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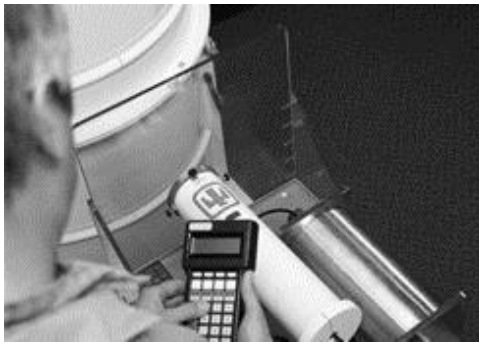

Radiation Protection Sample Diagnostics (RPSD) Gamma Spectroscopy Training: Session 001

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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Discussion topics

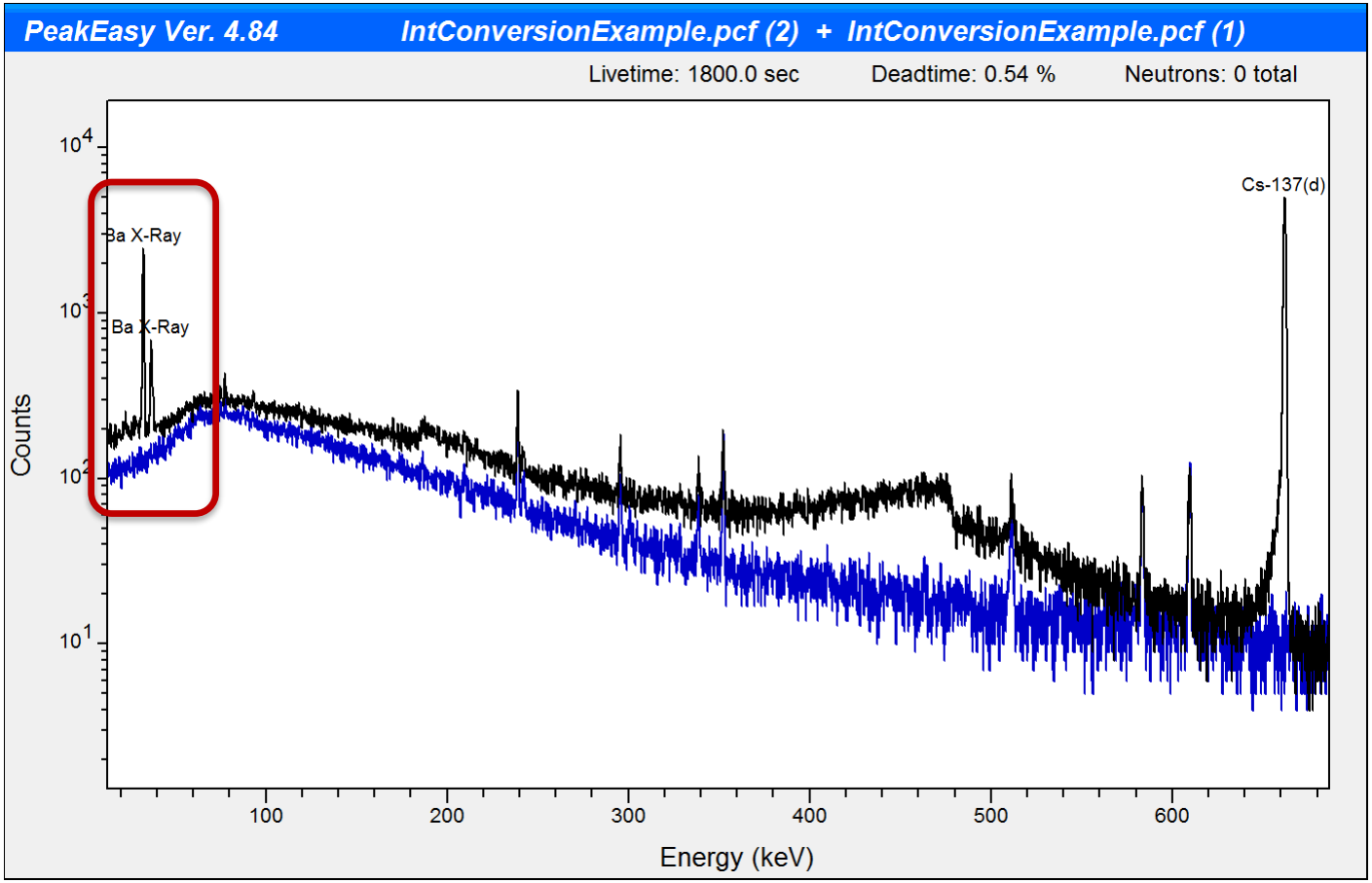
- Characteristic x-rays
- Quick detection limit calculations
- Age determination of materials
- Uranium isotopic mass and activity fractions
- Uranium gamma spectroscopy notes
- Plutonium isotopic mass and activity fractions
- Plutonium gamma spectroscopy notes
- Differential attenuation analysis

Characteristic x-rays from internal conversion

- Following radioactive decay, a decay product in an excited state is formed.
- Generally, the excited state emits a photon to return to ground state.
- Alternatively, an orbital electron can be ejected to return to ground state (internal conversion).
- Following internal conversion, outer orbital electrons fill the lower energy levels producing characteristic x-rays.
  - These x-rays are characteristic of the decay product element.
  - The ratio of internal conversion electrons to gamma emission photons is known as the internal conversion coefficient.

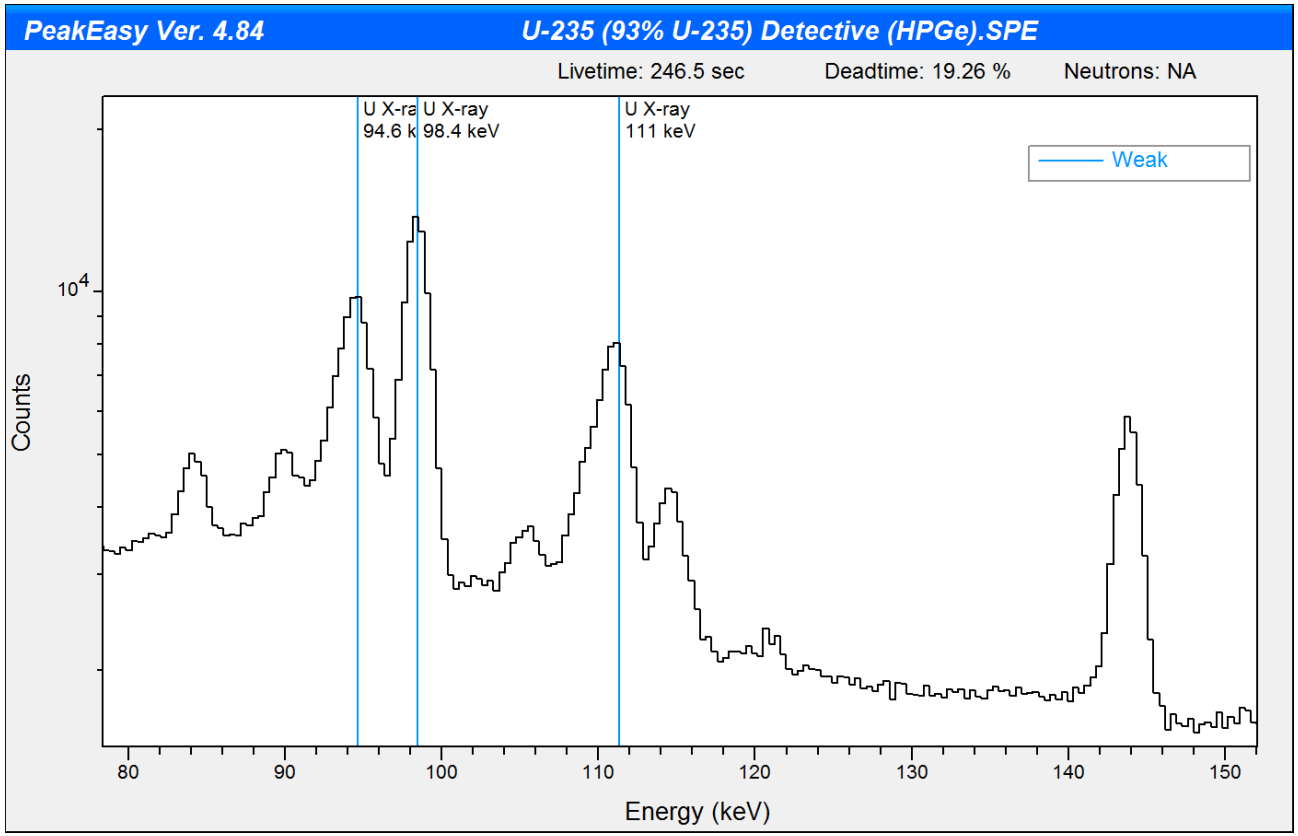
Characteristic x-rays from internal conversion

- Cs-137 decays by beta emission to the excited state of Ba-137m.
- The internal conversion process generates characteristic Ba x-rays.
- Spectrum: Unshielded Cs-137 with characteristic Ba x-rays.



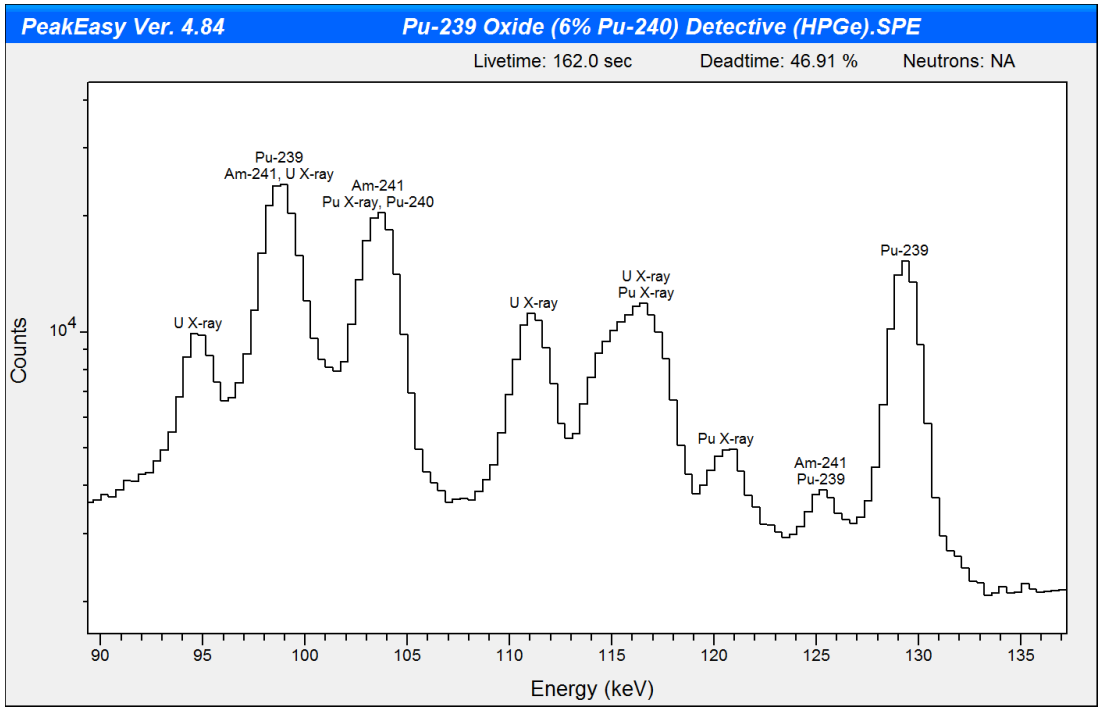
Characteristic x-rays from induced x-ray fluorescence

- X-ray fluorescence results from the ionization of atoms, for which the excited state returns to the ground state emitting x-ray photons characteristic of the element that was ionized.
- Spectrum: Highly enriched uranium (HEU) with uranium (U) x-rays from self-induced x-ray fluorescence.

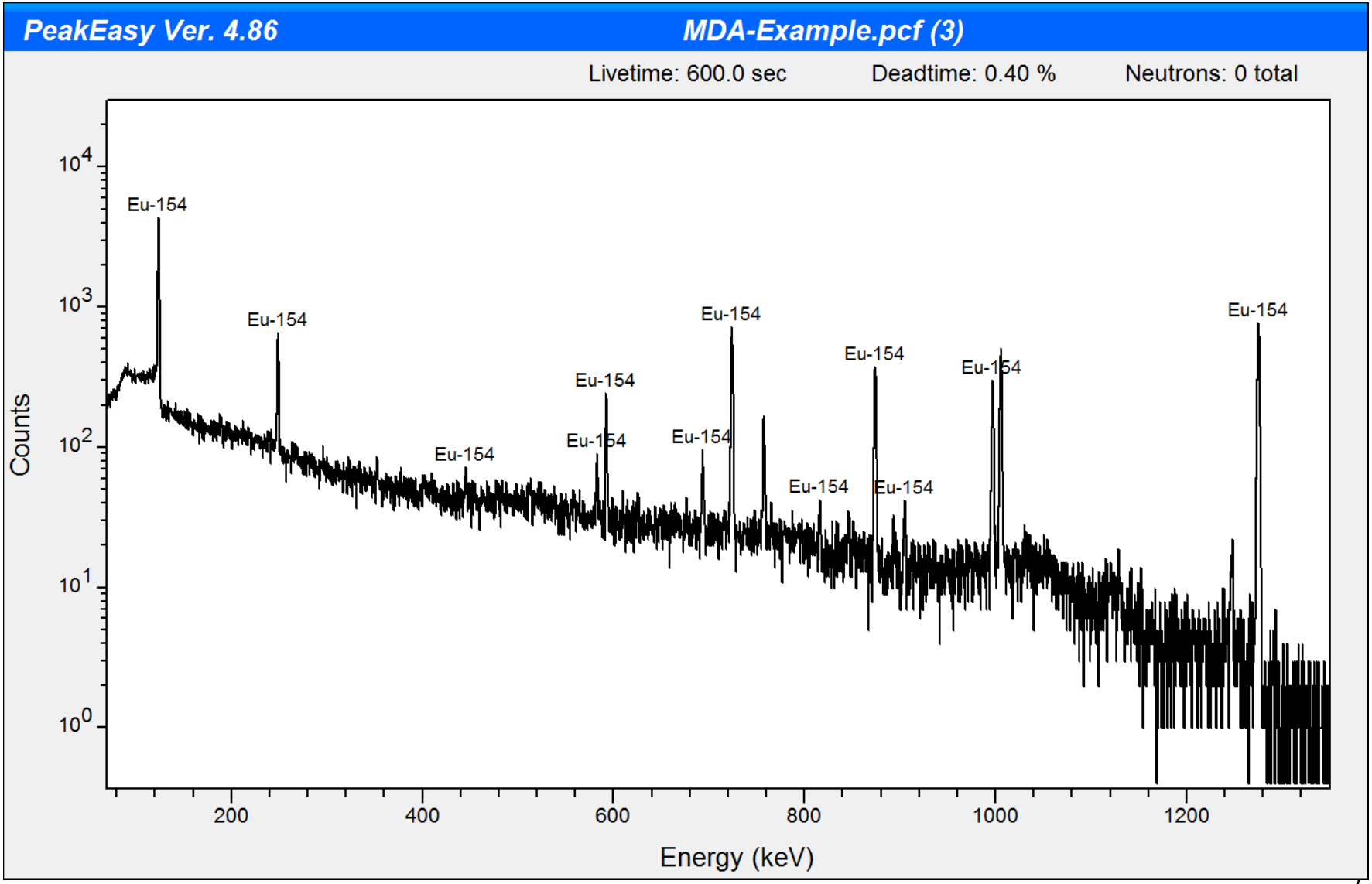


Parent and decay product element characteristic x-rays

- High-Z materials have high internal conversion coefficients which, as discussed, generate x-rays that are characteristic of the decay product element.
- In addition, high-mass or high-concentration radioactive samples produce self-induced x-ray fluorescence generating x-rays that are characteristic of the element that was ionized.
- Spectrum: Low burn-up plutonium (Pu) with parent (Pu) and decay product (U) element characteristic x-rays.



Quick detection limit calculations



Quick detection limit calculations

- A container with 500-grams of water was measured for 600-seconds with an ORTEC Detective EX-100 at 5-cm from the detector face.
  - ORTEC Detective EX-100: 65 mm x 50 mm deep, P-type HPGe based Hand-Held Radioisotope Identifier (RIID), Coaxial construction.

- An efficiency curve is available for this geometry at the measured distance.

Quick detection limit calculations

- Eu-154 has been determined to be present at approximately 100 nCi.

- Estimate the detection limit (L_d) for Ag-110m at 657.8 keV.

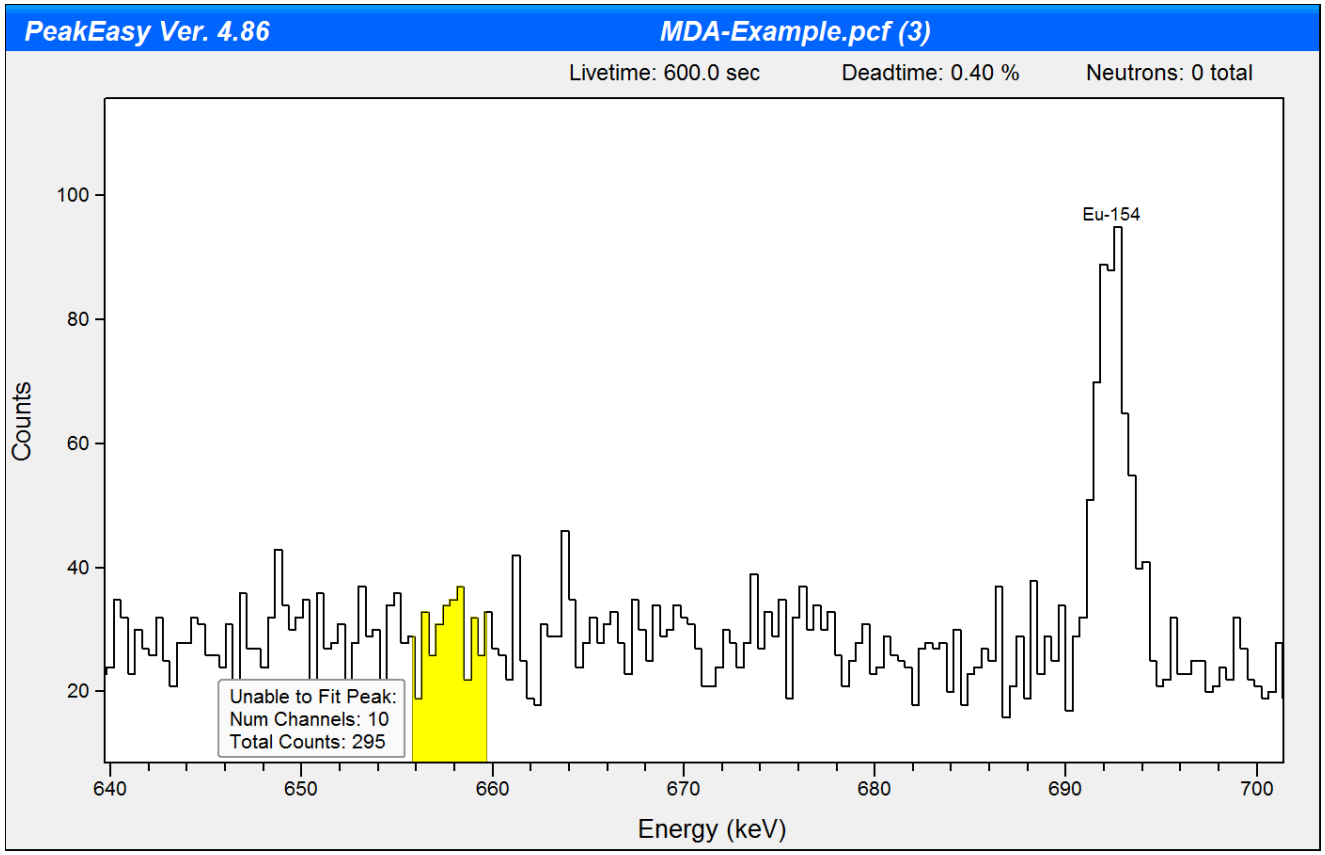
  - The absolute full-energy peak (FEP) efficiency at 657.8 keV is 9.25E-03 counts / gamma
  - The full width at half maximum (FWHM) at 657.8 keV is ≈ 1.75 keV.

Quick detection limit calculations

- The detection limit (L_d) is the minimum number of counts or activity you can be 95% confident of detecting in a sample with similar properties as the one to be counted.
  - A good overview of decision limits can be found in Chapter 5 of Practical Gamma-Ray Spectrometry, 2nd Edition, Gordon R. Gilmore.
- For a quick detection limit (L_d) estimate, use Lloyd Currie's formula $L_d = 4.66 \times \text{Background}^{1/2}$
  - Background = $2 \times \text{FWHM} \times 2.807\,\sigma / 2.355\,\sigma = 2.38\ \text{FWHM}$
  - Where FWHM = $2.355\,\sigma$ and $2.807\,\sigma$ is the coverage factor for 99.5% of the whole peak area

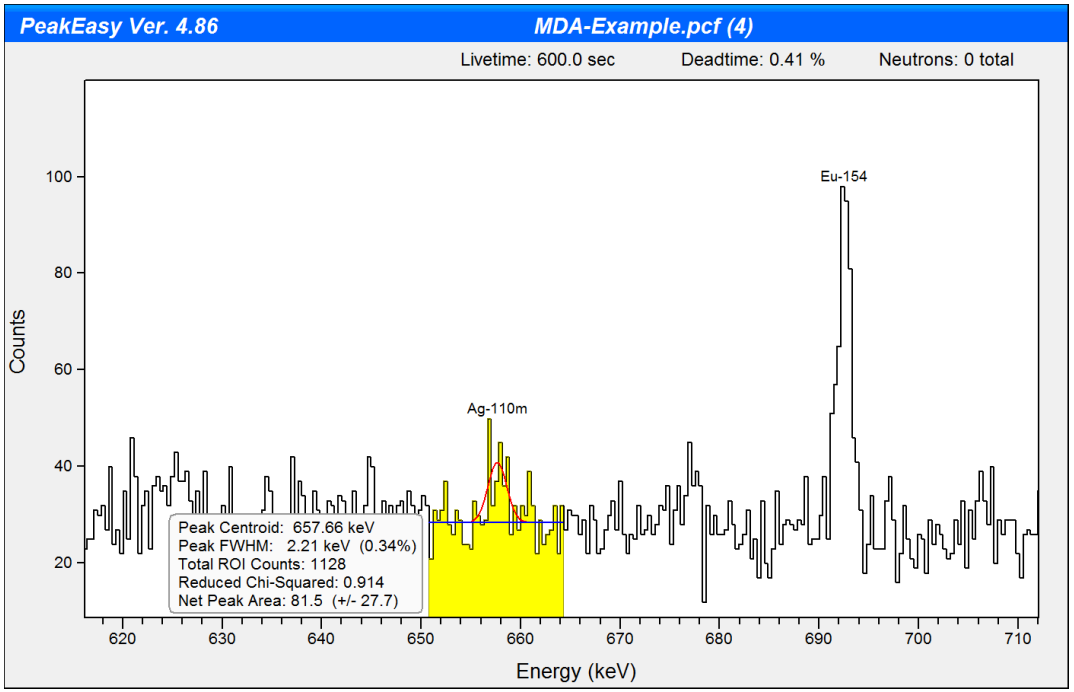
Quick detection limit calculations

- 657.8 keV background Region Of Interest (ROI) determination
  - Using a typical EX-100 FWHM at 657.8 keV of ≈ 1.75 keV, the background ROI (2.38 FWHM) is 655.7 keV to 659.7 keV which corresponds to ≈ 295 counts



Quick detection limit calculations

- The calculated detection limit (L_d) is 4.66 × (295 counts)$^{1/2}$ = 80 counts
- The calculated detection limit (L_d) converted to activity = 0.4 nCi
  - L_d = (80 counts / 600 seconds) × (gamma / 9.25E-03 counts) × (disintegration / 0.94 gammas) / (3.7E+01 nCi/Bq) = 0.4 nCi
- The spectrum below includes the addition of 0.4 nCi Ag-110m to the previous Eu-154 spectrum using the Gamma Detector Response and Analysis Software (GADRAS) inject tool.



Material age determinations

- Gamma spectroscopy can be a useful technique for age determinations (chronology) of materials.

- The age can be determined using gamma spectroscopy results for suitable parent/decay product nuclides in conjunction with decay/in-growth calculations.

Material age determinations

- For material age determinations, it is generally assumed that no decay product activity is present immediately following chemical separation (aka, 100% chemical separation).
  - When all of the decay product activity is not removed during the chemical separation process, the age calculated will appear to be longer than it actually is resulting in an age estimate that is biased high.

Material age determinations

- Equation: Assuming decay product nuclide activity is equal to 0 at the time of chemical separation.

$$A_D = B * A_{PO} * (\lambda_D/(\lambda_D-\lambda_P)) * (EXP(-\lambda_P * t) - EXP(-\lambda_D * t))$$

A_D is the decay product nuclide activity

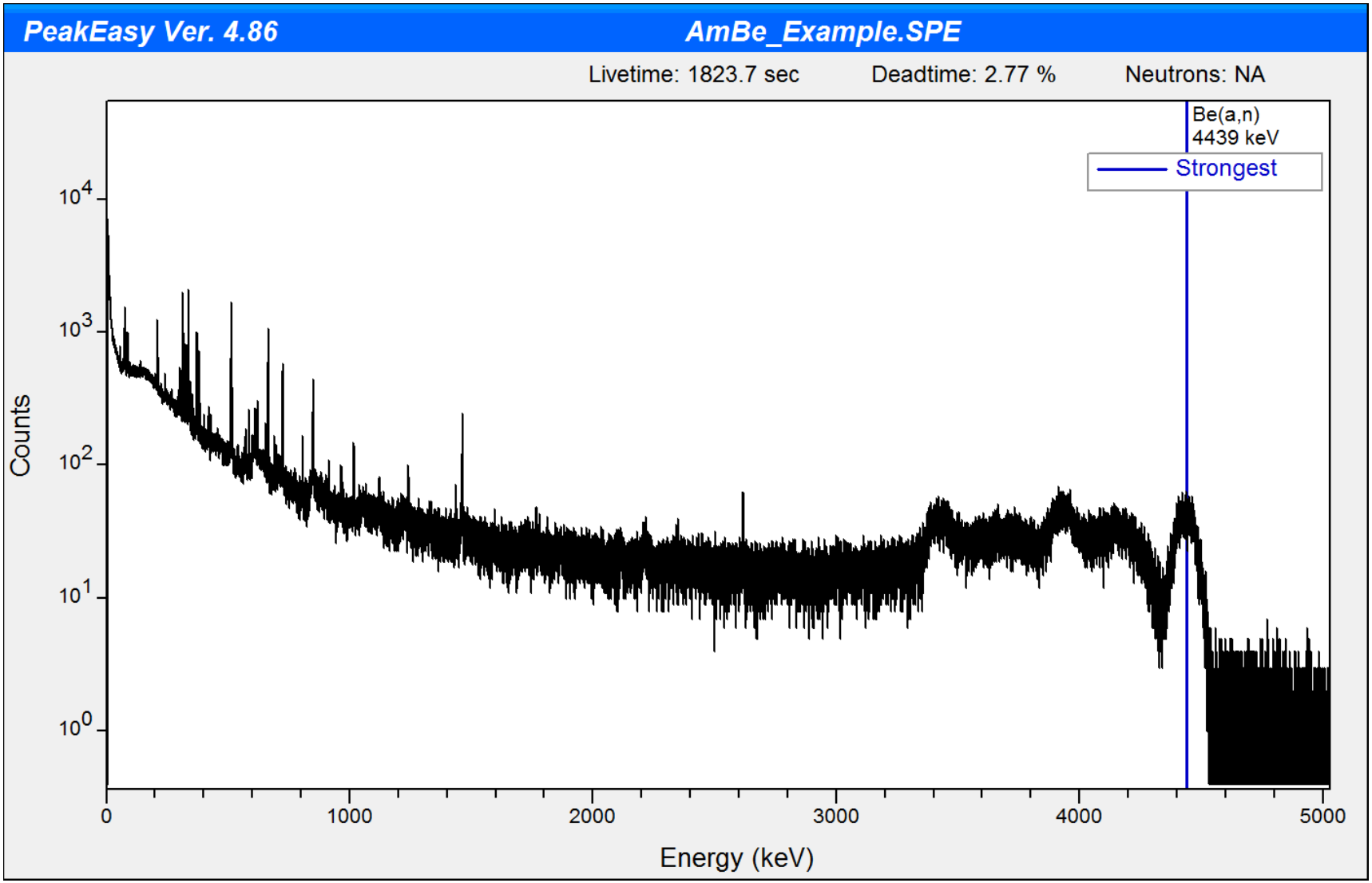
A_{PO} is the initial parent nuclide activity

λ is the decay constant in $days^{-1}$

t is the decay time in days

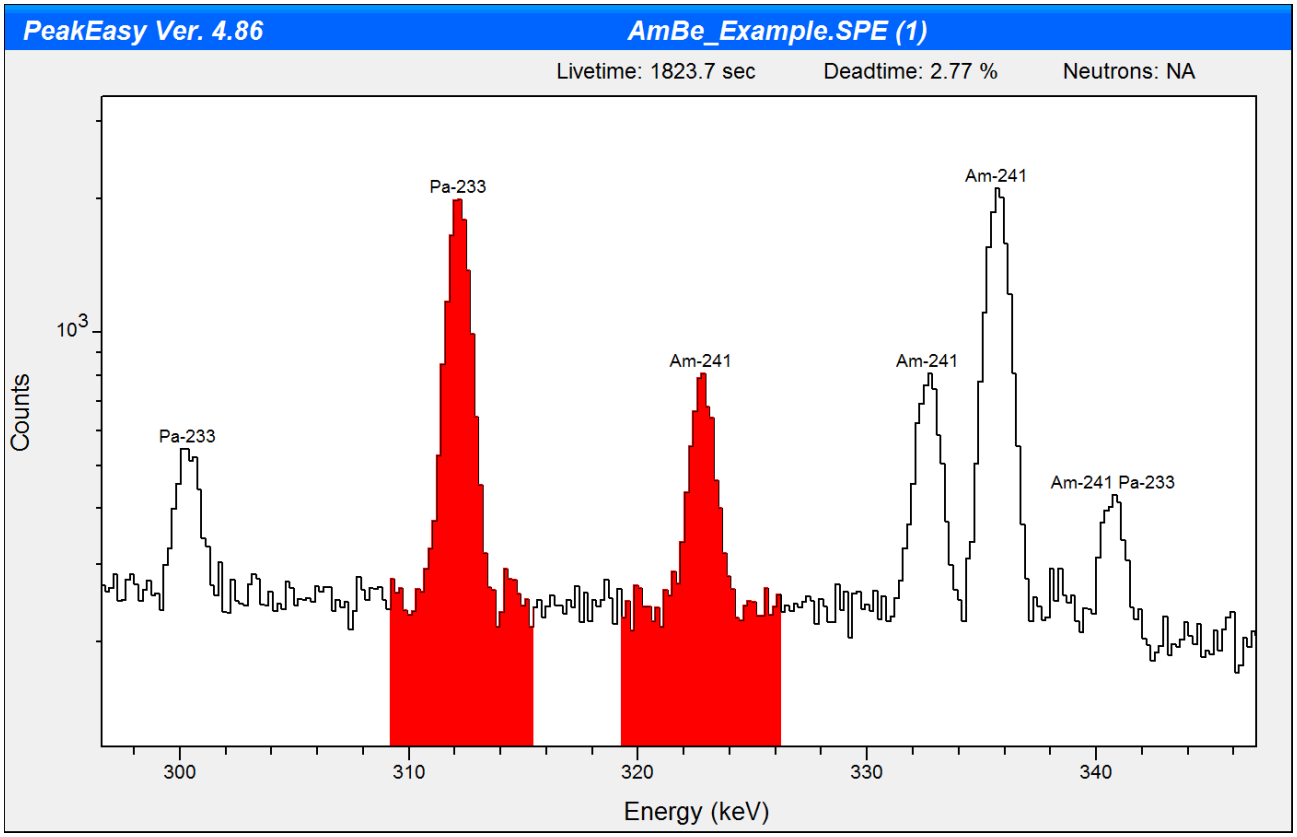
B is the branching fraction

Example: AmBe source: Be-9(α,nγ)C-12



Example: AmBe source: Be-9(α,nγ)C-12

- For a quick “rough” age estimate, assume the efficiency is the same for the 312.2 and 322.5 keV emissions from Pa-233 and Am-241, respectively, and normalize the peaks areas by the yield to determine the Np-237/Am-241 activity ratio.



Example: AmBe source: Be-9(α,nγ)C-12

- Full-energy peak area calculations.

Nuclide	Energy (keV)	Yield gps/dps	Net_Area counts	Net_Area Uncertainty	Counts/Yield
Am-241	322.5	1.52E-06	3583	92	2.36E+09
Pa-233	312.2	3.86E-01	11222	127	2.91E+04

- Np-237/Am-241 activity ratio estimate = 2.91E+04 / 2.36E+09 = 1.2E-05

Example: AmBe source: Be-9(α,nγ)C-12

- Using the calculated "rough" Np-237/Am-241 activity ratio of 1.2E-05 and assuming no Np-237 is present following chemical separation, the age is estimated at 35 years using the following table.

Time (Years)	Activity Ratio Np-237/Am-241	Activity Ratio Am-241/Np-237
1	3.24E-07	3.08E+06
2	6.49E-07	1.54E+06
3	9.74E-07	1.03E+06
4	1.30E-06	7.69E+05
5	1.63E-06	6.15E+05
10	3.27E-06	3.06E+05
15	4.92E-06	2.03E+05
20	6.58E-06	1.52E+05
25	8.26E-06	1.21E+05
30	9.95E-06	1.00E+05
35	1.17E-05	8.58E+04
40	1.34E-05	7.47E+04
45	1.51E-05	6.62E+04
50	1.69E-05	5.93E+04
55	1.86E-05	5.37E+04
60	2.04E-05	4.90E+04
65	2.22E-05	4.51E+04
70	2.40E-05	4.17E+04

Typical uranium mass and activity percentages

Typical Uranium Mass Percentages

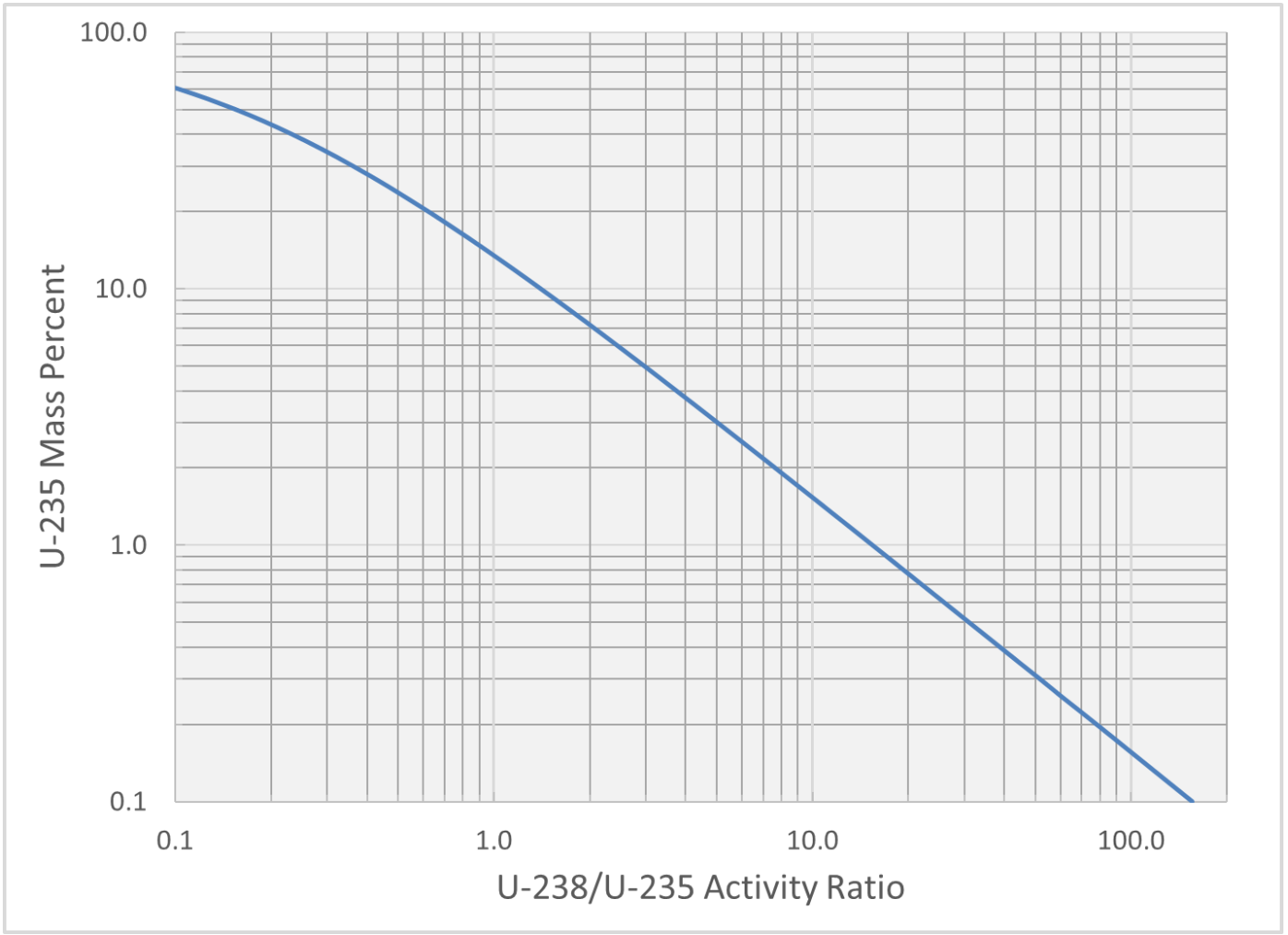
Nuclide	Depleted Uranium Mass %	Natural Uranium Mass %	3% Enriched Mass %	93.3% Enriched Mass %
U-238	99.7995%	99.2830%	96.9732%	5.8308%
U-235	0.1995%	0.7115%	3.0000%	93.3000%
U-234	0.0010%	0.0055%	0.0268%	0.8692%

Typical Uranium Activity Percentages

Nuclide	Depleted Uranium Activity %	Natural Uranium Activity %	3% Enriched Activity %	93.3% Enriched Activity %
U-238	84.039%	48.260%	15.858%	0.035%
U-235	1.080%	2.223%	3.153%	3.592%
U-234	14.900%	49.510%	80.989%	96.373%

- Although not listed above, U-232 and U-236 are produced during reactor irradiation and are present in U that has been reprocessed.
- Typical U enrichment processes enrich the lighter U isotopes (U-232, U-234, U-235) more readily than the heavier U isotopes (U-236, U-238).

Typical uranium mass and activity percentages



- Adapted from Rucker, T.L. & Johnson, C.M. J Radioanal Nucl Chem (1998) 235: 47.

Uranium gamma spectroscopy

- Primary U 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

- Typical U-238/U-235 activity ratios
  - Depleted uranium between 50:1 and 80:1.
  - Natural uranium is 22:1

Uranium gamma spectroscopy

- The U-238 gamma emissions listed previously (258.3, 742.8, 766.4, and 1001.0 keV) are from Pa-234m (and to a lesser extent Pa-234).
  - Unless the age on the U is known, equilibrium must be established between U-238 and Pa-234m (roughly 80 days) for accurate U-238 quantification and U isotopic analysis.
  - If equilibrium is assumed yet U-238/Pa-234m equilibrium has not been reached, the assessed U enrichment will be biased high.

Uranium gamma spectroscopy

- U-235 low energy emissions (143.8, 163.3, 185.7, and 205.3 keV) are easier to shield than U-238 higher energy emissions (258.3, 742.8, 766.4, and 1001.0 keV).
  - If shield and/or self-attenuation corrections are not made for U samples experiencing attenuation, the high energy gamma emissions from U-238 can dominate the spectrum and the assessed U enrichment will be biased low.
- U-232 emissions (238.6, 583.2, 727.3, 860.6, and 2614.5 keV) are commonly found in highly enriched uranium (HEU) gamma-ray spectra.

Typical plutonium type mass and activity percentages

Initial Mass Percentages

Nuclide	Weapons Grade Mass %	Reactor Grade Mass %	Heat Source Mass %
Pu-238	0.010%	1.500%	83.890%
Pu-239	93.771%	58.100%	13.800%
Pu-240	5.999%	24.100%	1.900%
Pu-241	0.200%	11.400%	0.320%
Pu-242	0.020%	4.900%	0.090%
Am-241	0.000%	0.000%	0.000%

Decay Corrected Mass Percentages at 20 Years

Nuclide	Weapons Grade Mass %	Reactor Grade Mass %	Heat Source Mass %
Pu-238	0.009%	1.286%	81.652%
Pu-239	93.783%	58.319%	15.723%
Pu-240	5.991%	24.154%	2.161%
Pu-241	0.076%	4.357%	0.139%
Pu-242	0.020%	4.921%	0.103%
Am-241	0.122%	6.962%	0.222%

Initial Activity Percentages

Nuclide	Weapons Grade Activity %	Reactor Grade Activity %	Heat Source Activity %
Pu-238	0.611%	2.118%	97.664%
Pu-239	20.761%	0.297%	0.058%
Pu-240	4.858%	0.451%	0.029%
Pu-241	73.770%	97.133%	2.248%
Pu-242	0.000%	0.002%	0.000%
Am-241	0.000%	0.000%	0.000%

Decay Corrected Activity Percentages at 20 Years

Nuclide	Weapons Grade Activity %	Reactor Grade Activity %	Heat Source Activity %
Pu-238	0.937%	4.358%	98.828%
Pu-239	37.262%	0.716%	0.069%
Pu-240	8.707%	1.084%	0.035%
Pu-241	50.419%	89.110%	1.014%
Pu-242	0.001%	0.004%	0.000%
Am-241	2.675%	4.727%	0.054%

- A Pu production reactor irradiates U-238 for a short time, then processes it to chemically remove the Pu isotopes.
- The longer the exposure time in the reactor (higher burnup), the more Pu-238, Pu-240, Pu-241, and Pu-242 are produced relative to Pu-239.
- Pu-241 increases more rapidly than Pu-240 as burnup increases.
- Following chemical separation, Am-241 builds-in from the decay of Pu-241 (14.4 y half-life).

Plutonium gamma spectroscopy notes

- The PeakEasy Library “Plutonium” folder contains useful reference spectra (as a function of burn-up, shielding, and detector resolution).

- When reporting Am-241 mass fractions, the convention is to report the mass fraction of Am-241 relative to the total amount of Pu mass present (Am-241 mass / total Pu mass).

- When the measured Pu-239/Pu-240 mass ratio is less than 5, do not simply use the measured Pu-240/(Pu-239+Pu-240) mass fraction to estimate the Pu-240 mass fraction in the plutonium.

  - To better estimate the Pu-240 mass fraction, corrections should be made for the contributions from other Pu isotopes (Pu-238, Pu-241, Pu-242) present.

Plutonium gamma spectroscopy notes

- For unshielded Pu samples, consider the use of graded shielding (Cd-Cu or Sn-Cu) to eliminate/reduce Am-241 59.5 keV gamma emissions in the spectrum.
  - May allow closer sample to detector distances to be used (reducing detection limits and increasing higher energy emission count rates).
  - Reduces “true coincidence” with other low-energy photon emissions.
  - For portable gamma spectroscopy systems, I have used graded shield thicknesses between 30 to 60 mil of Cd or Sn in conjunction with 15 to 30 mil of Cu.

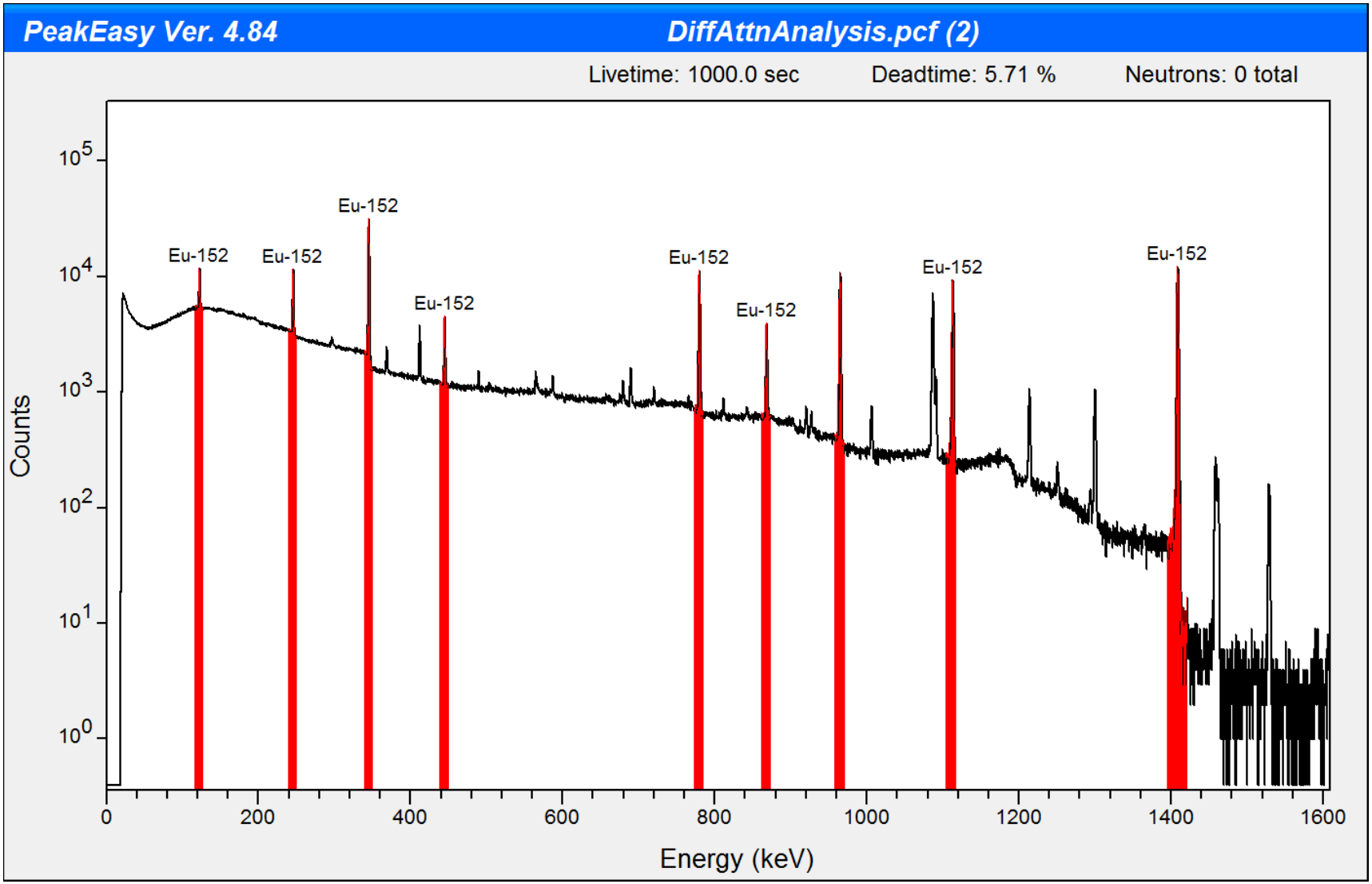
Differential attenuation analysis

- Differential peak analysis utilizes the difference in attenuation suffered by gamma rays of different energies from the same nuclide to estimate the effective matrix density and thickness.

- To assist in understanding the concept of differential attenuation analysis, the following example is presented.

  - A 1000 second measurement with an ORTEC Detective EX-100 is performed on a Eu-152 source of unknown activity in a steel container of unknown thickness at a distance of 25-cm from the detector endcap to the center of the container.

Differential attenuation analysis



Differential attenuation analysis

- Assuming the thickness of steel is 1-cm and 3-cm, respectively, the following activities were calculated.

			1-cm Fe	3-cm Fe
Nuclide	Energy (keV)	Counts	Activity (μCi)	Activity (μCi)
Eu-152	244.7	41,088	23	164
Eu-152	344.3	151,578	25	126
Eu-152	444.0	17,336	27	109
Eu-152	778.9	56,889	27	79
Eu-152	867.4	18,300	28	77
Eu-152	964.1	61,214	29	75
Eu-152	1112.1	54,758	30	74
Eu-152	1408.0	79,696	33	73

- Based on differential attenuation analysis, 1-cm provides too little attenuation, since estimated activities are increasing with energy, and 3-cm provides too much attenuation, since estimated activities are decreasing with energy.

Differential attenuation analysis

- Accordingly, the thickness of steel is optimized until the calculated activities at the different energies are approximately equal.

			1.7-cm Fe
Nuclide	Energy (keV)	Counts	Activity (µCi)
Eu-152	244.7	41,088	46
Eu-152	344.3	151,578	45
Eu-152	444.0	17,336	43
Eu-152	778.9	56,889	39
Eu-152	867.4	18,300	40
Eu-152	964.1	61,214	40
Eu-152	1112.1	54,758	41
Eu-152	1408.0	79,696	44

- Actual Eu-152 activity = 50 µCi, Thickness of the iron container = 1.9-cm

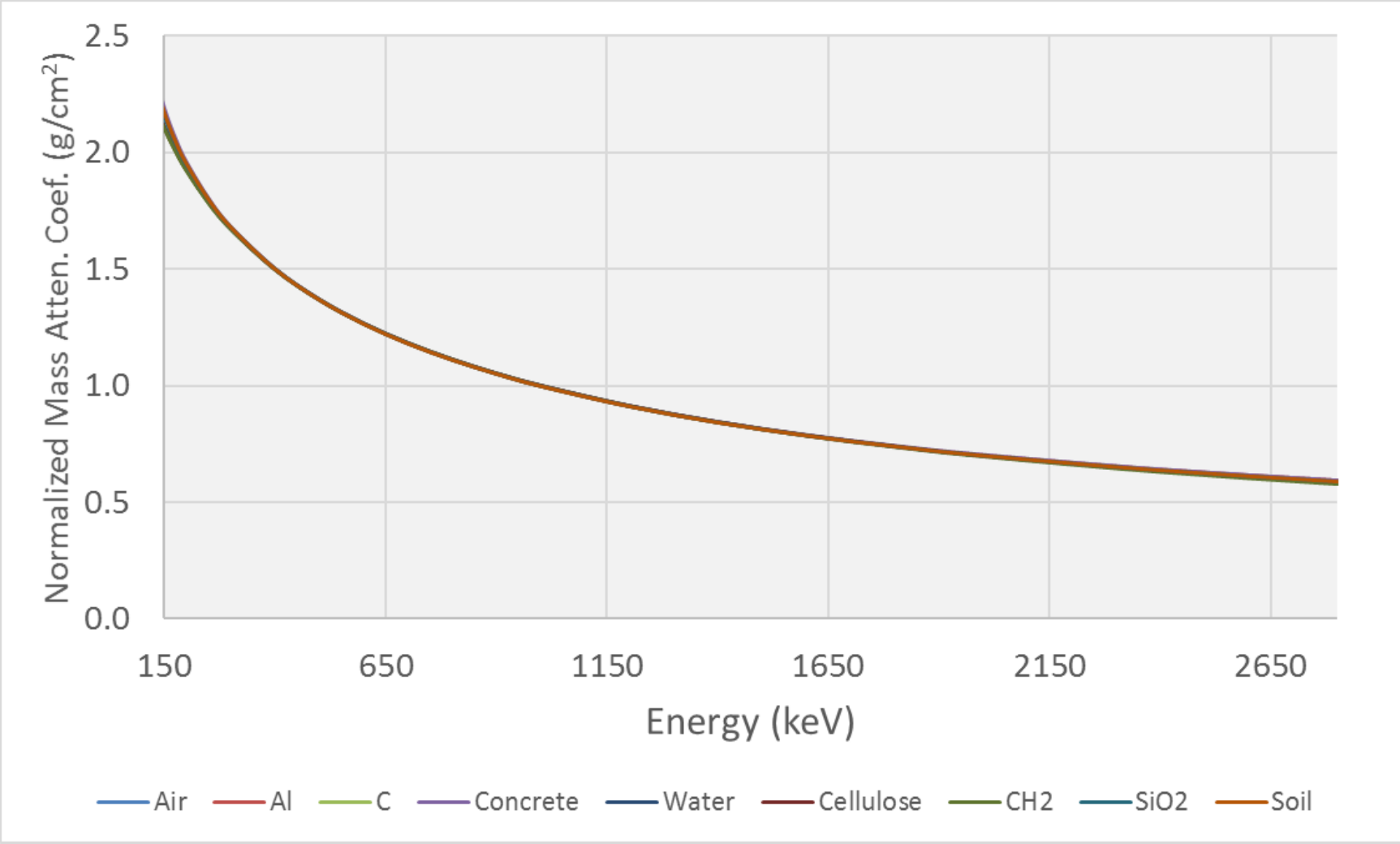
Note: Calculations were performed with SimpleMass.xls, a Los Alamos National Laboratory (LANL) EXCEL and Visual Basic application for full-energy peak analysis of shielded and unshielded point sources.

Differential attenuation analysis

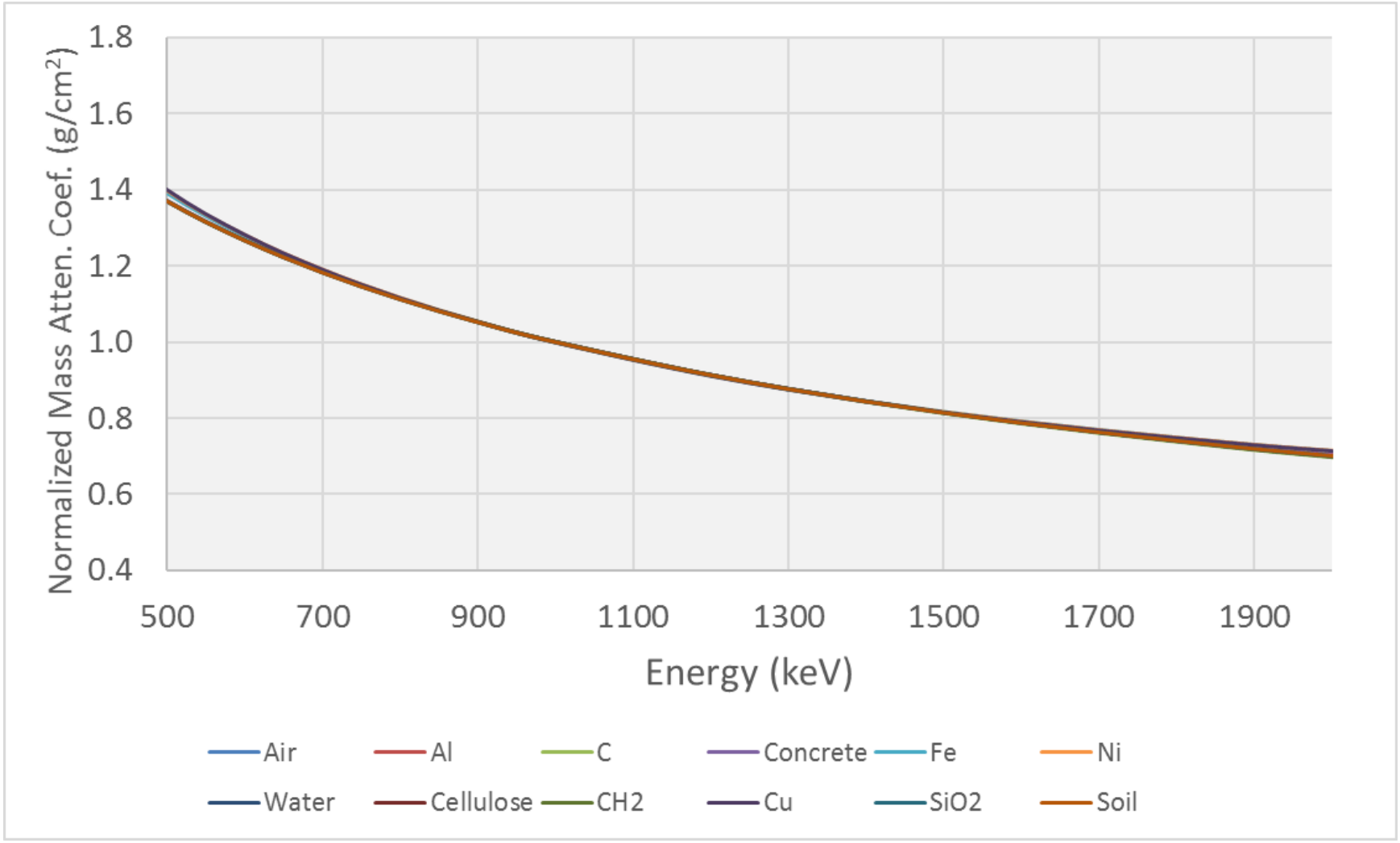
Additional notes

- Normalized mass attenuation coefficients values are nearly independent of material between 150 and 3000 keV for several common materials.
  - Al, concrete, water, cellulose ($C_6H_{10}O_5$), plastics (CH_2), glass (SiO_2), soil, and air.
  - Fe, Cu, and Ni can be added to the list for energies between roughly 500 to 2000 keV.
- In these instances, you can approximate the “right” activity, but not the “right” shield thickness, using the “wrong” material.

Differential attenuation analysis



Differential attenuation analysis



Differential attenuation analysis

- Eu-152 activity estimates performed using the following emissions:
  - 444.0, 778.9, 867.4, 964.1, 1112.1, 1408.0 keV

- Optimized shielding using different Z

Material	Activity Estimate (µCi)	% Error
Fe	45	-10%
Al	49	-2%
Concrete	48	-4%
Water	49	-3%
Cellulose	49	-2%
CH2	50	0%
SiO2	48	-4%
Soil	48	-4%
Air	50	0%
Cu	43	-14%
Ni	45	-10%

References

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