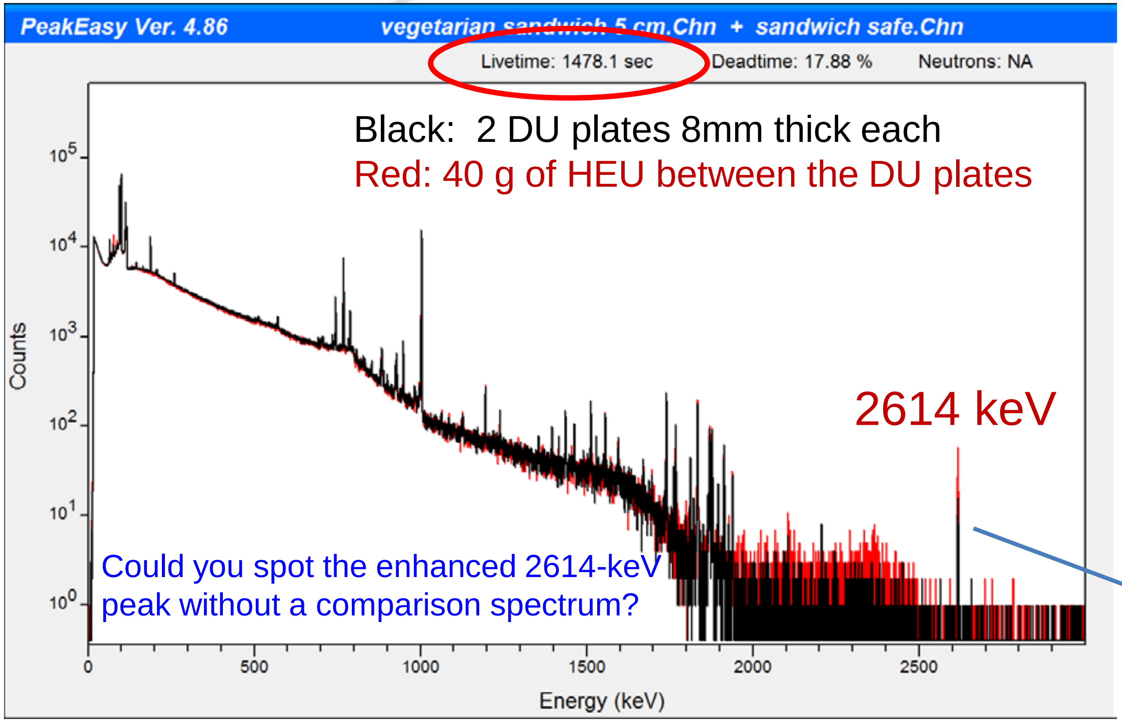
$^{238}U(n,2n) \rightarrow {}^{237}U(\beta-) \rightarrow {}^{237}Np(n,2n) \rightarrow {}^{236m}Np(\beta-) \rightarrow {}^{236}Pu(\alpha) \rightarrow {}^{232}U$

2614 keV
861 keV
583 keV

*Peurrung, A.J., "Predicting ^{232}U Content in Uranium", PNNL Document 12075

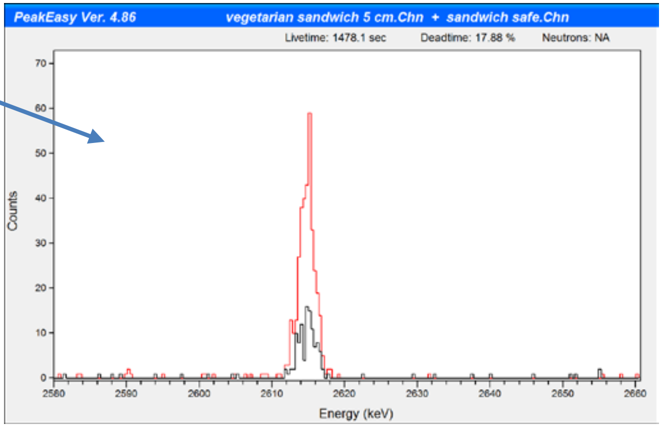
NNSA National Nuclear Security Administration

Shielded Recycled HEU (HPGe)



In the black spectrum we only have counts at 2614 from the detector intrinsic radiation and the natural background.

The red spectrum has additional counts at 2614 keV from ^{232}U.

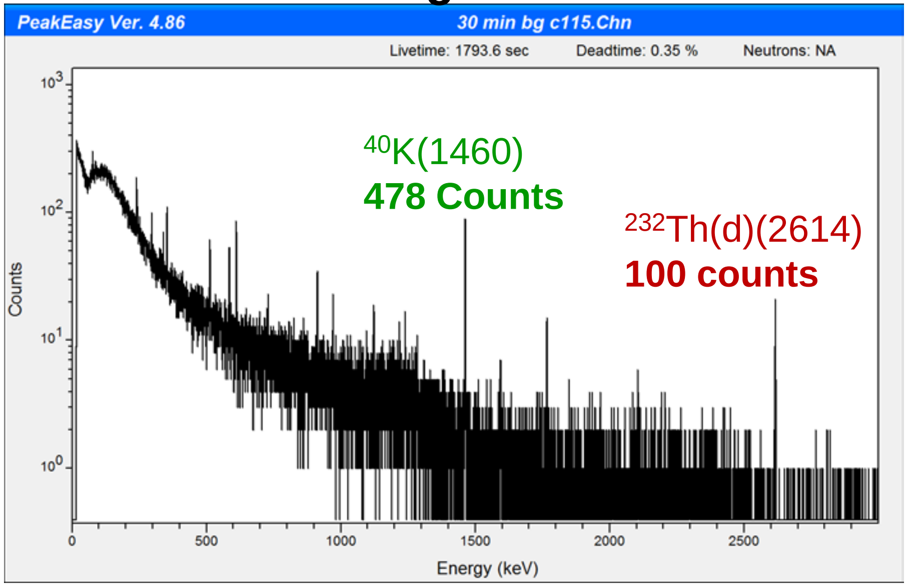


The superior peak-to-Compton ratio of the HPGe detector used to take these spectra further emphasizes the <u>enhanced 2614-keV peak</u>.

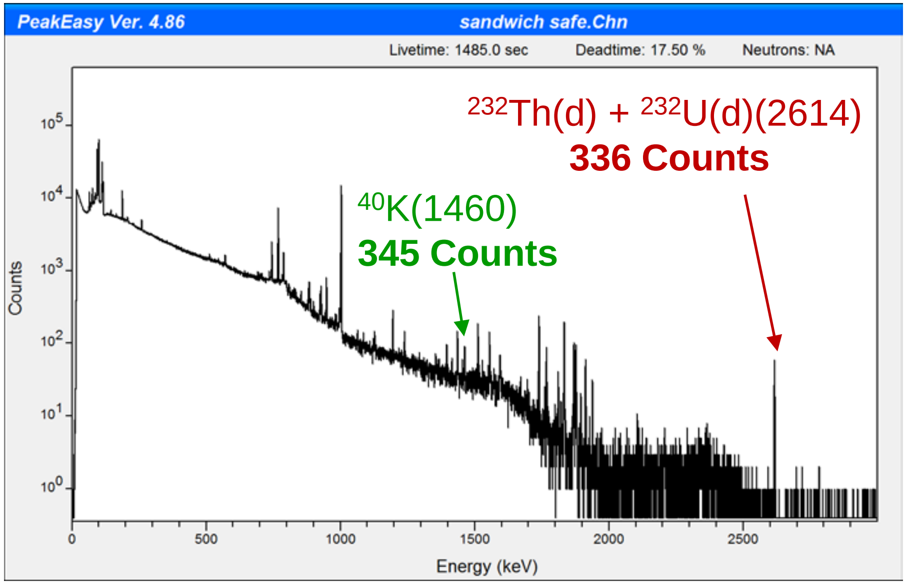
The Enhanced 2614-keV Peak

If the 2614-keV peak has the more counts than the 1460-keV peak from ^{40}K in the absence of a thorium source, then suspect ^{232}U.

Background



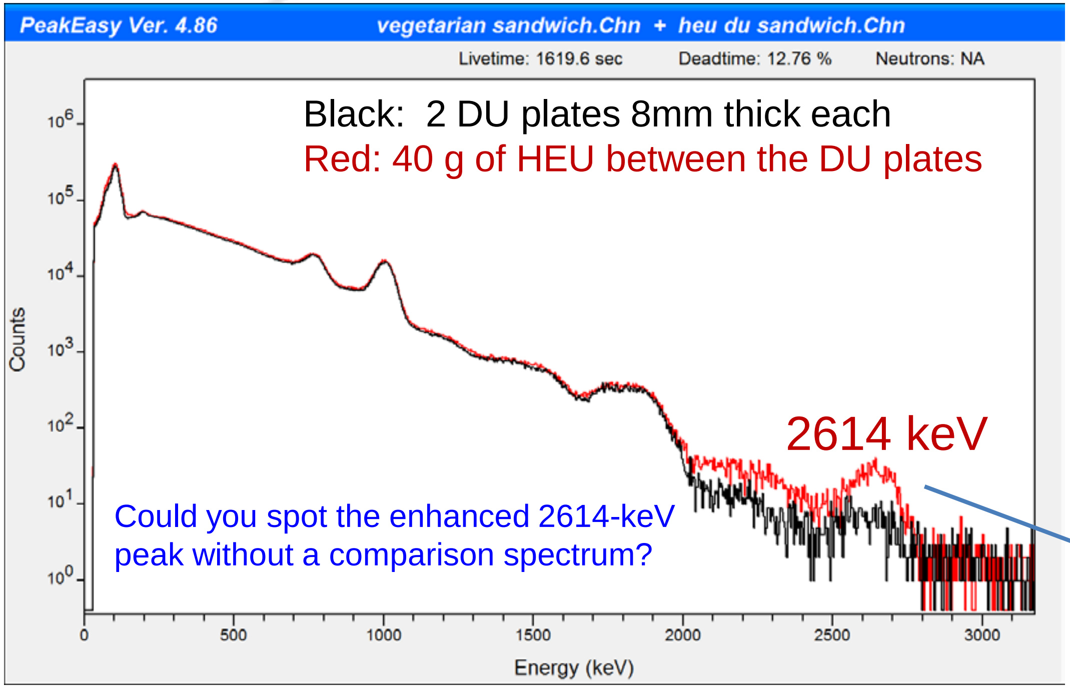
HEU behind DU



In the background, the ratio of 2614:1460 counts is < 1:4

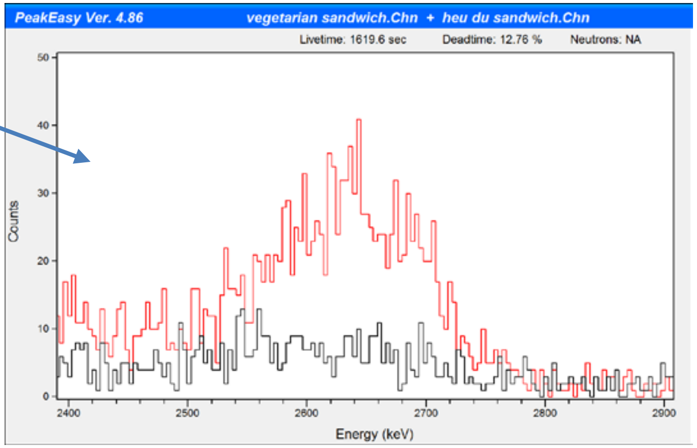
In the shielded (recycled) HEU case, the ratio of 2614:1460 counts is ~ 1:1

Shielded Recycled HEU (NaI)



In the black spectrum we only have counts at 2614 from the detector intrinsic radiation and the natural background.

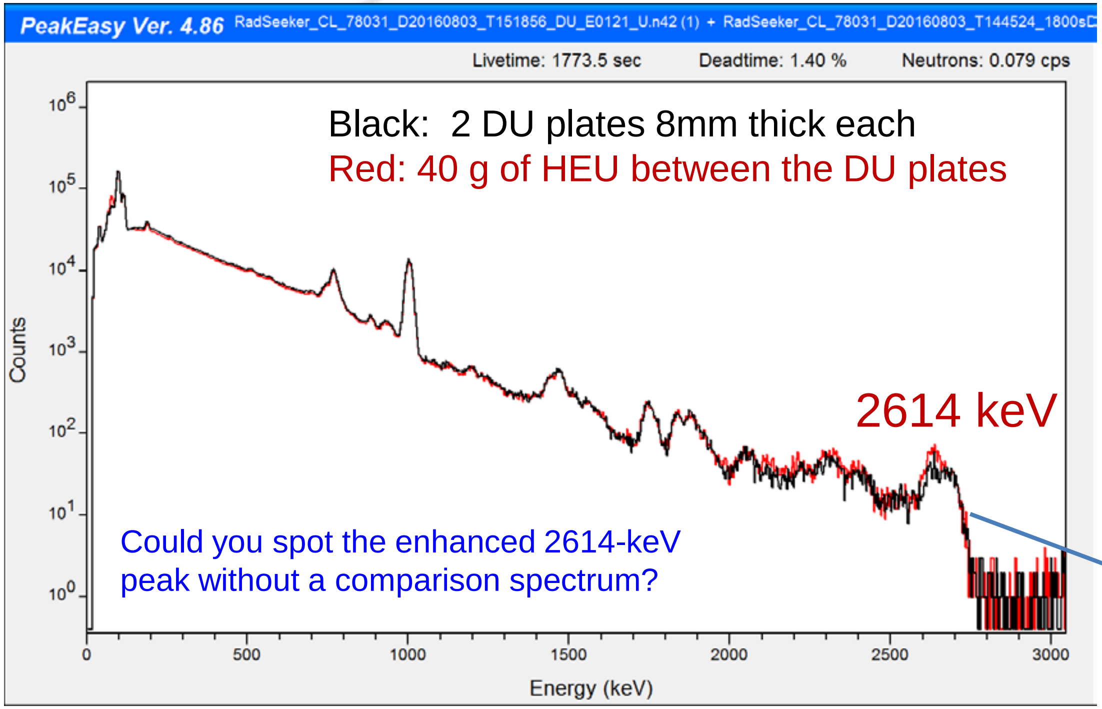
The red spectrum has additional counts at 2614 keV from ^{232}U.



Without intrinsic radiation such as with the LaBr3 detector, the NaI spectra, clearly show the enhanced 2614-keV peak.

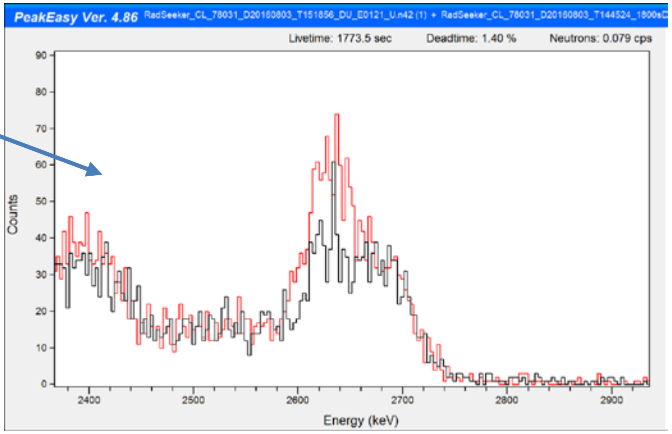
Operated by Los Alamos National Security, LLC for the U.S. Department of Energy's NNSA

Shielded Recycled HEU (LaBr3)



In the black spectrum we only have counts at 2614 from the detector intrinsic radiation and the natural background.

The red spectrum has additional counts at 2614 keV from ^{232}U.



The intrinsic radiation from the LaBr3 detector crystal makes the presence of an <u>enhanced 2614-keV peak</u> more subtle than for other detector types.

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Plutonium Isotopic Information

Example of Typical Isotopic Composition for Weapon-Grade Plutonium (WGPu) and Reactor-Grade Plutonium (RGPu)

	% ^{238}Pu	**% ^{239}Pu**	**% ^{240}Pu**	% ^{241}Pu	% ^{242}Pu	% ^{241}Am
WGPu	0.01	**93.6**	**6.3**	0.07	0.04	0.3
RGPu	1.0	**65.8**	**26.7**	2.1	4.4	6.3

- Burnup(BU) is a measure of how much energy has been extracted from the primary nuclear fuel. For us, it basically means:
  - How long has the Pu been in the reactor?
  - Low BU = WGPu
  - High BU = RGPu

  (compare this to the lingo for LEU and HEU)

- ^{236}Pu production in a reactor:
  - ^{235}U(n,g) → ^{236}U(n,γ) → ^{237}U(β-) → ^{237}Np(n,2n) ^{236m}Np(β-) → ^{236}Pu(α) → ^{232}U
  - High BU : more ^{236}Pu

Nuclide Transitions in a Reactor

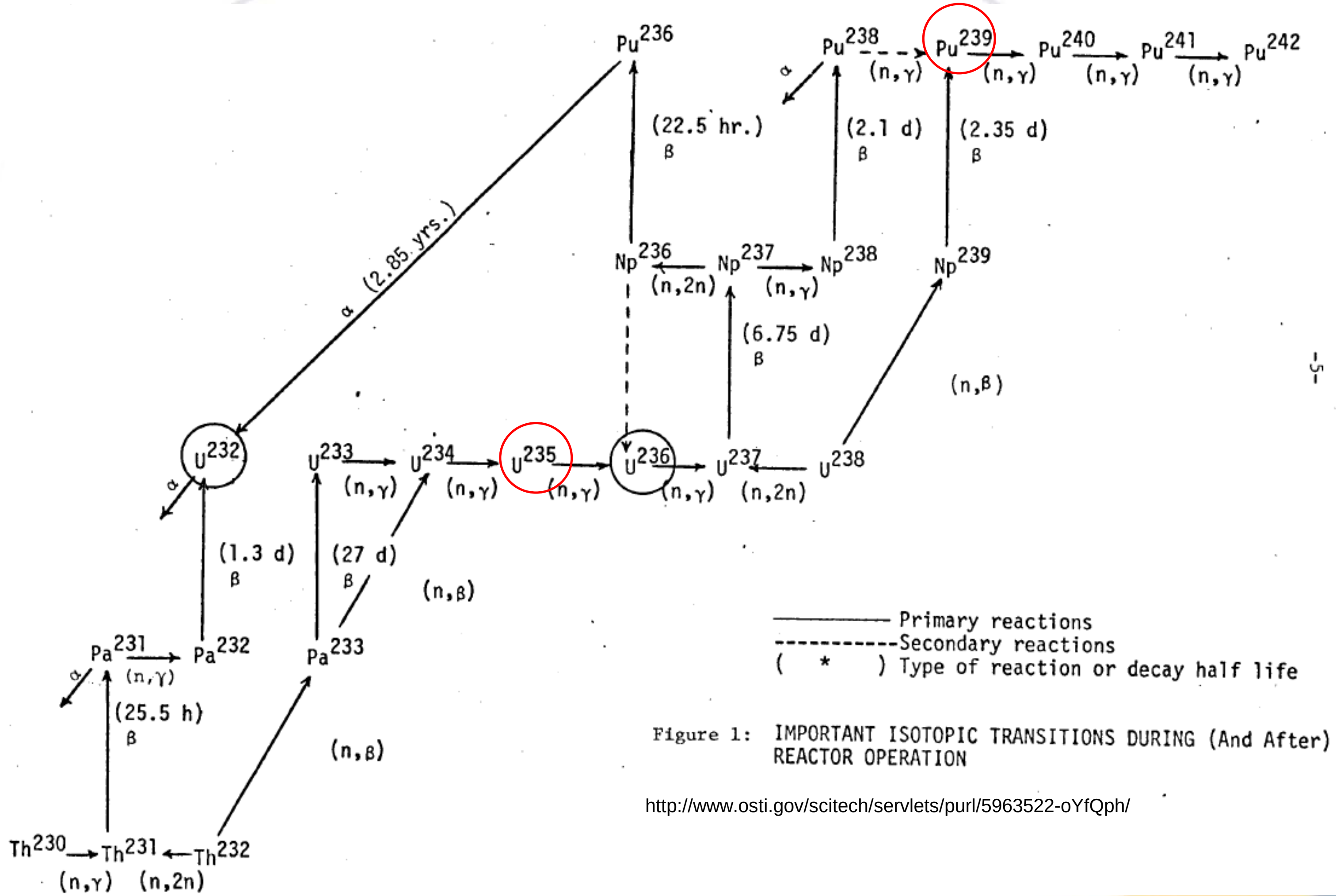
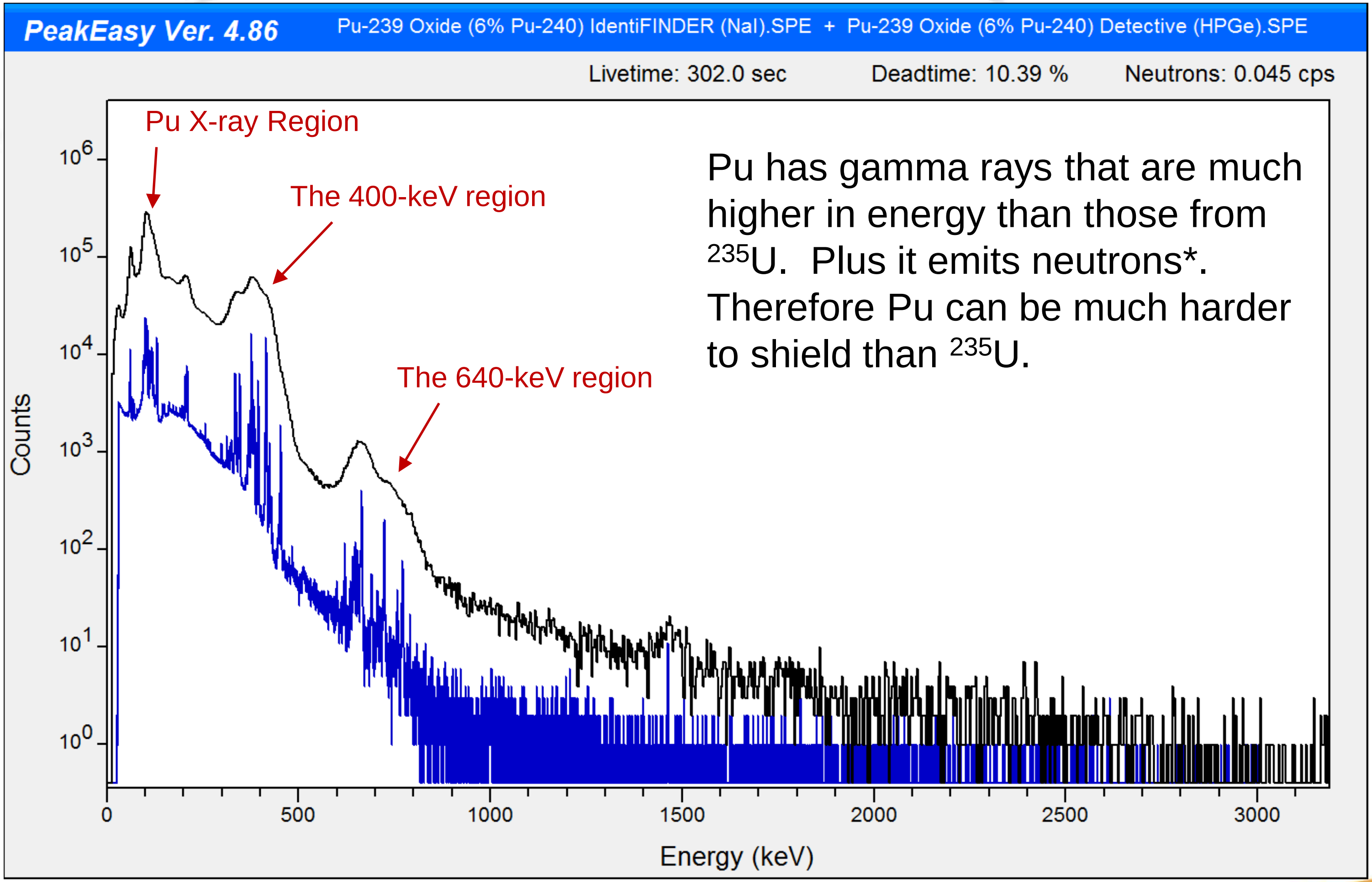


Figure 1: IMPORTANT ISOTOPIC TRANSITIONS DURING (And After) REACTOR OPERATION

http://www.osti.gov/scitech/servlets/purl/5963522-oYfQph/

Plutonium (NaI vs. HPGe)



Pu has gamma rays that are much higher in energy than those from ^{235}U. Plus it emits neutrons*. Therefore Pu can be much harder to shield than ^{235}U.

* The mass for this Pu item is only 10g so the neutron signal is weak.

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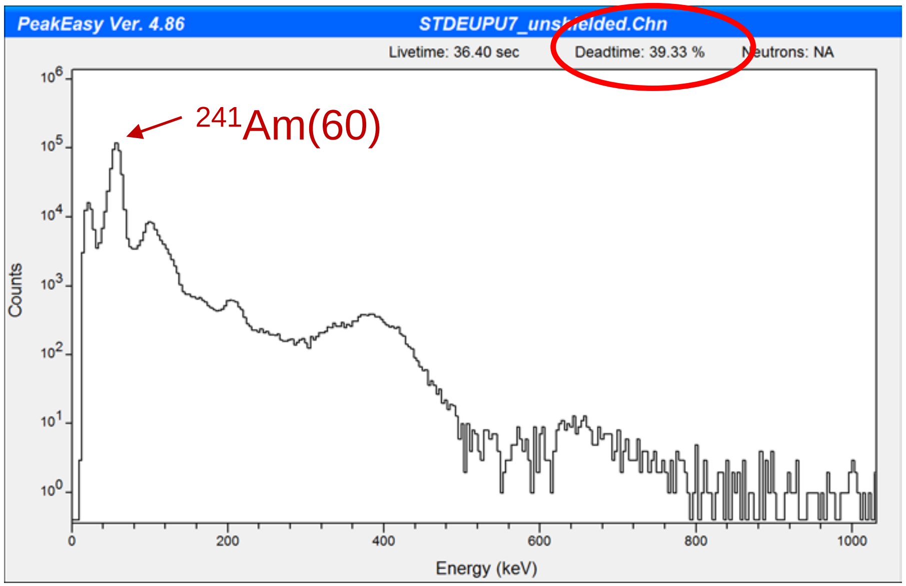
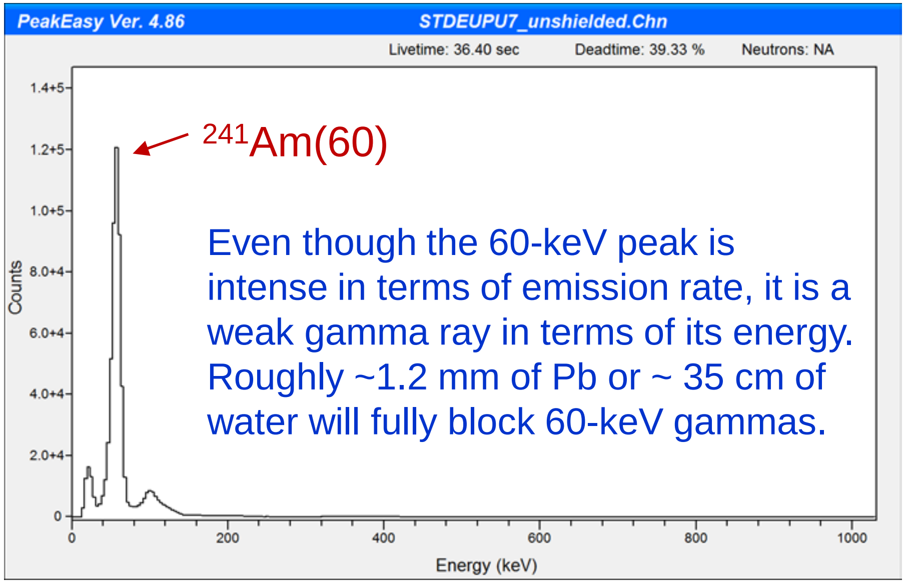
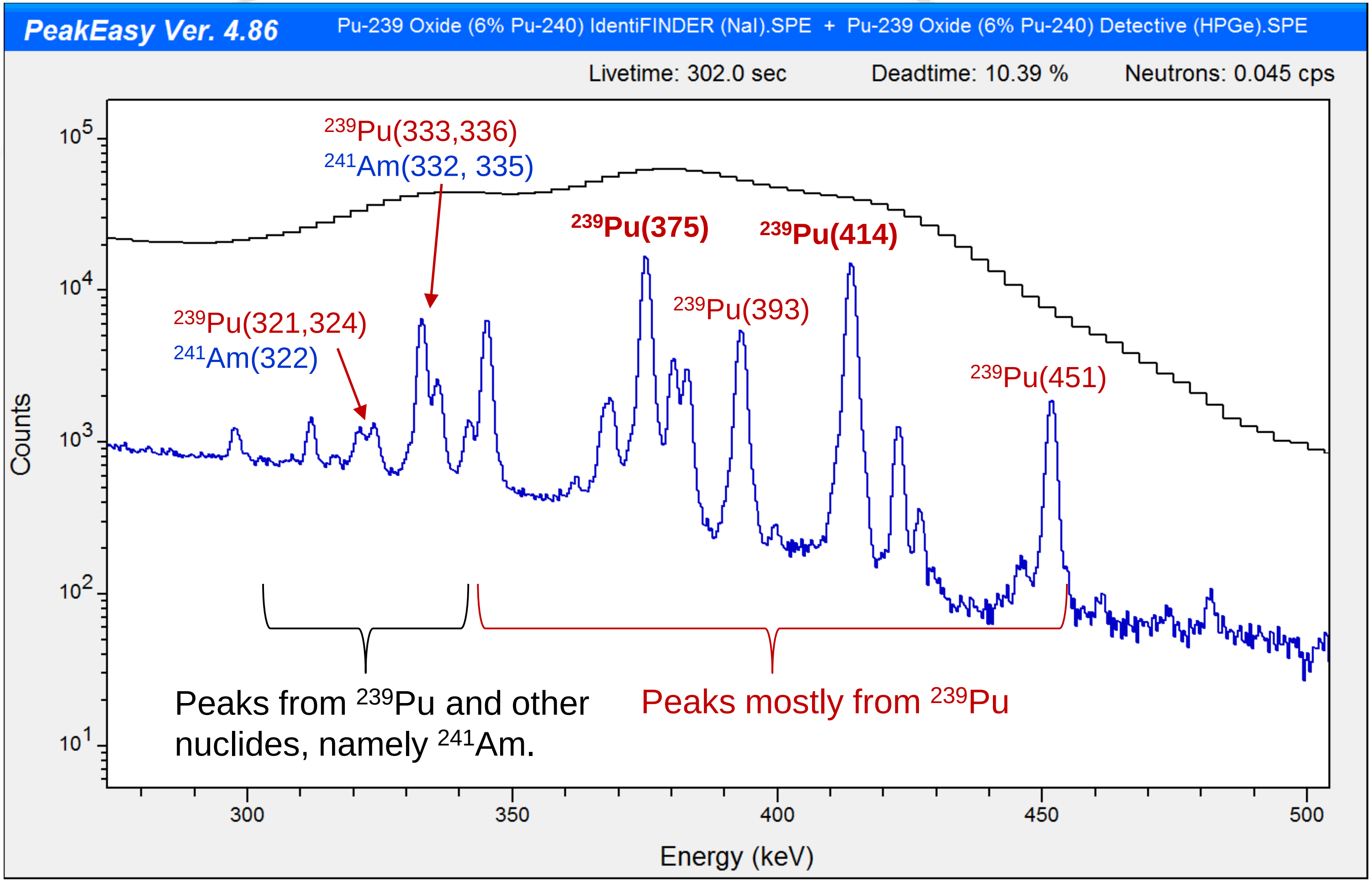
The 60-keV Peak from ^{241}Am

^{241}Am comes from the decay of ^{241}Pu. Normally for unshielded or lightly shielded plutonium, **^{241}Am will dominate** your spectrum and the gamma dose that one would receive.





Plutonium 400-keV Region



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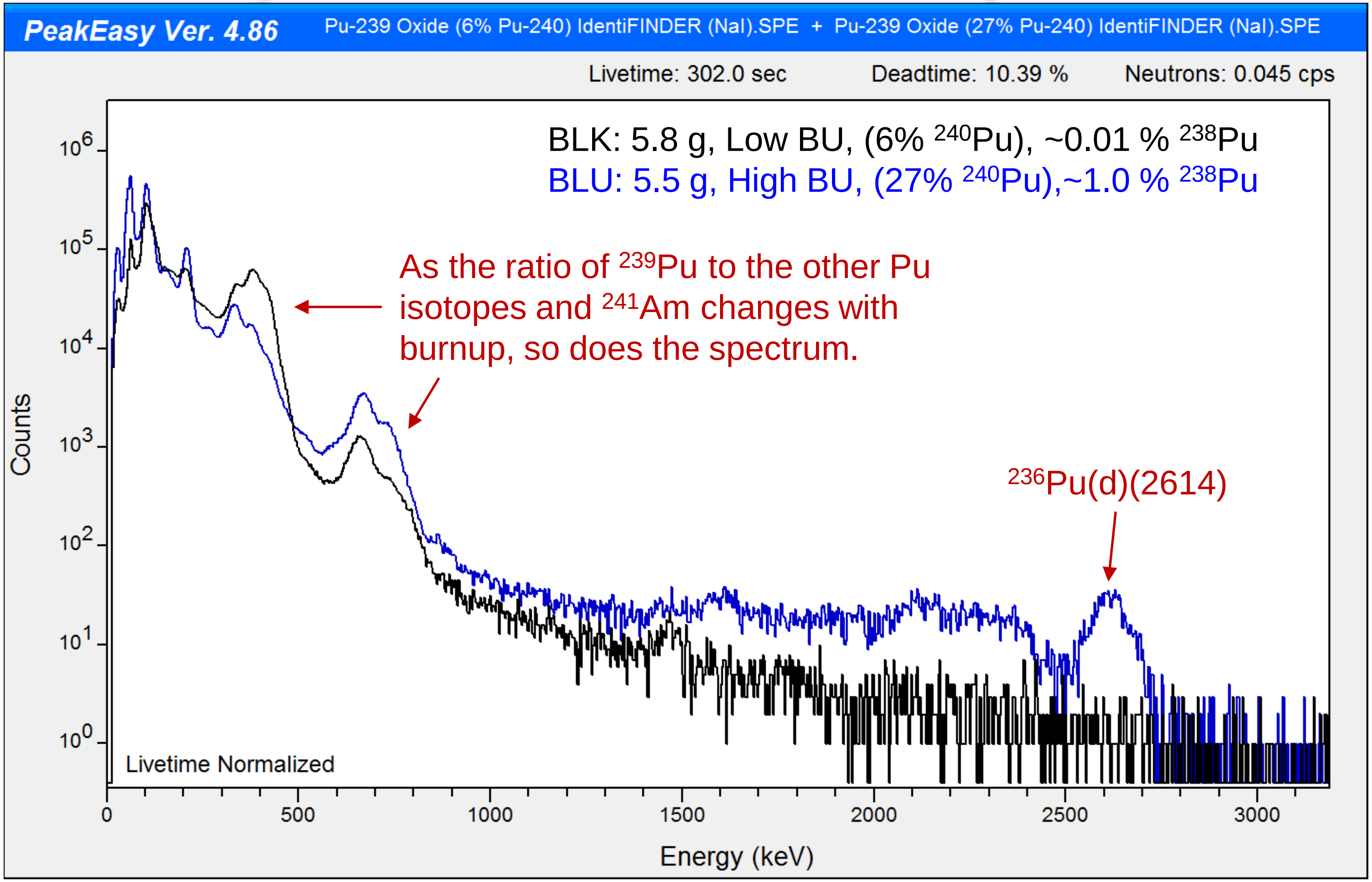
Spectral Variations from Burnup



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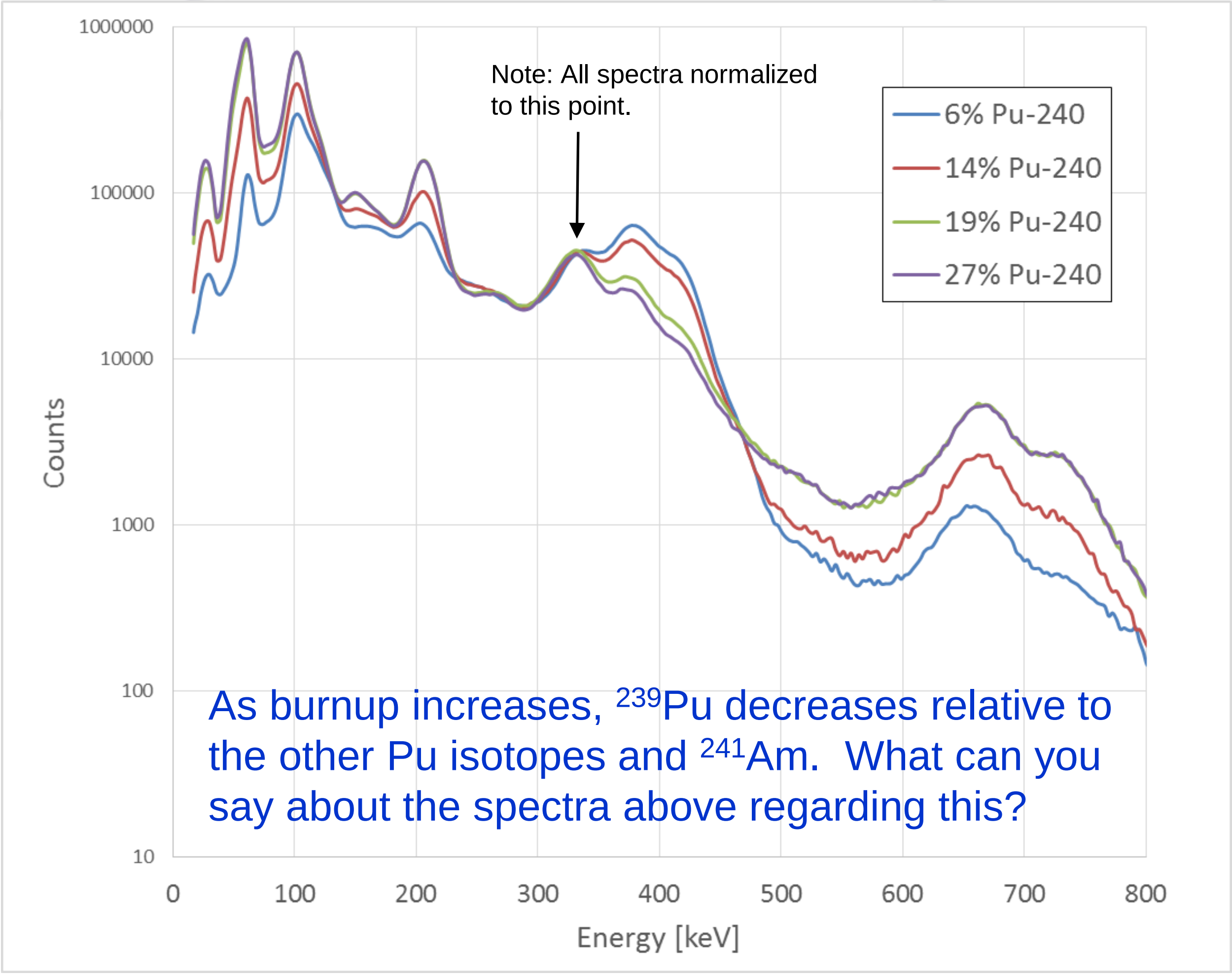
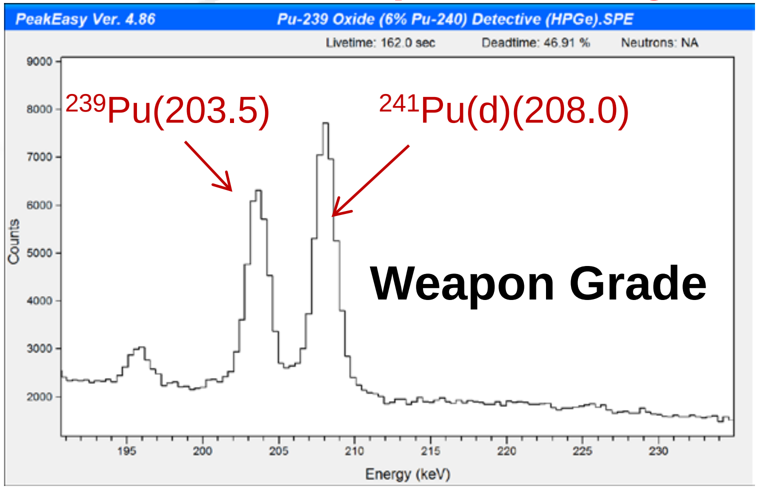
Burnup Comparison (NaI)

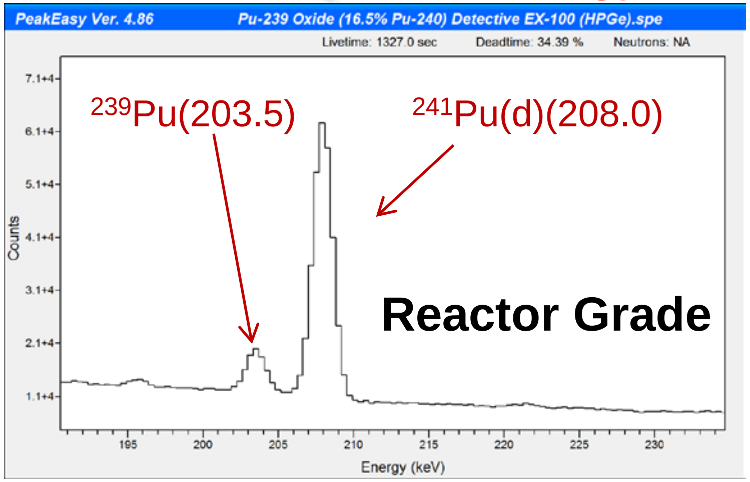


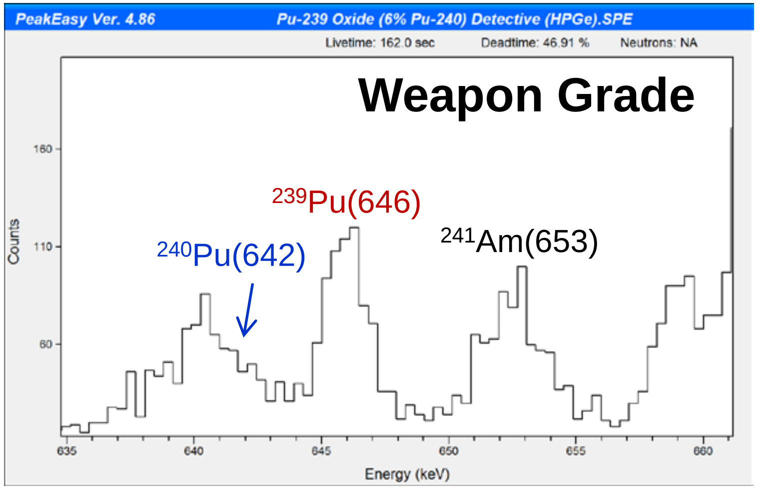
As burnup increases, ^{239}Pu decreases relative to the other Pu isotopes and ^{241}Am. What can you say about the spectra above regarding this?

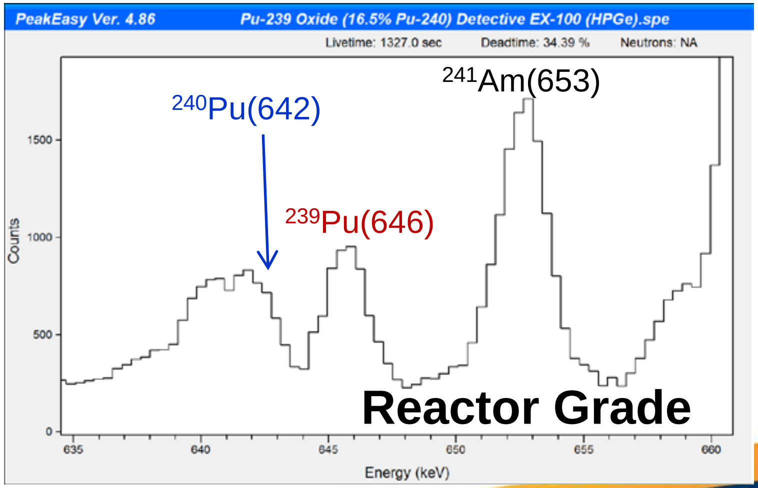
Estimating Burnup with HPGe

Note: The peaks being compared here are close in energy!

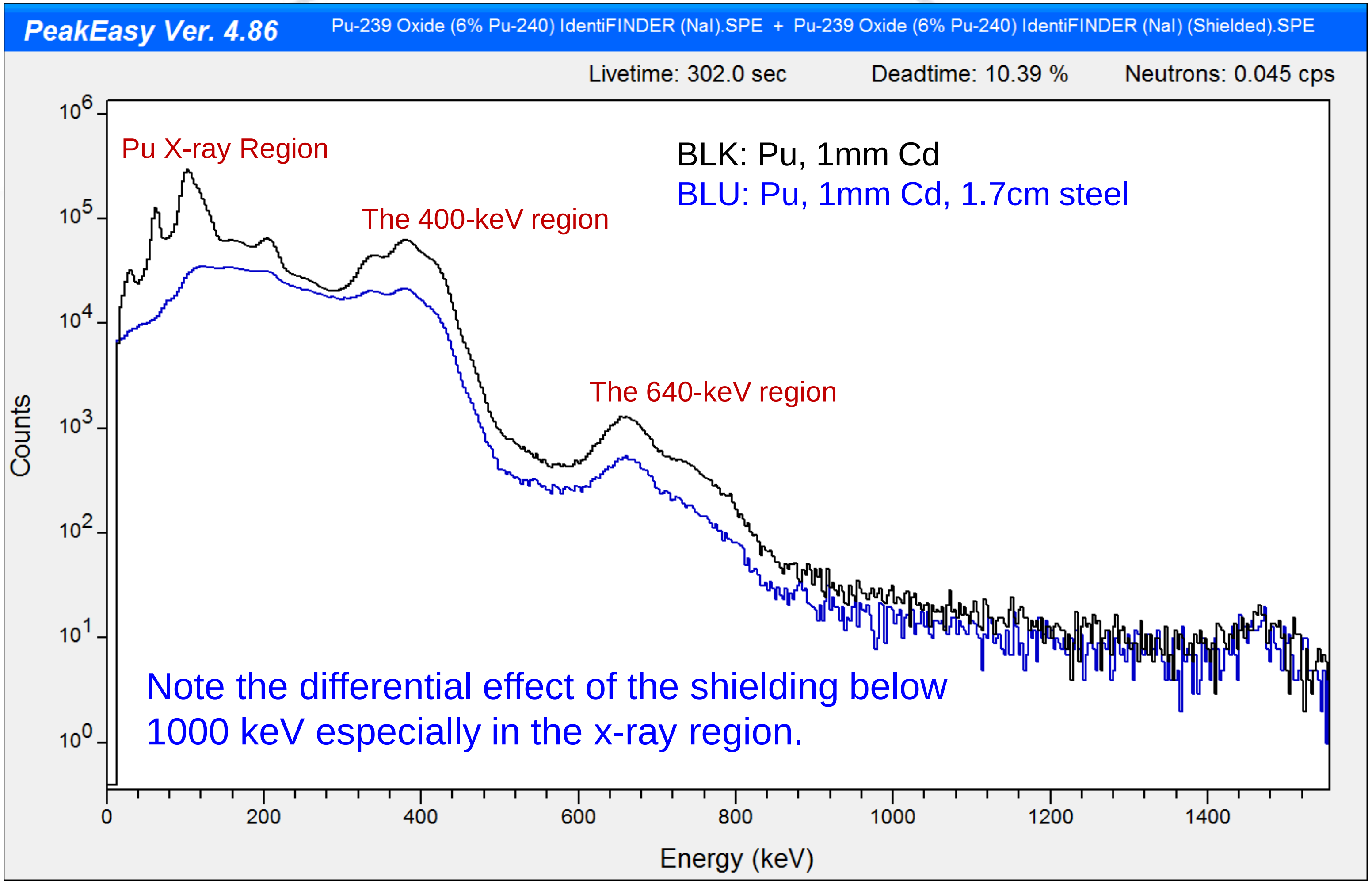








Shielded Plutonium



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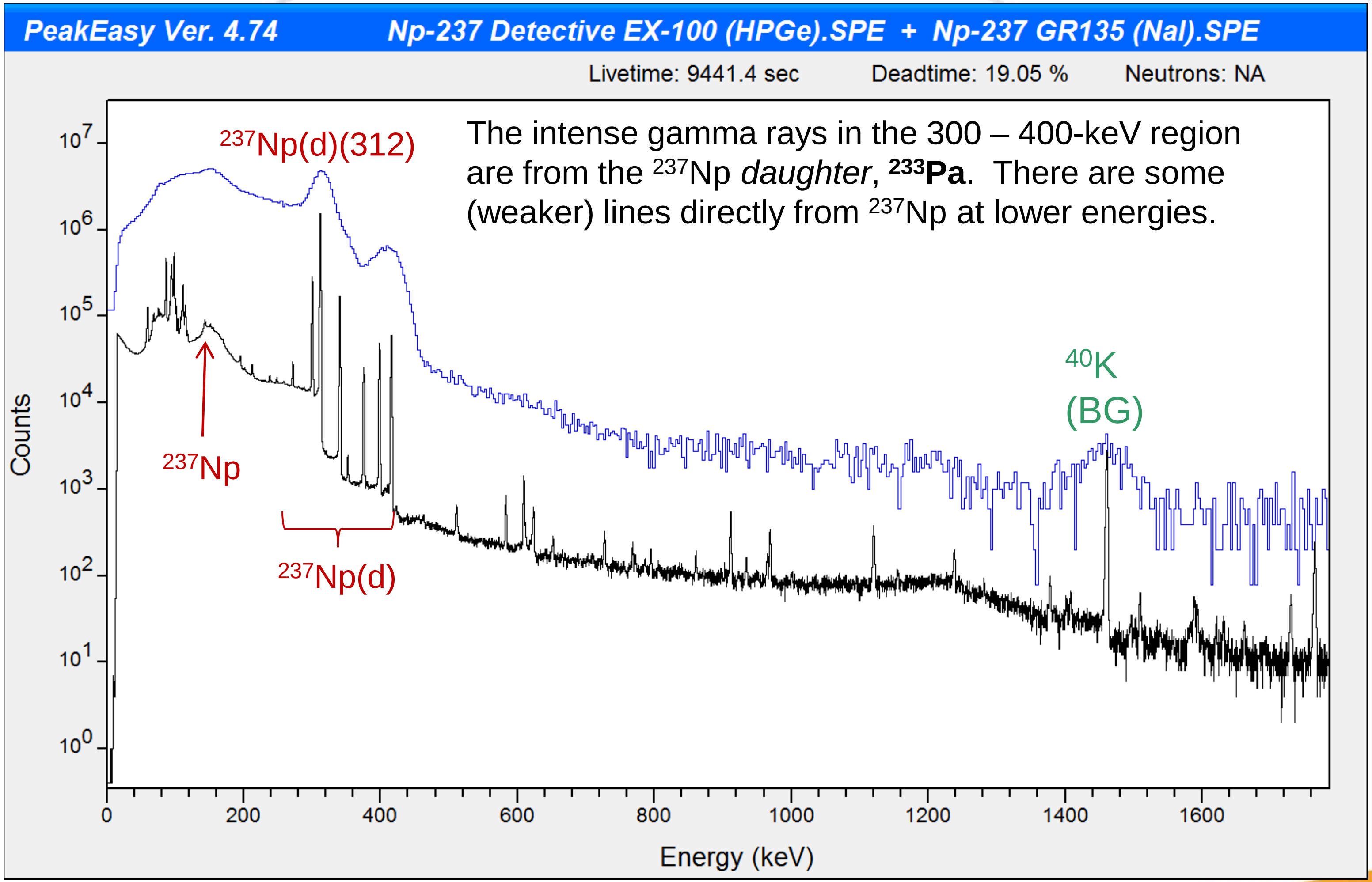
^{237}Np Spectrum



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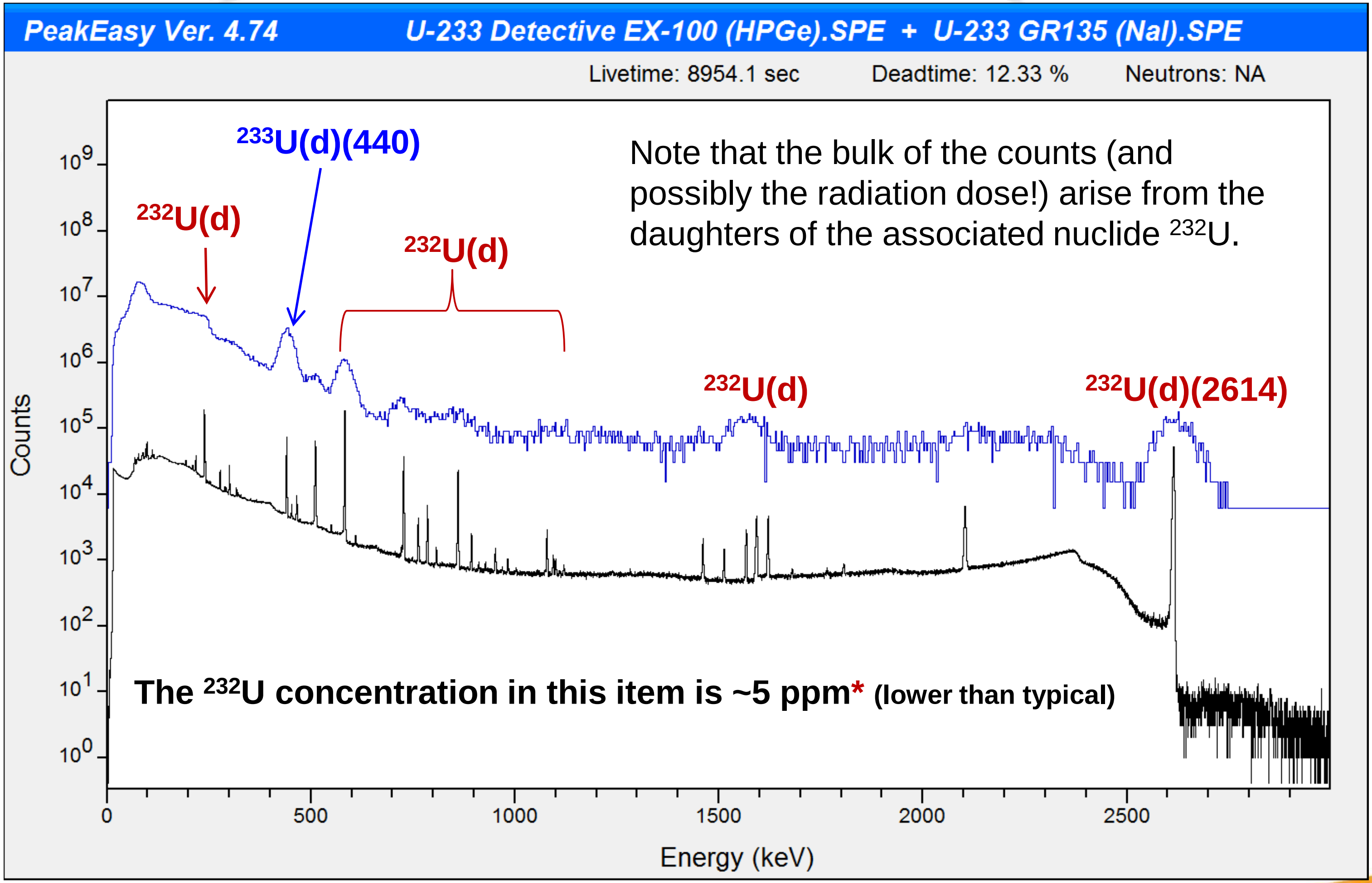
^{233}U Spectrum



* age assumed to be 40 years

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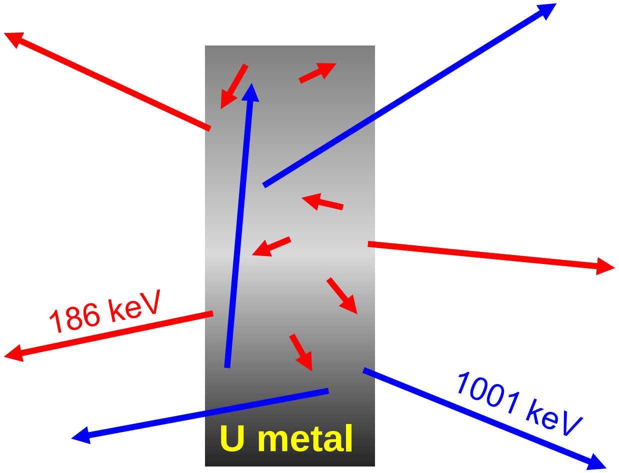
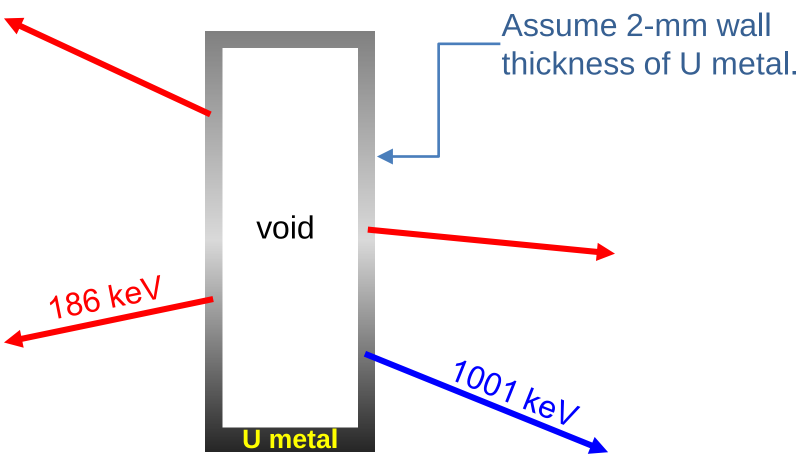
Self-Attenuation

SNM metal is high Z, high ρ material. It is very good at attenuating gamma rays, even those from itself.

Red Arrows: 186 keV
Blue Arrows: 1001 keV

~ 2.5 mm of U metal totally blocks 186 keV
~ 50 mm of U metal totally blocks 1001 keV





At our detector we see the *same amount of 186-keV* gammas for both cases above. But we see less of the 1001-keV gammas <u>in a relative sense</u> from the thin-walled box than the solid block.

Closing Thoughts

- SNM materials have definite spectral signatures that should be readily recognizable to analysts in both bare and shielded configurations.
- One can estimate burnup of plutonium using certain pairs of peaks that are a few keV apart.
- In most cases, one cannot reliably estimate uranium enrichment in an analogous way to the estimation of plutonium burnup.
- The origin of the most intense peaks from some SNM items may be indirect and from ‘associated nuclides’
- Indirect SNM signatures sometimes have commonalities with the natural gamma-ray background.