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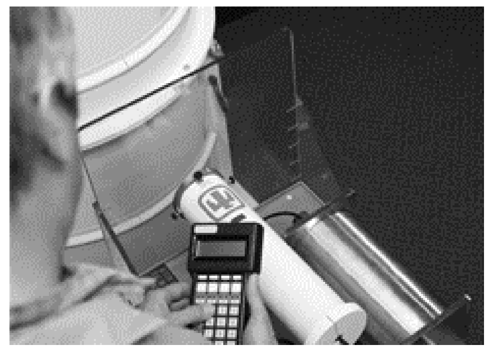

Relative Efficiency Curves Demystified

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Overview

- Relative efficiency curves are the fundamental basis of uranium and plutonium isotopic software programs such as Los Alamos National Laboratory (LANL) Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) and Lawrence Livermore National Laboratory (LLNL) Multi-Group Analysis (MGA).

- Relative efficiency curves are used to determine nuclide activity ratios or nuclide mass ratios (not absolute activity or mass).

- Benefit: Relative efficiency curves require no measurement of calibration sources and "self-correct" for geometry and attenuation (shield attenuation and self attenuation).

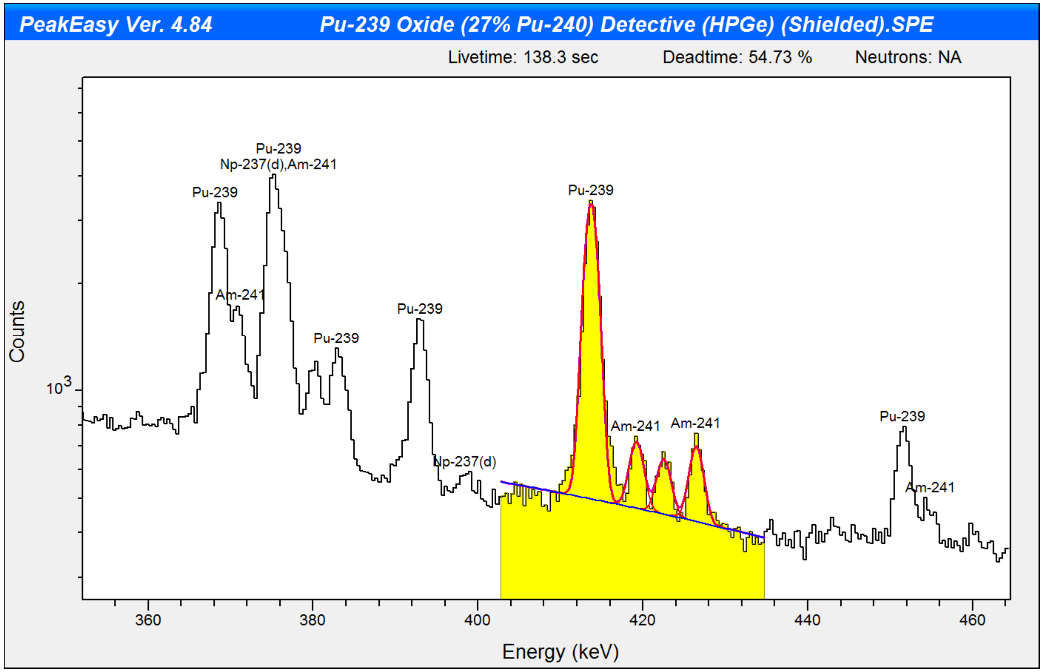
- This presentation will focus on relative efficiency curves as applied to the determination of uranium isotopics.

Basics

- Closely spaced full-energy peak (FEP) pairs
  - When full-energy peaks from two different nuclides are very close in energy the relative detection efficiency is essentially the same.
  - As such, the relative activity of the nuclides can be well estimated by taking the ratio of the net full-energy peak counts divided by the yield of each nuclide.

Example A: High burnup plutonium

- Using closely spaced full-energy peak pairs, estimate the relative Am-241 to Pu-239 activity.

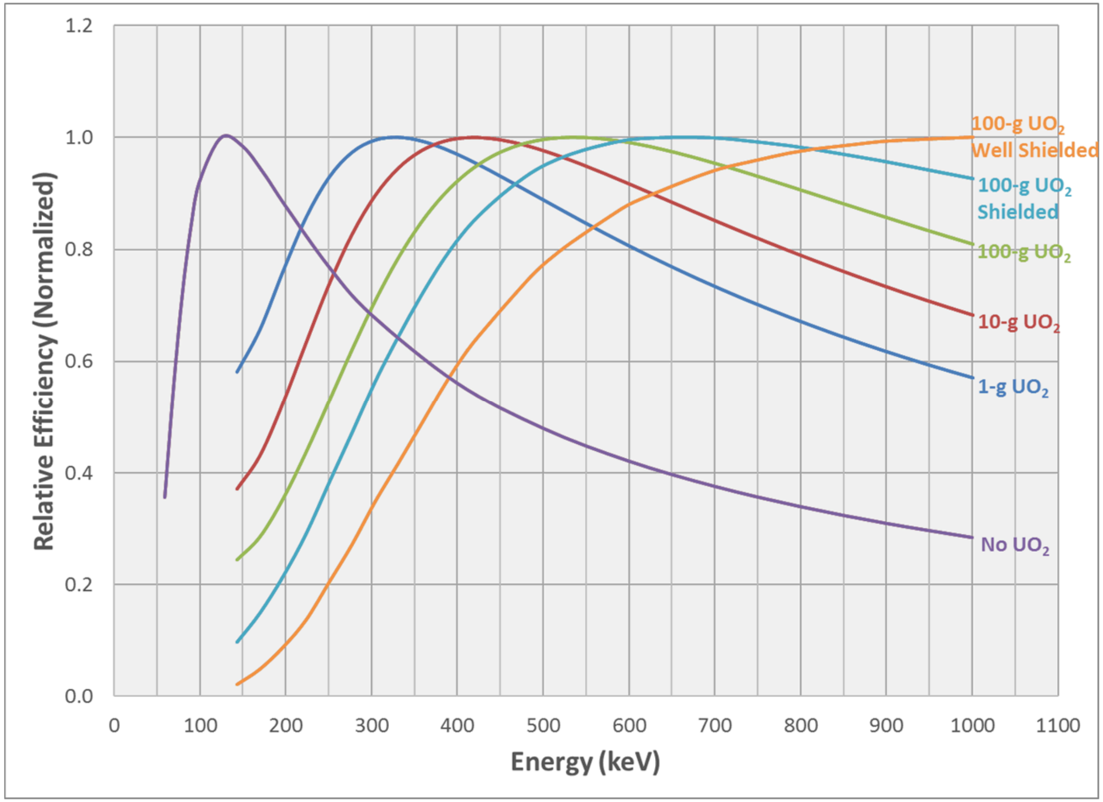


Nuclide	Energy (keV)	Yield gps/dps	Net_Area counts	Counts / Yield
Am-241	419.3	2.87E-07	1410	4.91E+09
Pu-239	413.7	1.47E-05	16529	1.13E+09

- Relative Am-241 to Pu-239 activity = (4.91E+09 / 1.13E+09) = 4.4

Relative efficiency curve shapes

- The shape of the relative efficiency curve is based on the detector efficiency, item geometry, shield attenuation, and self attenuation.
- Accordingly, changes in detector efficiency, item geometry, shield attenuation, and self attenuation are reflected by changes in the shape of the relative efficiency curve.



Generation of a relative efficiency curve

- To generate an effective relative efficiency curve, you must have a nuclide with full-energy peaks than span the energy range of interest or multiple nuclides with full-energy peaks with sufficient overlapping or nearly overlapping energy ranges for the energy range of interest.

- In addition, the isotopic composition throughout the sample must be the same for a relative efficiency curve to be valid.

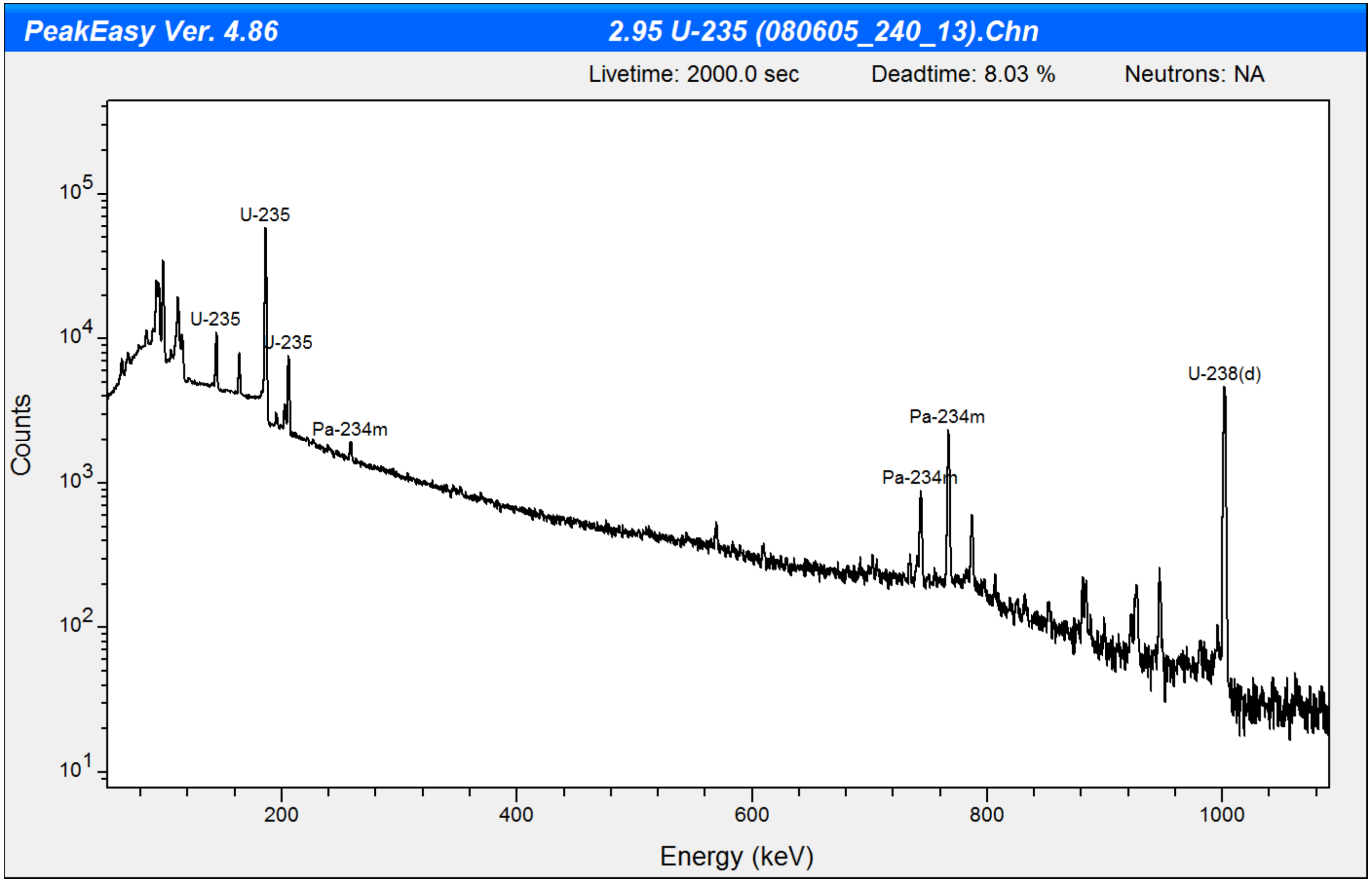
Uranium relative efficiency analysis

- Primary uranium gamma emissions between 120 and 1010 keV.
  - U-235: 143.8, 163.3, 185.7, and 205.3 keV
  - U-238: 258.3, 742.8, 766.4, and 1001.0 keV
  - U-232: 238.6, 583.2, 727.3, and 860.6 keV
  - U-234: 120.9 keV

- Note: U-232 is produced during reactor irradiation and is present in uranium that has been reprocessed.

Example B

- An example of the process using an ORTEC Detective measurement of a known uranium standard is presented for illustrative purposes.



Example B

1. For a selected nuclide, divide the net full-energy peak counts by the yield for each gamma emission.

Nuclide	Energy keV	Yield Unitless	Net_Area Counts	Counts / Yield
U-235	143.76	1.10E-01	27974	2.54E+05
U-235	163.33	5.08E-02	16719	3.29E+05
U-235	185.72	5.72E-01	238763	4.17E+05
U-235	205.31	5.01E-02	23549	4.70E+05

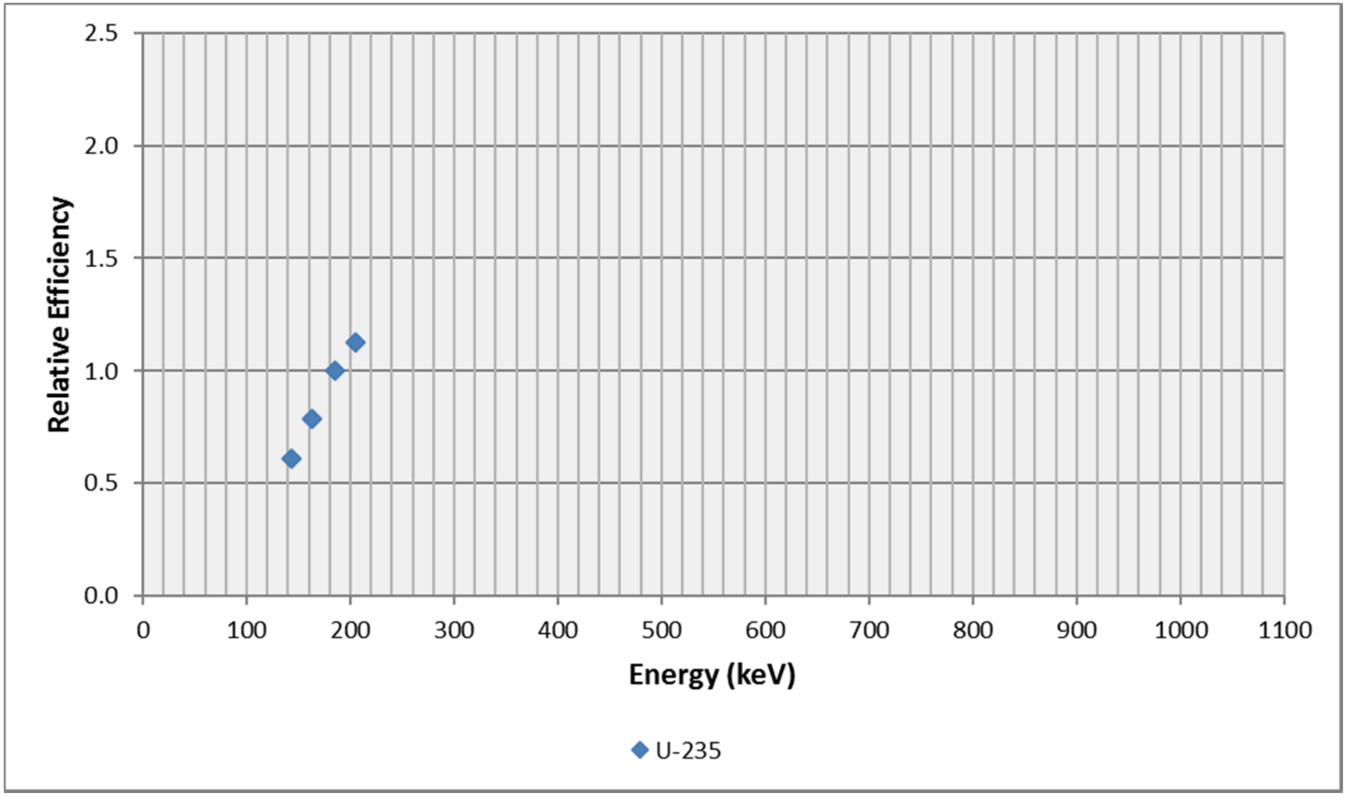
2. For the selected nuclide, normalize the results using one of the selected nuclide gamma emissions.

Nuclide	Energy keV	Yield Unitless	Net_Area Counts	Counts / Yield	Measured Rel Eff
U-235	143.76	1.10E-01	27974	2.54E+05	0.6092
U-235	163.33	5.08E-02	16719	3.29E+05	0.7884
U-235	185.72	5.72E-01	238763	4.17E+05	1.0000
U-235	205.31	5.01E-02	23549	4.70E+05	1.1261

- Results normalized to the U-235 185.7 keV emission.

Example B

3. Plot the measured relative efficiency.

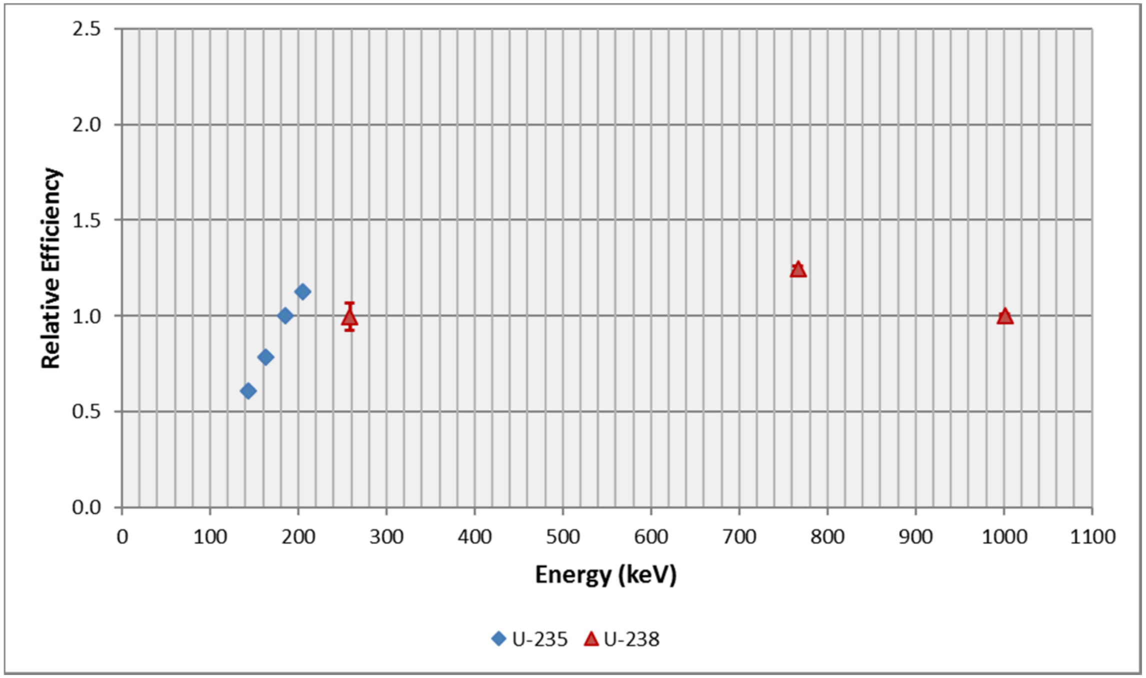


Example B

4. Perform steps 1, 2, and 3 on remaining nuclides.

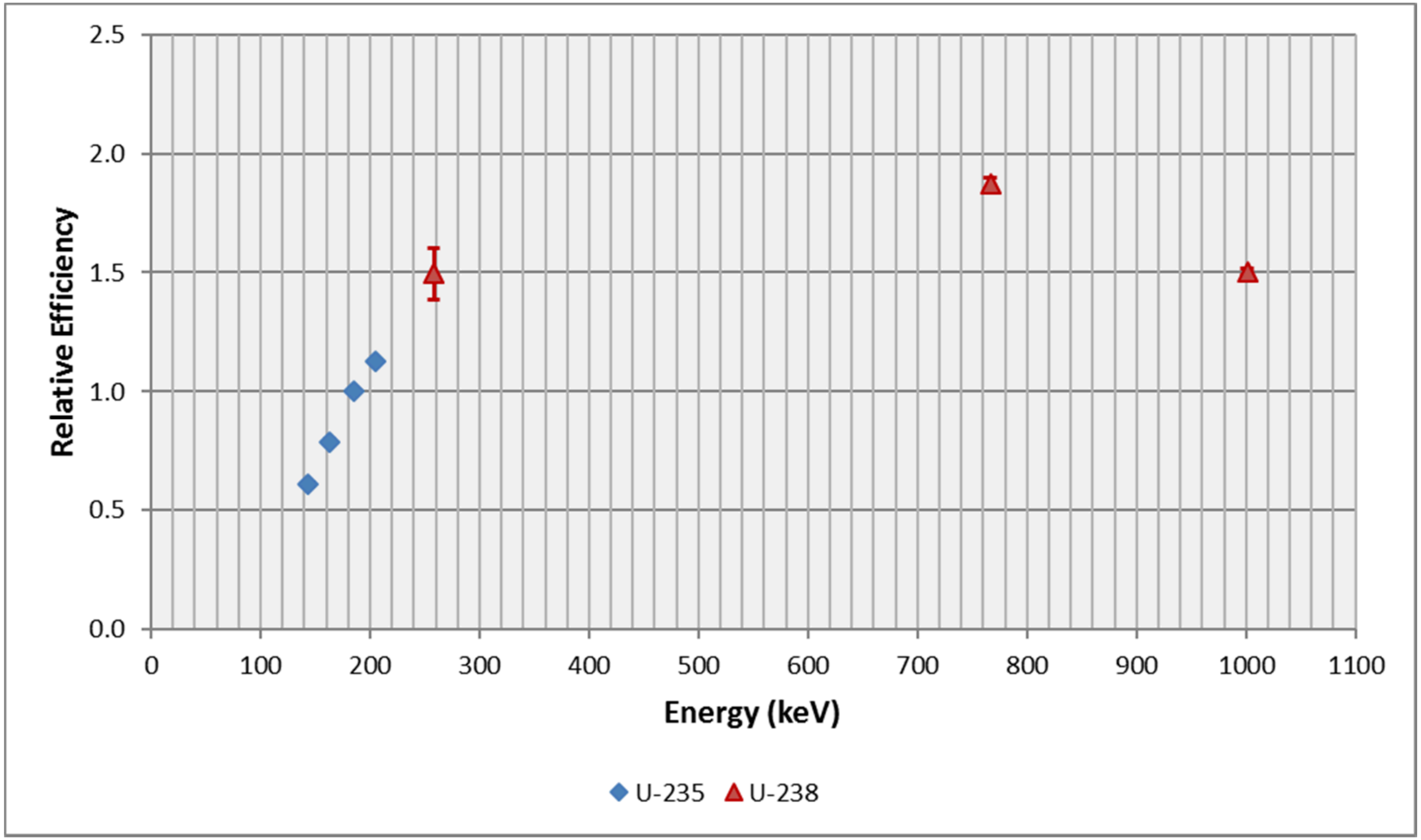
Nuclide	Energy keV	Yield Unitless	Net_Area Counts	Counts / Yield	Measured Rel Eff
U-238	258.26	7.27E-04	2285	3.14E+06	0.9961
U-238	766.36	2.94E-03	11561	3.94E+06	1.2480
U-238	1001.03	8.36E-03	26372	3.16E+06	1.0000

- Results normalized to the U-238 1001.0 keV emission.



Example B

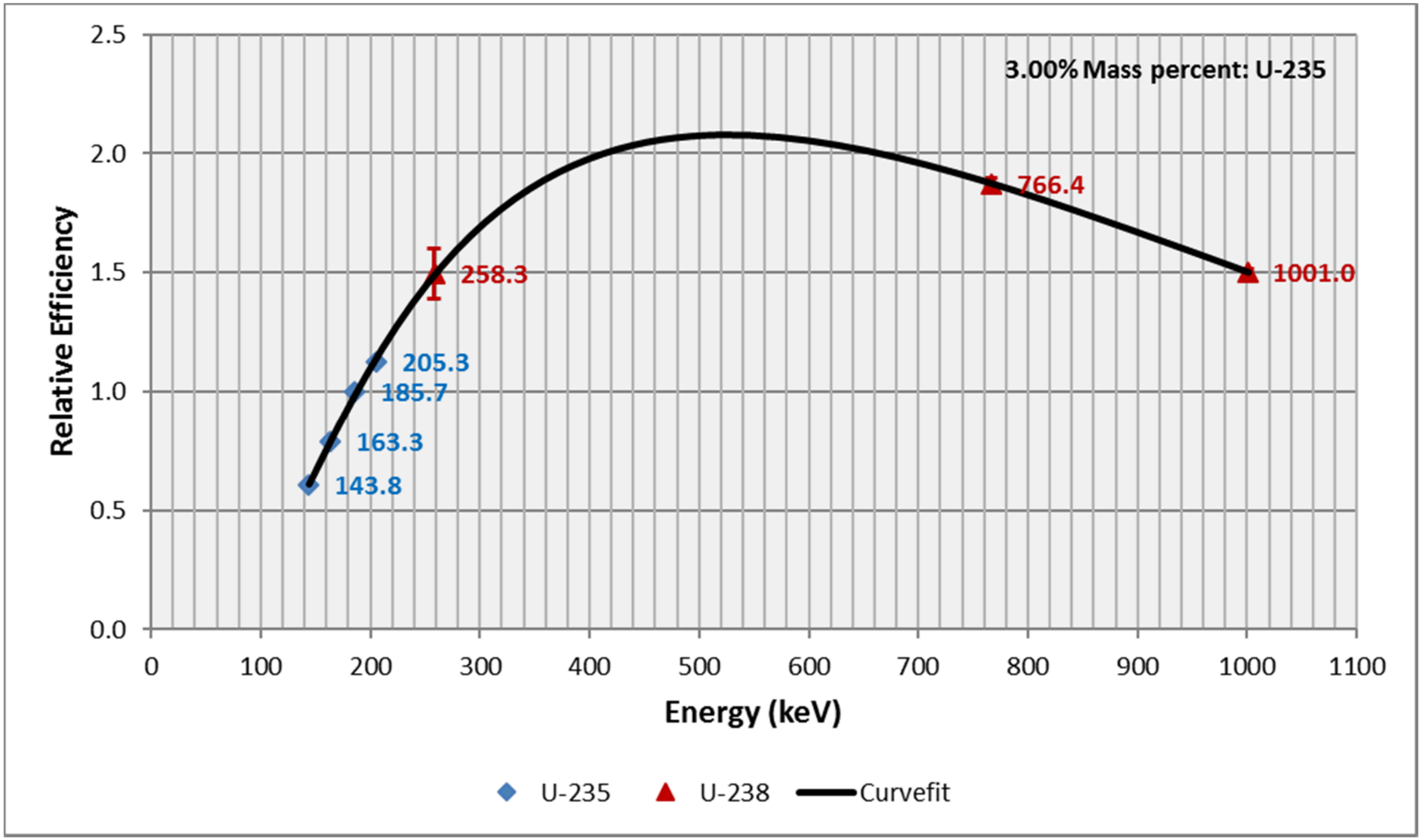
5. Scale nuclide efficiencies relative to the first selected nuclide.



- Note: The 258.3 keV U-238 peak is used to link to the low energy U-235 emissions to the high energy U-238 emissions.

Example B

6. Curve-fit the data to determine the relative efficiency as a function of energy.



- Note: Actual 2.95% mass percent U-235.
- Analysis performed using SNL_Relative_Eff_V007_Uiso.xlsx.

Additional discussion

- A similar process would generally be used for analysis of other depleted, natural, and low enriched uranium samples.

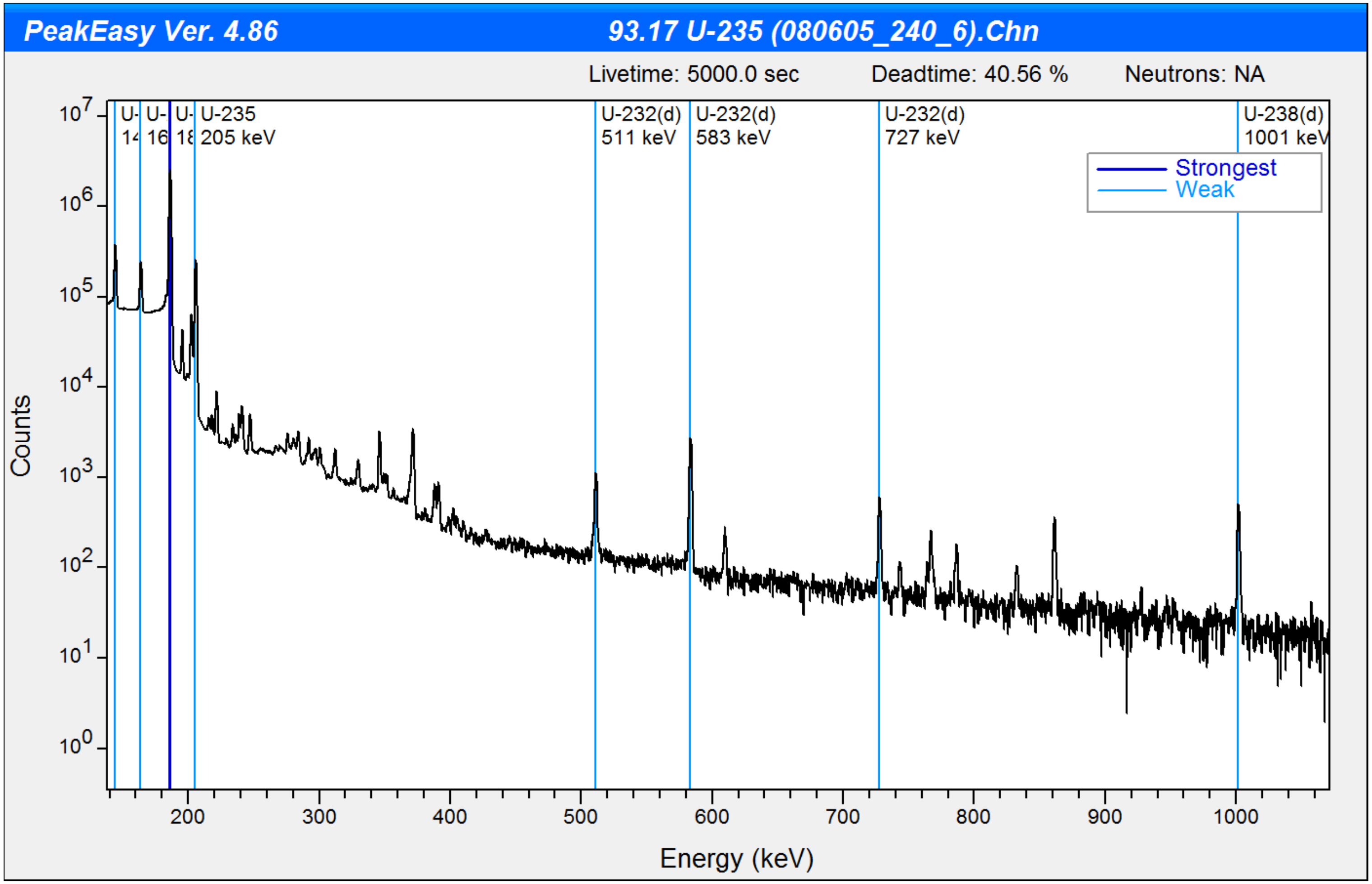
- For highly enriched uranium samples, U-232 emissions (238.6, 583.2, 727.3, and 860.6 keV) are commonly used to bridge the gap in the relative efficiency curve between the U-235 low energy emissions (143.8, 163.3, 185.7, and 205.3 keV) and the U-238 high energy emissions (742.8, 766.4, and 1001.0 keV).

- The U-238 emissions used for relative efficiency curve generation are from Pa-234m (and to a lesser extent Pa-234).

  - Unless the age on the uranium is known, equilibrium must be established between U-238 and Pa-234m (roughly 80 days) for accurate U-238 quantification and uranium isotopic analysis.
  - If equilibrium is assumed yet U-238/Pa-234m equilibrium has not been reached, the assessed uranium enrichment will be biased high.

Example C: Highly enriched uranium (HEU)

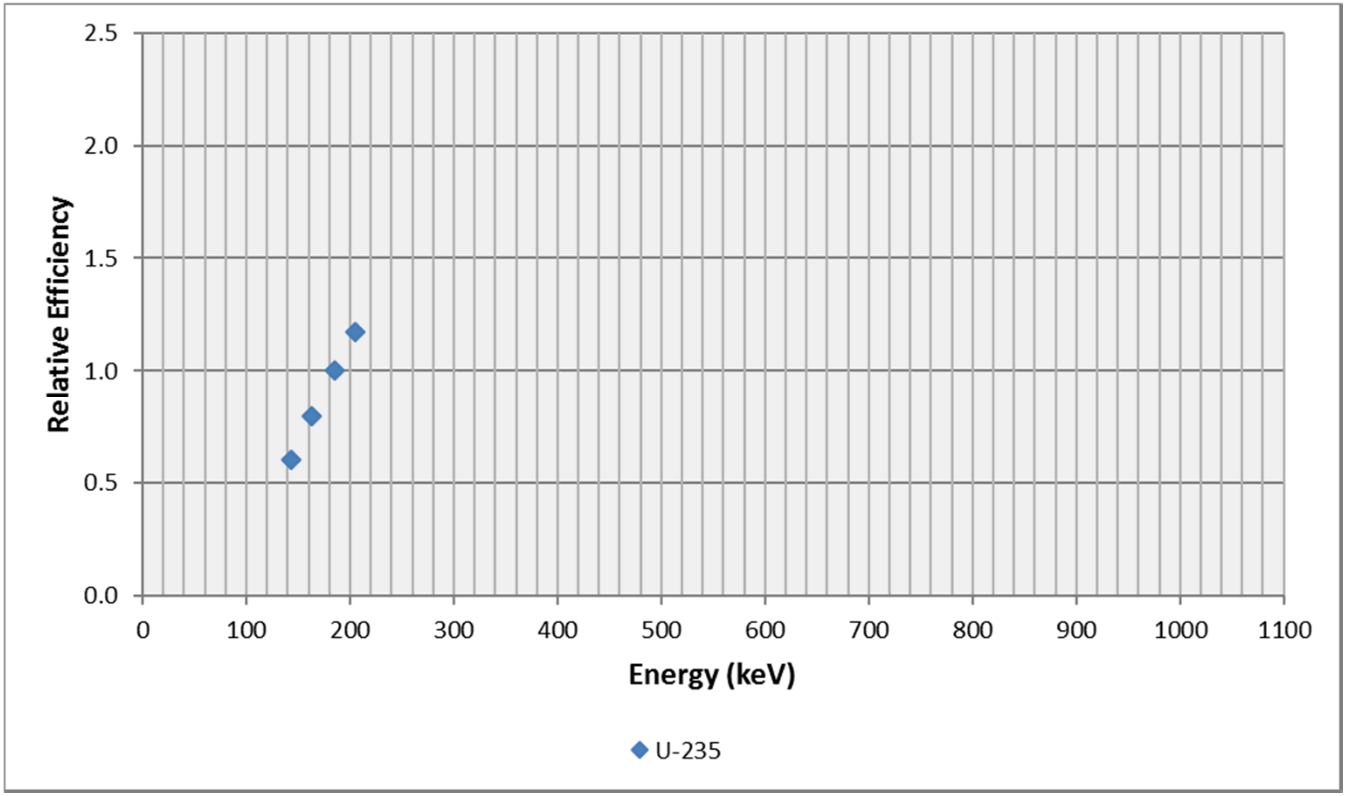
- ORTEC Detective measurement of a known HEU standard.



Example C

- U-235

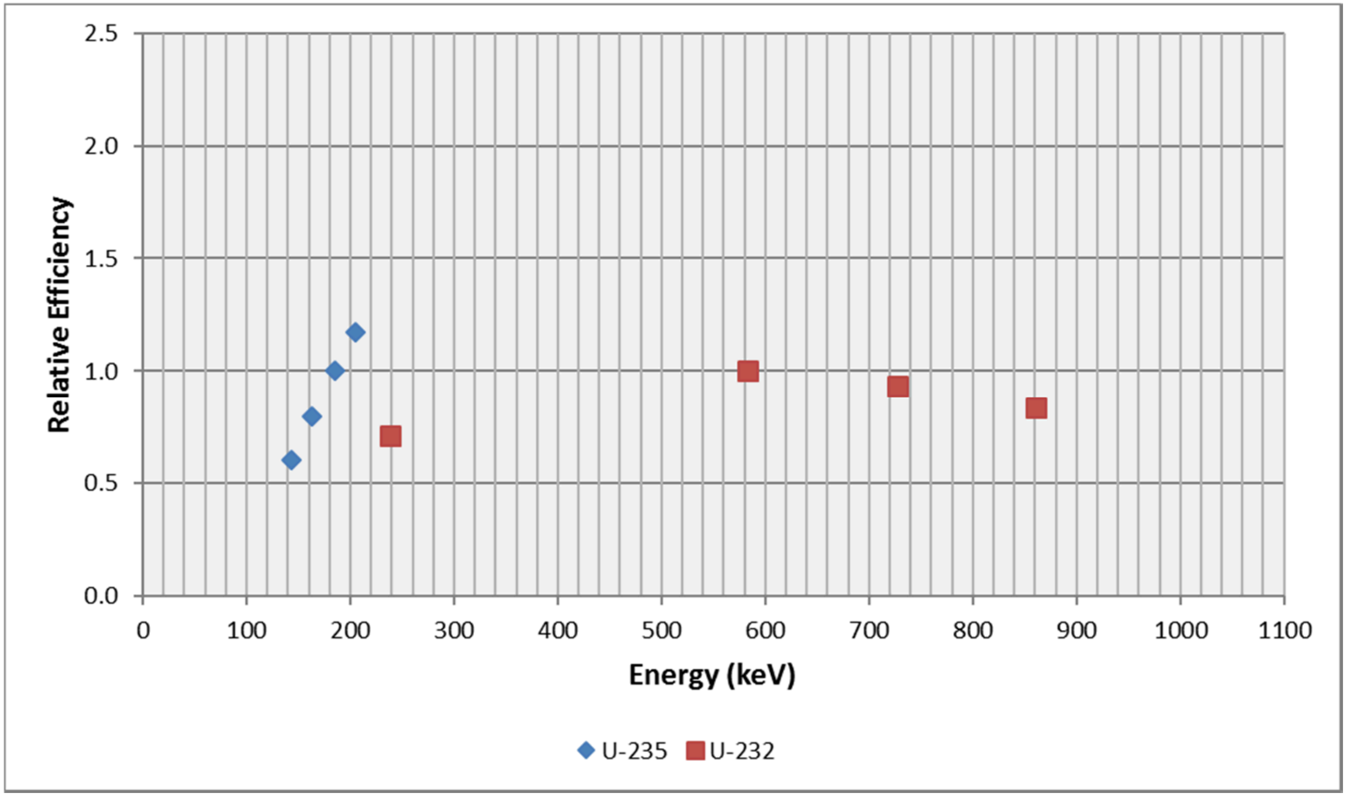
Nuclide	Energy keV	Yield Unitless	Net_Area Counts	Counts / Yield	Measured Rel Eff
U-235	143.76	1.10E-01	1282939	1.17E+07	0.6022
U-235	163.33	5.08E-02	785728	1.55E+07	0.7986
U-235	185.72	5.72E-01	11077932	1.94E+07	1.0000
U-235	205.31	5.01E-02	1138432	2.27E+07	1.1733



Example C

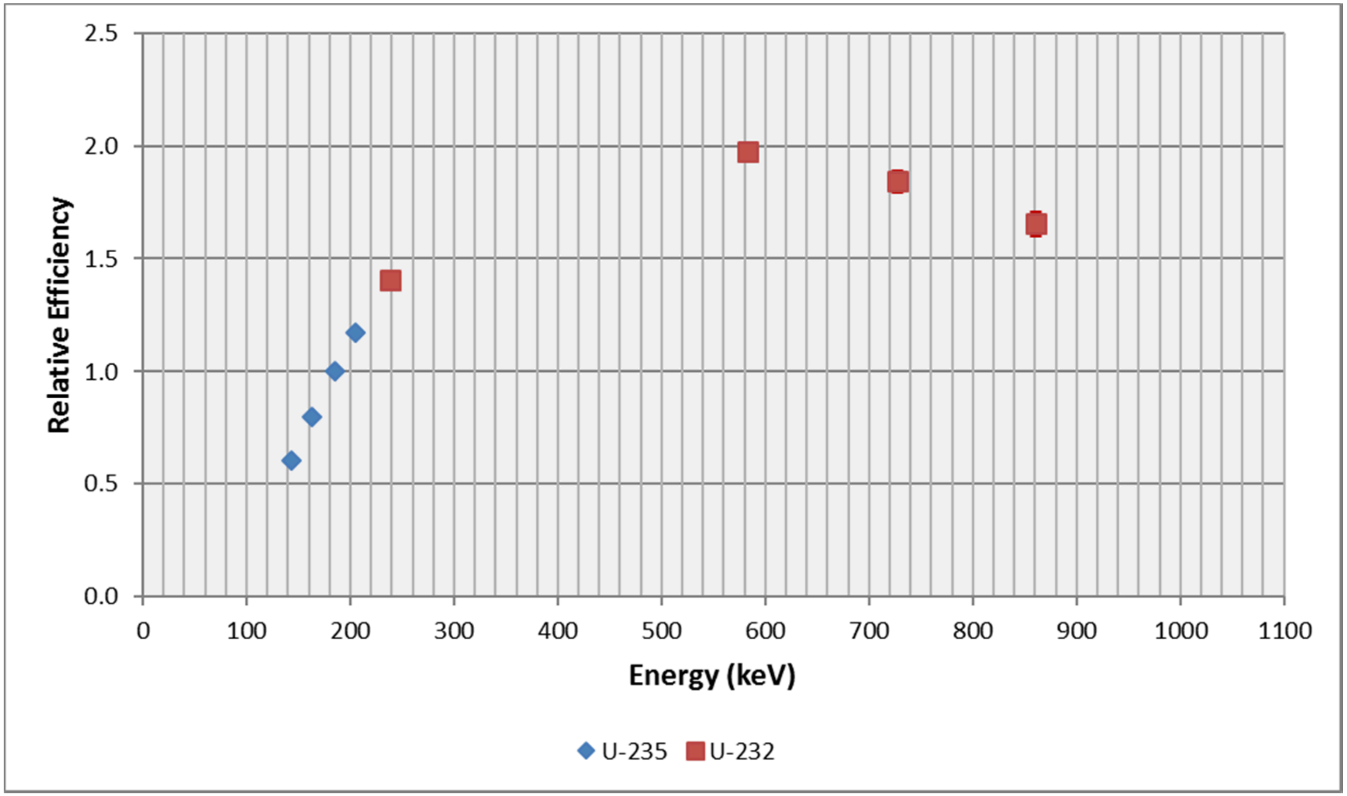
- U-232 without activity scaling

Nuclide	Energy keV	Yield Unitless	Net_Area Counts	Counts / Yield	Measured Rel Eff
U-232	238.63	4.33E-01	14899	3.44E+04	0.7123
U-232	583.19	3.04E-01	14685	4.83E+04	1.0000
U-232	727.33	6.58E-02	2964	4.50E+04	0.9325
U-232	860.56	4.47E-02	1808	4.04E+04	0.8373



Additional discussion

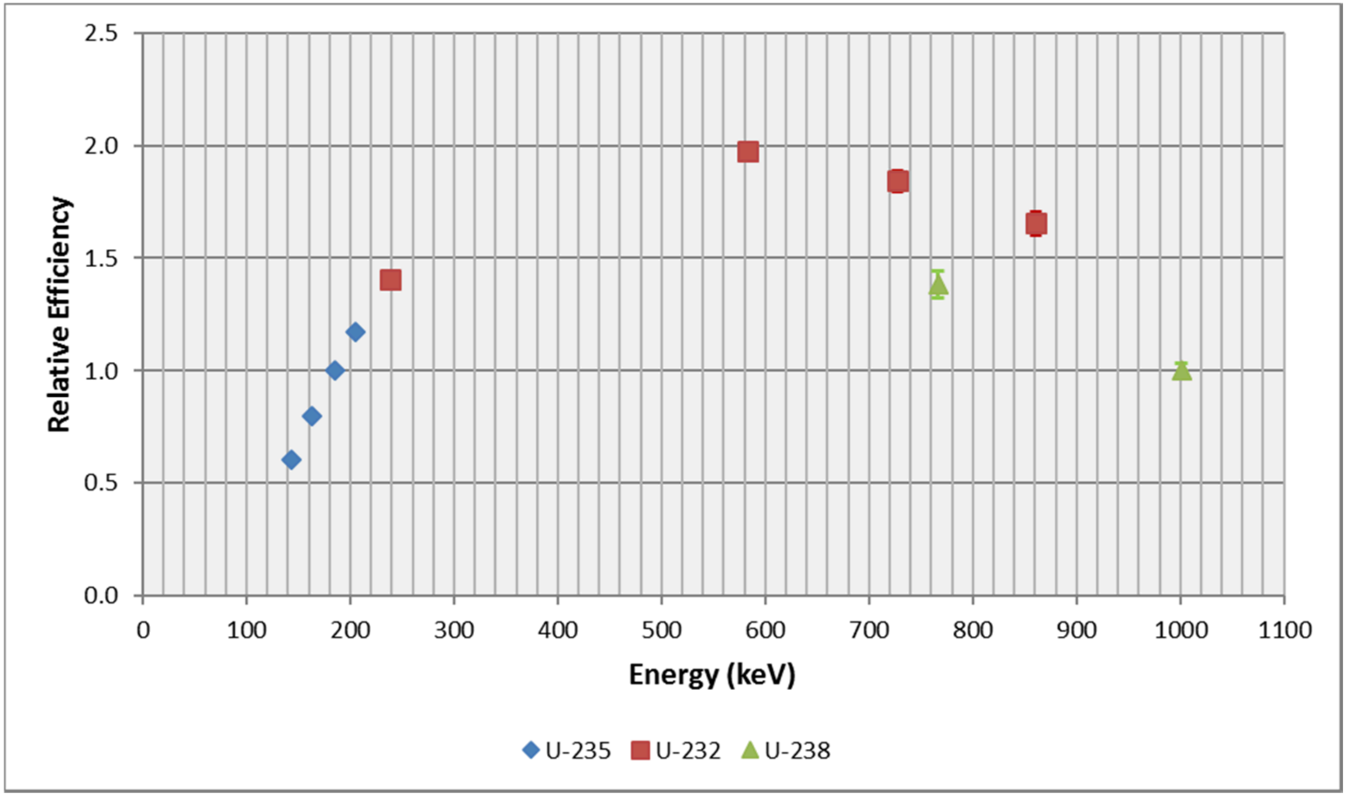
- U-232 activity scaled to U-235



Example C

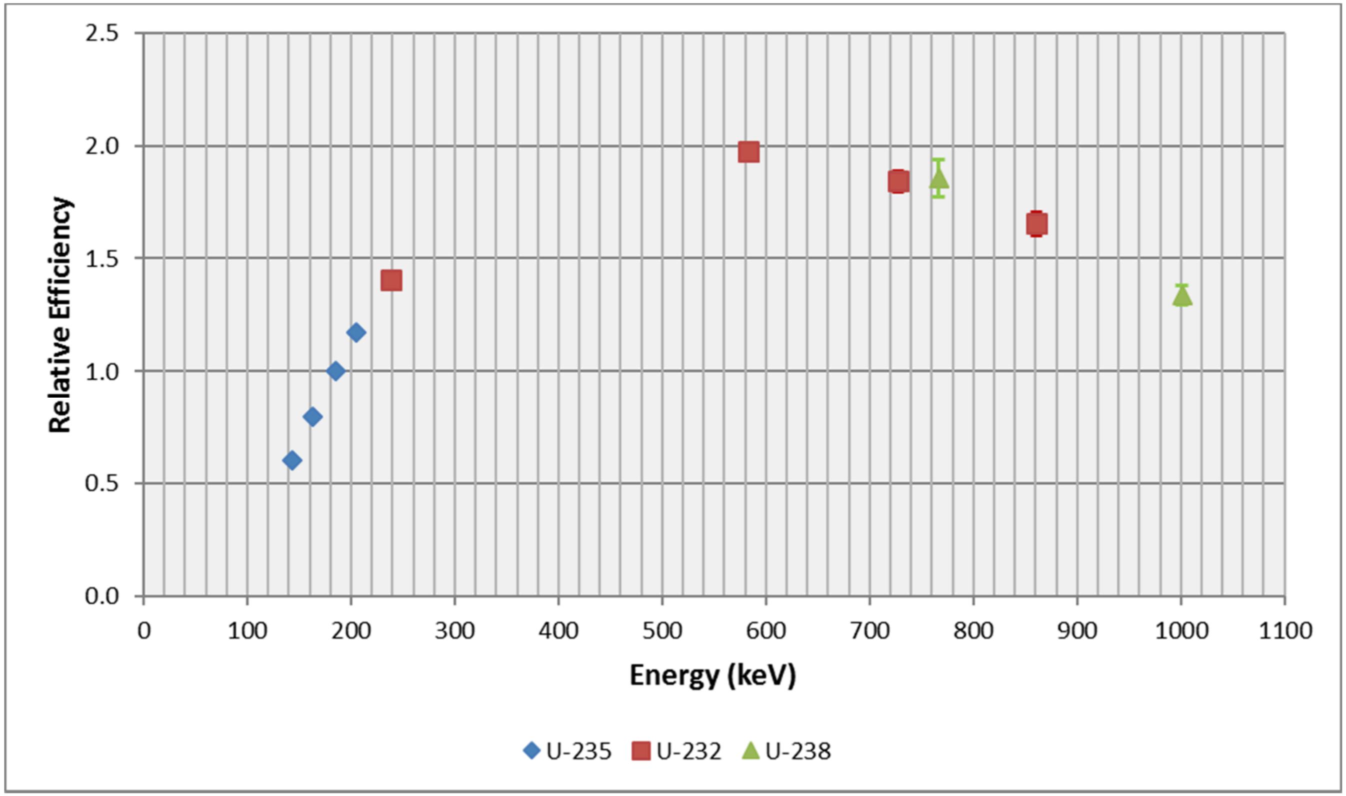
- U-238 without activity scaling

Nuclide	Energy keV	Yield Unitless	Net_Area Counts	Counts / Yield	Measured Rel Eff
U-238	766.36	2.94E-03	1298	4.42E+05	1.3856
U-238	1001.03	8.36E-03	2667	3.19E+05	1.0000



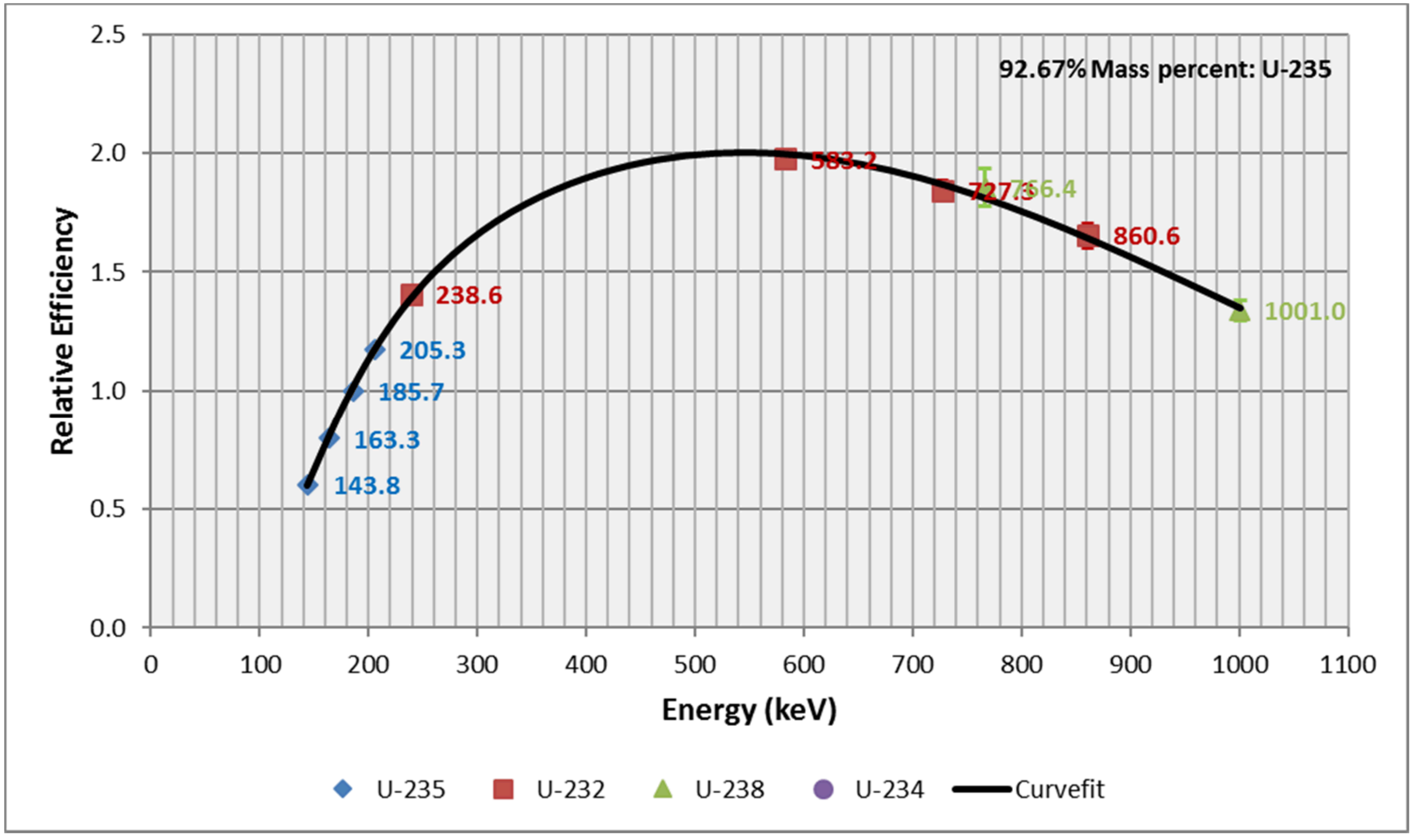
Example C

- U-238 activity scaled to U-235 and U-232



Example C

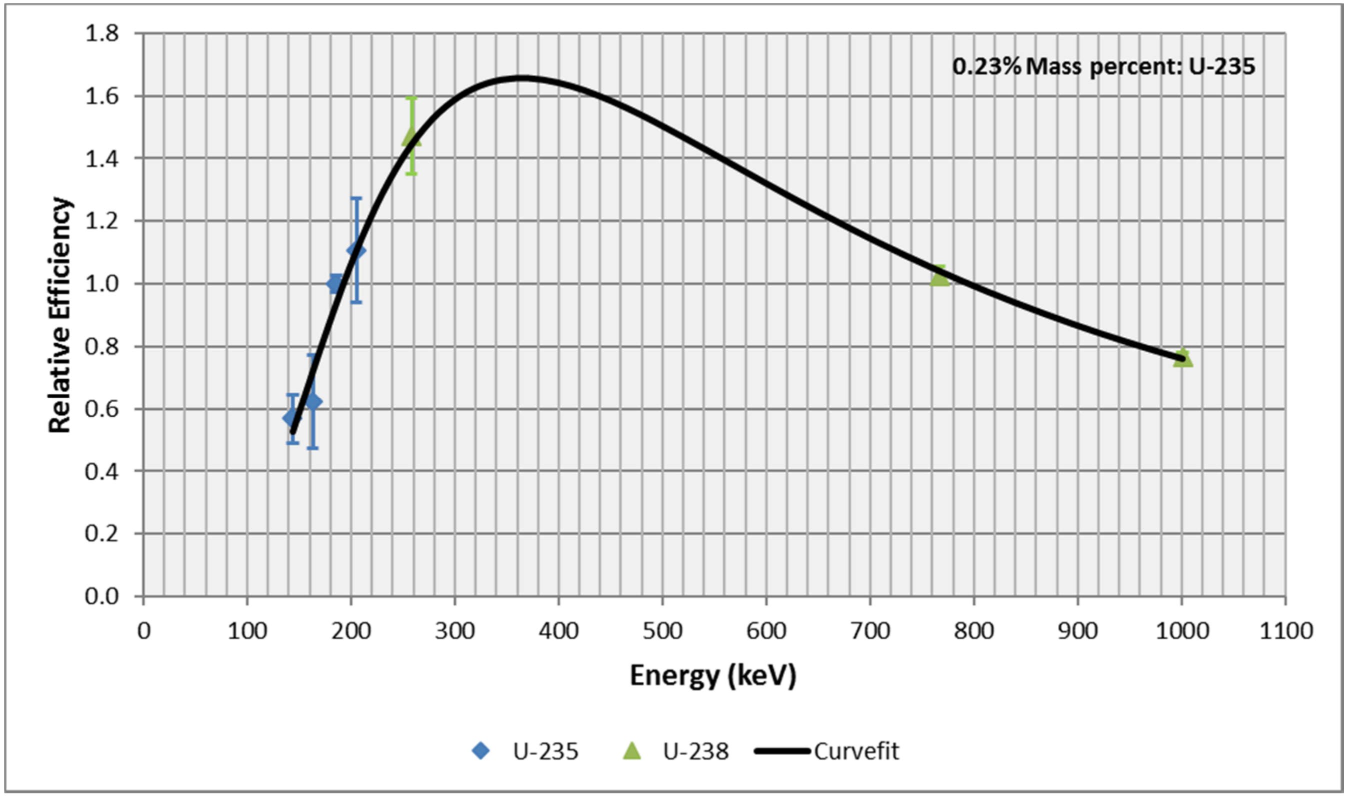
- Final curve-fit



- Note: Actual 93.17% mass percent U-235.
- Analysis performed using SNL_Relative_Eff_V007_Uiso.xlsx.

Example D: Real world example

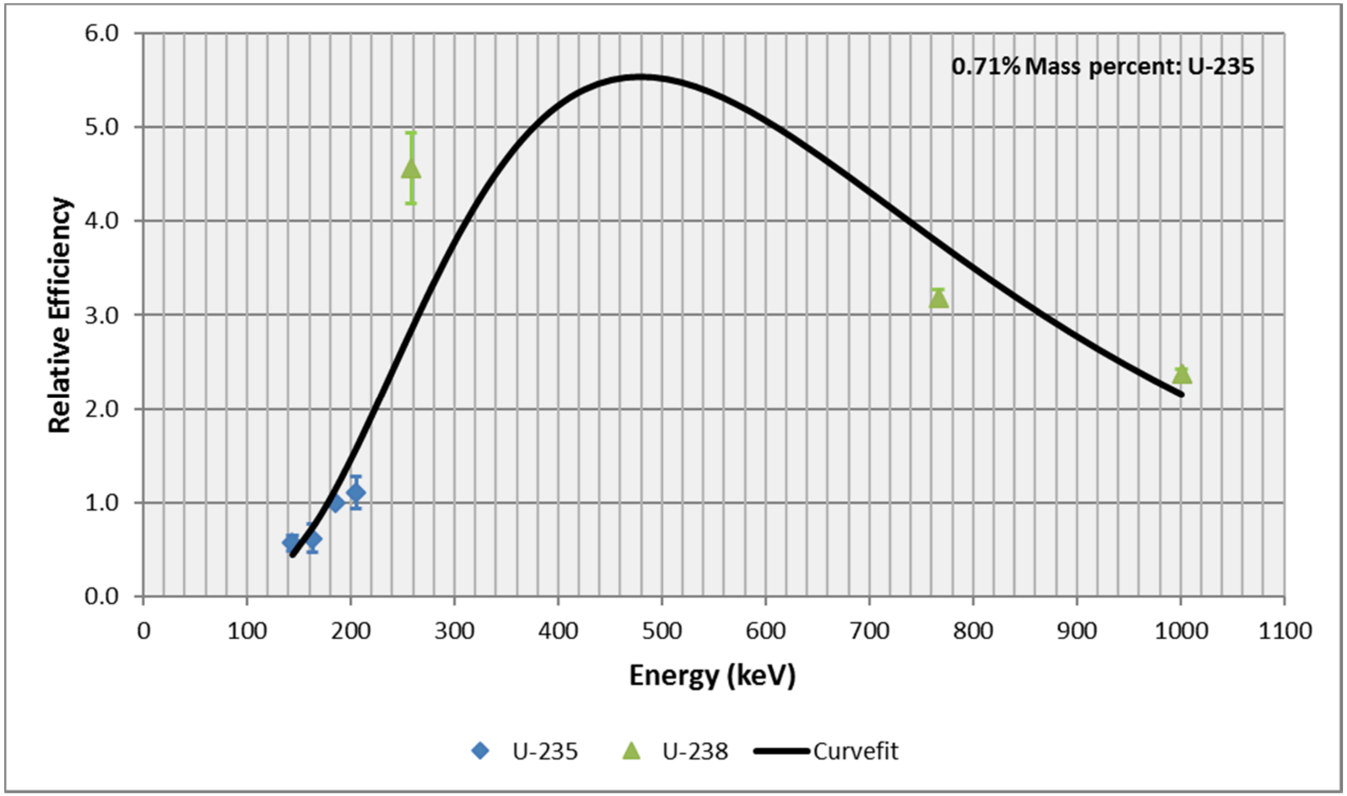
- Uranium solution with unknown isotopics.
- Best fit is consistent with depleted uranium.



- Could it be natural uranium?

Example D: Real world example

- A relative efficiency curve fit forcing the U-235 mass percentage to 0.71% shows natural uranium is not likely.



Additional thoughts

- Remember counting statistics determine the ability to obtain precise results.
- Review full-energy peak fits for reasonableness.
- Review the spectrum for interferences.
- If U-232 is used in the relative efficiency curve, it might be necessary to perform proper background subtraction to accurately determine U-232 full-energy peak areas.

Additional thoughts

- Consider the use of graded shielding (Cd-Cu or Sn-Cu) to eliminate/reduce low energy emissions (e.g., Am-241 59.5 keV emissions in plutonium).
  - May allow closer sample to detector distances to be used (reducing detection limits and increasing higher energy emission count rates).
  - Reduces “true coincidence” with a low-energy photon emissions.
- Planer HPGe detectors are typically used for low energy analysis and coaxial HPGe detectors are used for higher energy analysis.
  - Planar HPGe detectors usually achieve the best energy resolution, because of their low capacitance, and are therefore preferred for analysis of the complex low-energy gamma-ray and x-ray spectra of uranium and plutonium.

Additional thoughts (Reference: LA-14018)

- "Random summing is assumed to result in equal losses throughout the entire spectrum. Thus, isotopic ratio methods should be unaffected. This assumption may not hold precisely because peak widths increase with energy, but it appears that the losses occurring from random summing do not materially affect isotopic ratio measurements."

- However, random summing can make isotopic analysis more difficult.

  - For example, if high dead times are present with large amounts of Am-241, then random sum peaks from Am-241 (59.5 keV with 99.0 and 103.0 keV) can further complicate the assessment of the 160.3 keV Pu-240 peak area used for the determination of plutonium isotopics.

Additional thoughts (Reference: LA-14018)

- "Coincidence summing is isotope dependent and does not cancel out as random summing does. The effect is proportional to the solid angle of the detector as seen from the sample and can be reduced by increasing the sample-to-detector distance."

  - "Coincidence summing effects are present in low-enriched uranium measurements analyzed in the 120-1001 keV energy range. The 258-keV gamma ray from the U-238 daughter Pa-234m is particularly affected."

  - "Coincidence summing is not recognized to be a problem with plutonium measurements."