

Automated SNM Analysis for Gamma-Ray Spectra

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Objective

➤ Provide automated spectral analysis tool for SNM:

- Determine isotopic compositions and uncertainty estimates for plutonium and uranium
- Determine effective atomic number and areal density of shielding materials
- Estimate outside radii of equivalent spheres
- Estimate void radius if the material is thin enough to be distinguishable from solid mass

➤ If plutonium is present:

- Estimate the age of the material
- Determine if the material is metallic or oxide based on gamma rays emitted following $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ reaction
- Estimate concentrations of fluorine and beryllium



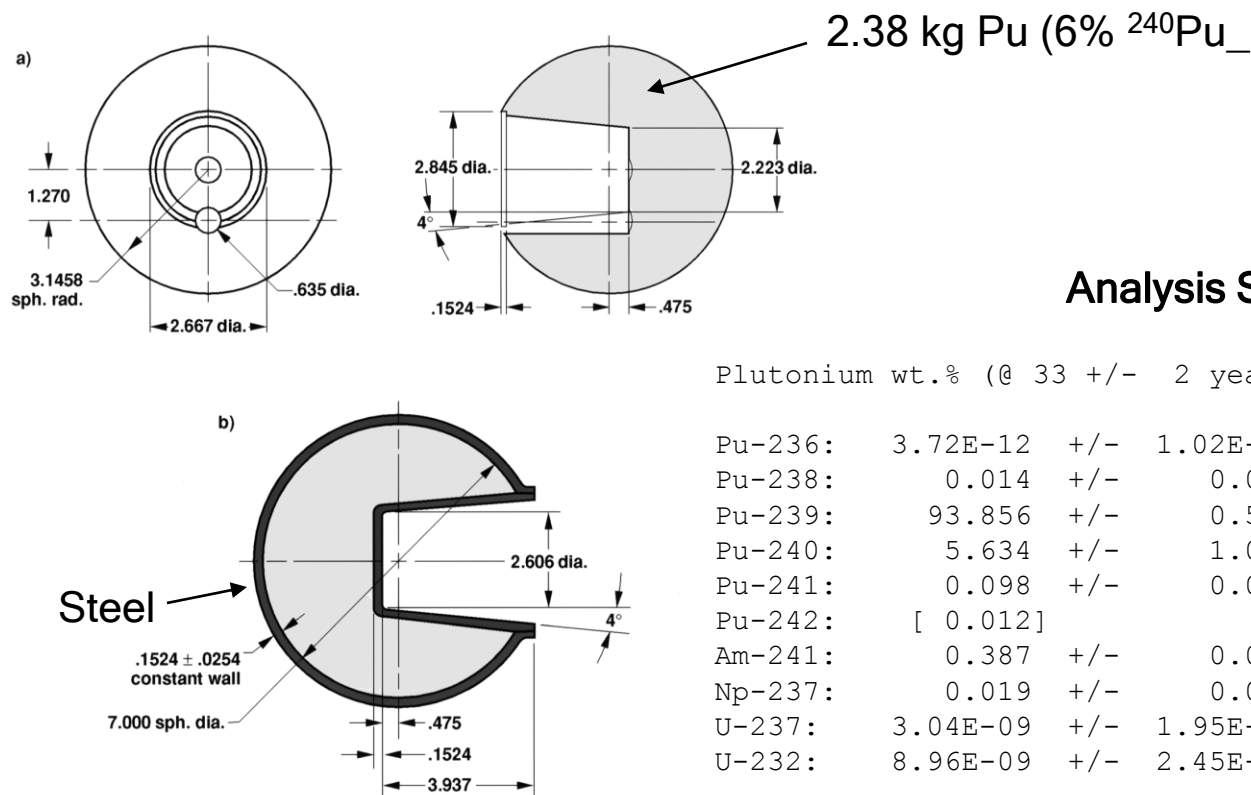
Analysis Method

- **Spectra are analyzed by a full-spectrum analysis method, which includes continuum as well as photopeaks**
 - Matching the entire spectrum utilizes all of the available statistical content
 - Continuum provides information about shielding materials
- **A response function is used to compute spectral shapes associated with gamma rays that are incident on the detector**
 - Photopeaks are represented by skewed Gaussians, and parameters are adjusted based upon the spectrum that is being analyzed
 - The response function also describes radiation that is scattered into the detector
- **Plutonium and uranium are analyzed independently, and both elements may be present in the source**
 - Plutonium and uranium are modeled as spheres with single external material that is characterized by an effective atomic number and areal density
 - Up to 46 parameters are estimated by nonlinear regression

Development History

- **The analysis method derived from the Minimum Mass Method (MME), which was developed in 1999 for international monitoring of SNM**
 - Simple approximation of the detector response and scattering by intervening materials
 - Low-angle scattering reveals areal density
 - Differential attenuation gives atomic number
 - Simplicity facilitated inspection of the code
 - Objectives were to show that the inspected materials were consistent with weapons-grade SNM and that the quantities exceeded a minimum threshold without quantitative mass estimate
- **The MME method evolved into the SNMAnalysis algorithm, which is part of the Gamma Detector Response and Analysis Software (GADRAS)**
 - Accurate response function was applied
 - Modified to derive quantitative information
 - Used by emergency responders at many US national laboratories
 - The SNMAnalysis algorithm is only exposed by GADRAS, but a simplified application could be developed that does not include the additional features that make the application Official Use Only

Analysis of Detective Spectrum for LLNL Pu Ball



Analysis Summary

Plutonium wt.% (@ 33 +/- 2 years) _____ @ t=0 _____

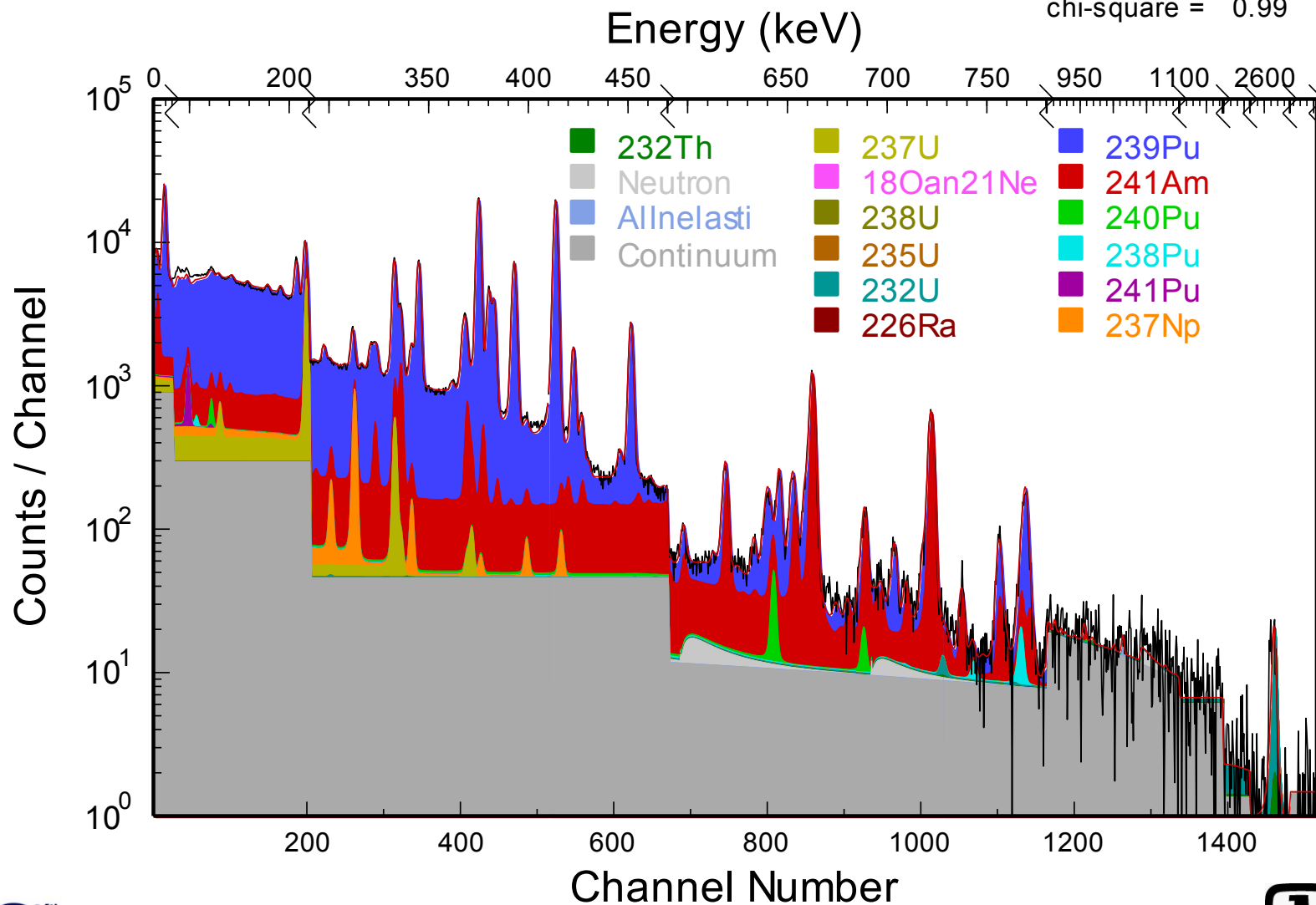
Pu-236:	3.72E-12	+/-	1.02E-12	8.97E-09	+/-	3.57E-09
Pu-238:	0.014	+/-	0.006	0.018	+/-	0.007
Pu-239:	93.856	+/-	0.585	93.952	+/-	0.585
Pu-240:	5.634	+/-	1.018	5.647	+/-	1.020
Pu-241:	0.098	+/-	0.006	0.505	+/-	0.032
Pu-242:	[0.012]			[0.012]		
Am-241:	0.387	+/-	0.007			
Np-237:	0.019	+/-	0.002			
U-237:	3.04E-09	+/-	1.95E-10			
U-232:	8.96E-09	+/-	2.45E-09			

Confidence for measured ^{180}O - α - ^{21}Ne gammas: -0.4 sigma
 Measured (a,n) relative to expected for oxide: -2.2 sigma
 Chemical form: indeterminate
 External shielding: AN = 29 +/- 1; AD = 4.1 +/- 0.1
 Outer radius if plutonium is spherical: 3.8 cm
 Estimated void radius: 2.2 cm
 Estimated mass if delta-phase plutonium: 2.9 kg

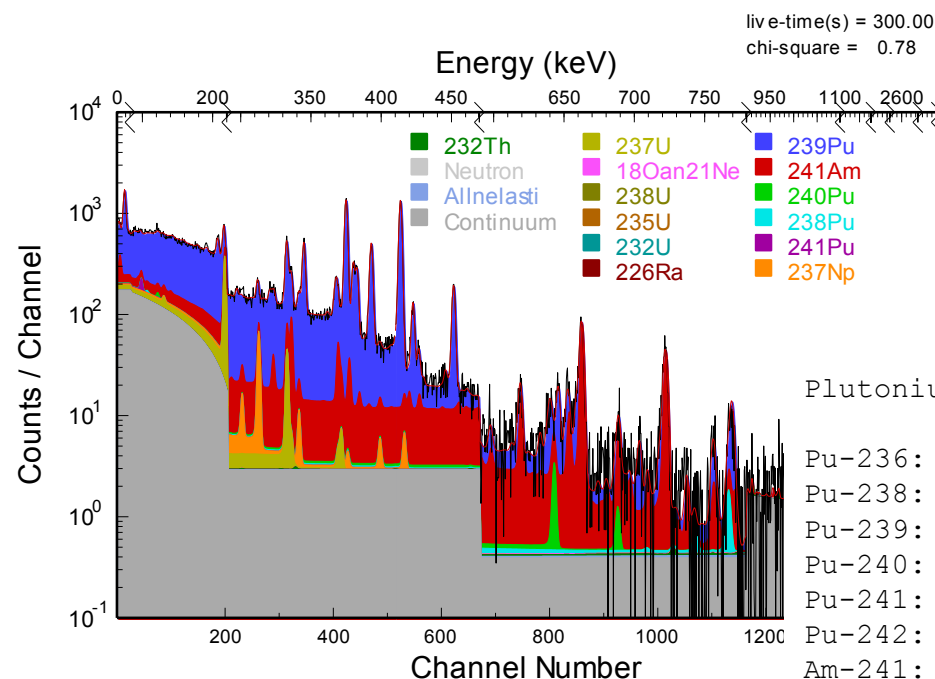
Spectral Display for LLNL Pu Ball

live-time(s) = 2820

chi-square = 0.99



Detective Spectrum for LLNL Pu Ball in PE



2.38 kg Pu, 6% ^{240}Pu

External shielding:

- 1.2 g/cm² Fe
- 4.2 g/cm² PE

Plutonium wt.% (@ 31 +/- 3 years)						@ t=0
Pu-236:	0.00E+00	+/-	8.09E-12	0.00E+00	+/-	1.45E-08
Pu-238:	0.022	+/-	0.022	0.027	+/-	0.028
Pu-239:	93.724	+/-	0.994	93.820	+/-	0.995
Pu-240:	5.759	+/-	3.576	5.766	+/-	3.588
Pu-241:	0.115	+/-	0.011	0.502	+/-	0.113
Pu-242:	[0.012]			[0.012]		
Am-241:	0.369	+/-	0.017			
Np-237:	0.021	+/-	0.004			
U-237:	3.56E-09	+/-	3.44E-10			
U-232:	0.00E+00	+/-	1.07E-08			

Confidence for measured 180-an-21Ne gammas: 0.2 sigma
 Measured (a,n) relative to expected for oxide: -0.1 sigma
 Chemical form: indeterminate
 External shielding: AN = 20 +/- 1; AD = 9.7 +/- 0.2
 Outer radius if plutonium is spherical: 4.1 cm
 Estimated void radius: 3.0 cm
 Estimated mass if delta-phase plutonium: 2.7 kg

High-Burnup Plutonium Oxide

Plutonium wt.% (@ 3 +/- 0 years) _____ @ t=0 _____

Pu-236:	8.47E-07	+/-	1.52E-08	1.80E-06	+/-	1.41E-06
Pu-238:	1.225	+/-	0.031	1.256	+/-	0.032
Pu-239:	60.703	+/-	0.632	60.709	+/-	0.632
Pu-240:	27.639	+/-	0.291	27.648	+/-	0.291
Pu-241:	6.185	+/-	0.139	7.191	+/-	0.165
Pu-242:	[3.246]			[3.246]		
Am-241:	1.001	+/-	0.017			
Np-237:	0.007	+/-	0.003			
U-237:	1.92E-07	+/-	4.31E-09			
U-232:	9.54E-07	+/-	1.71E-08			

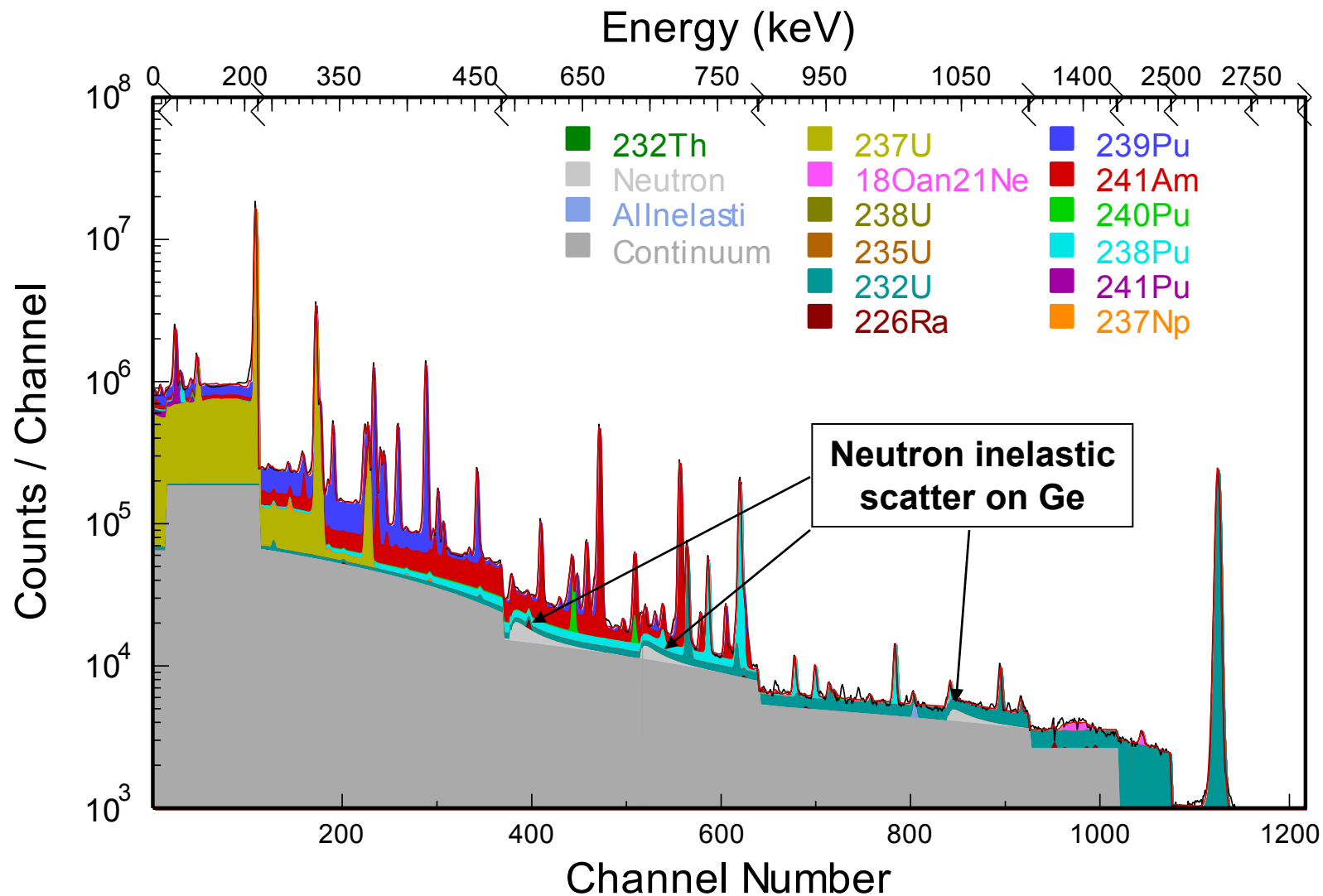
Confidence for measured 18O-an-21Ne gammas: 14.7 sigma
 Measured (a,n) relative to expected for oxide: 1.7 sigma
 Alpha-Fluorine peak: 1273.4 keV @ 6.7 sigma
 Alpha-Beryllium peak: 4438.0 keV @ 99.9 sigma
 Aluminum-inelastic peak: 1014.0 keV @ 25.4 sigma
 Chemical form: OXIDE
 External shielding: AN = 21 +/- 1; AD = 6.5 +/- 0.3
 Outer radius if plutonium is spherical: 8.8 cm
 Estimated mass of PuO2 if 3 g/cc oxide sphere: 8.7 kg
 Estimated fluorine concentration: 18 ppm
 Estimated beryllium concentration: 0 ppm

Actual Source:

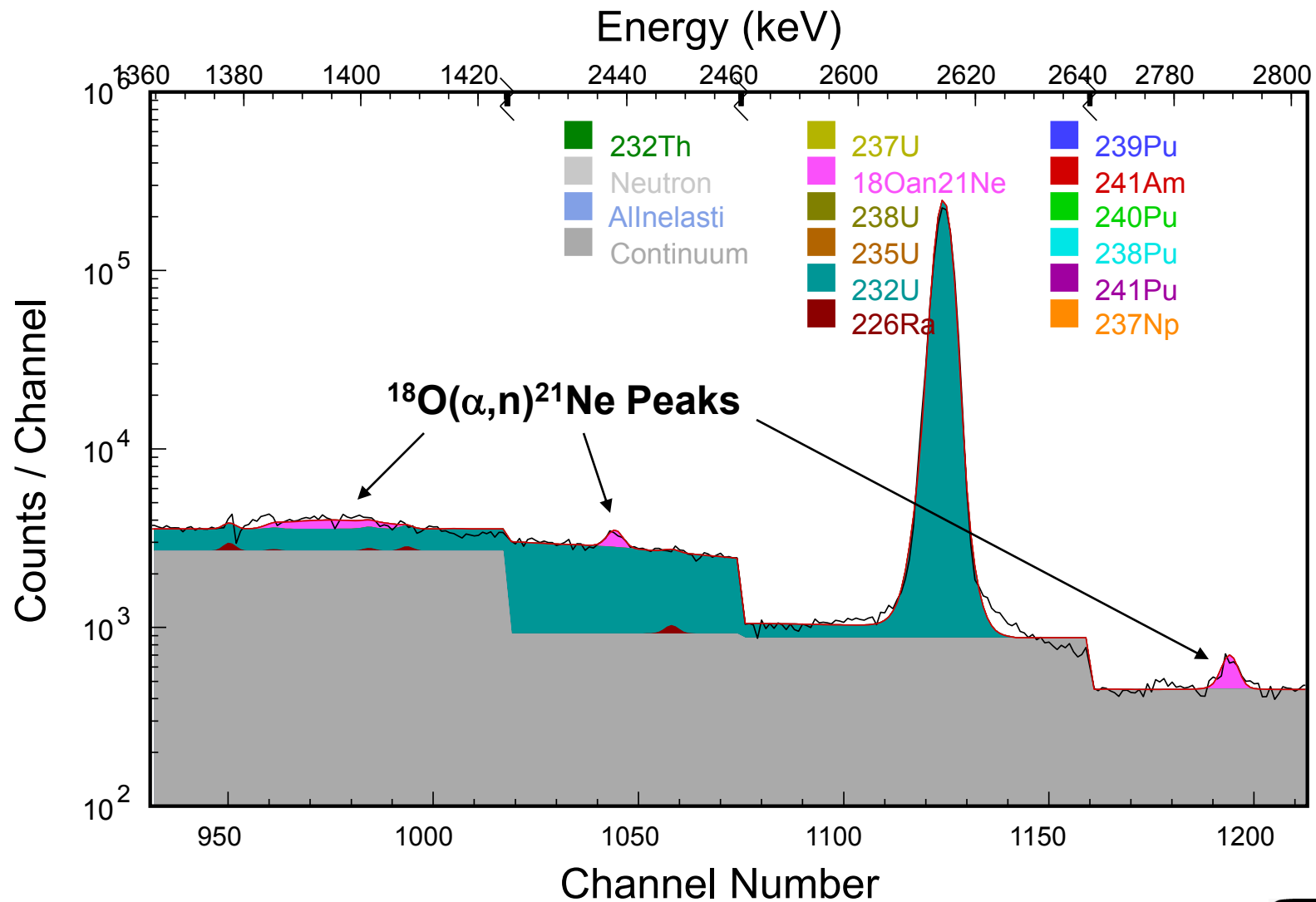
- 3 years old
- 26.8% ²⁴⁰Pu
- 3-mm-thick Fe container
- Oxide
- 7.12 kg PuO2

There is no evidence that uranium is present.

Spectrum for High-Burnup Plutonium Oxide

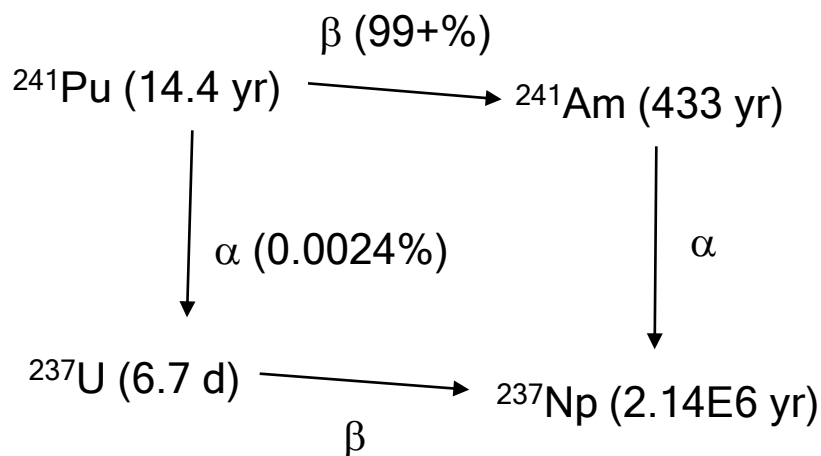
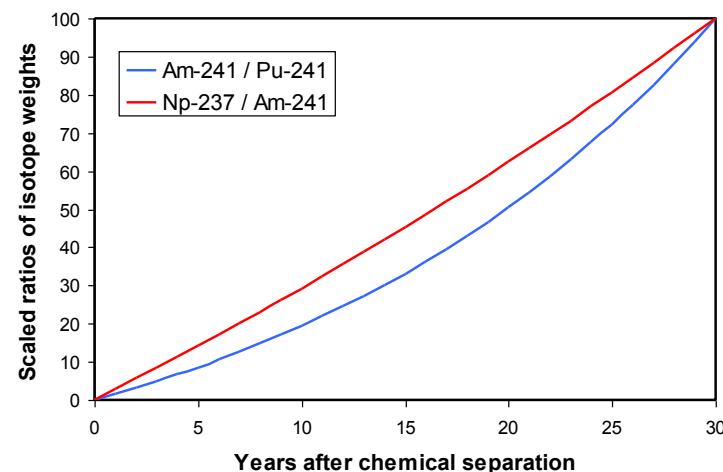
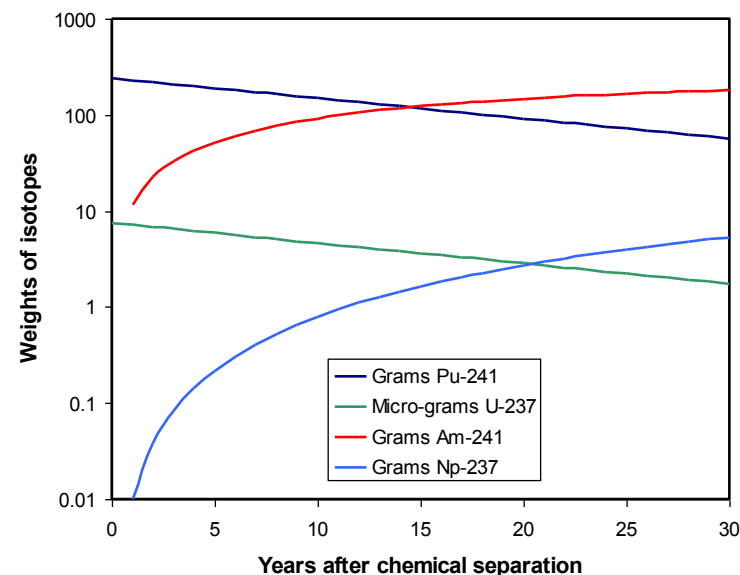


Alpha-Oxygen Region for High-Burnup PuO_2

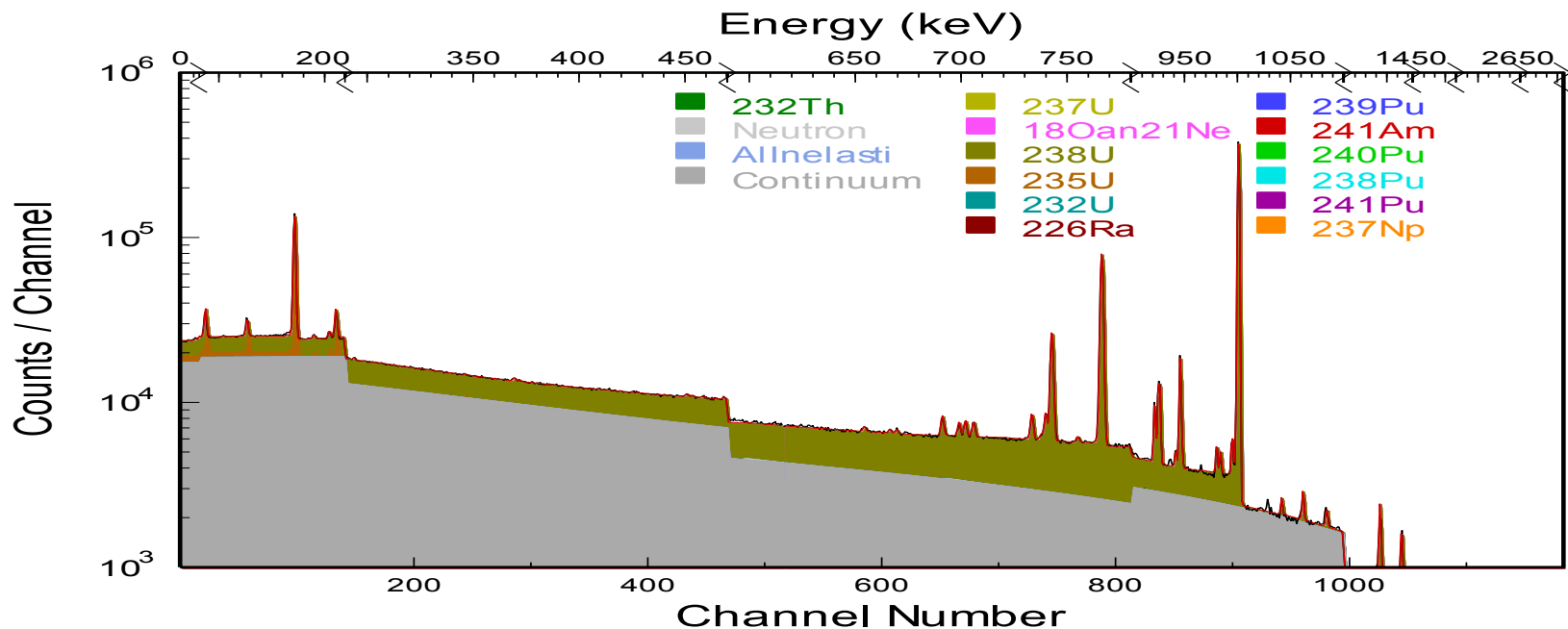


Plutonium Age Estimate

- Plutonium age (time after chemical separation) is estimated
- The ratios of $^{241}\text{Am}:$ ^{241}Pu and $^{237}\text{Np}:$ ^{241}Am are used to estimate age
 - The relative uncertainties in these ratios of the individual estimates are used to combine the age estimates
 - Because of its short half-life, ^{237}U is in equilibrium with ^{241}Pu . Therefore, the ^{237}U activity is used to derive the mass of ^{241}Pu .



Analysis of 7.4-kg Natural Uranium Sphere



Uranium wt. %

U-232:	0.00E+00	+/-	4.63E-12
U-235:	0.900	+/-	0.119
U-238:	99.100	+/-	0.119

Actual: 0.72% ²³⁵U

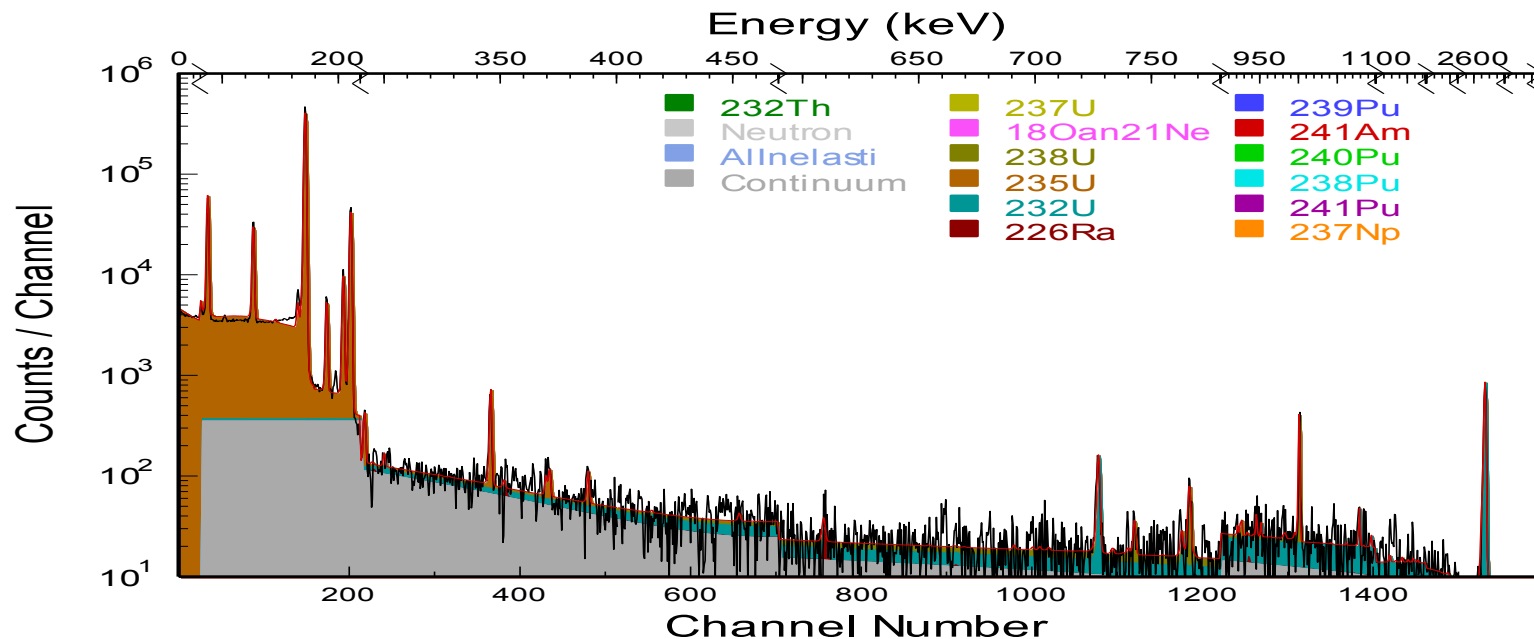
External shielding: AN = 41 +/- 11; AD = 1.2 +/- 1.0

Outer radius if uranium is spherical: 4.8 cm

Estimated void radius: 2.9 cm

Estimated mass if metallic uranium: 6.8 kg

Analysis of 2.1-kg HEU Test Object (with cavity)



Uranium wt. %			
U-232:	8.25E-09	+/-	3.10E-10
U-235:	94.698	+/-	1.438
U-238:	5.302	+/-	1.438

Actual: 93.5% ²³⁵U

External shielding: AN = 94 +/- 13; AD = 0.0 +/- 0.2
 Outer radius if uranium is spherical: 2.6 cm
 Estimated void radius: 1.5 cm
 Estimated mass if metallic uranium: 1.1 kg



Conclusions

- **The SNMAnalysis tool utilizes photopeaks as well as the continuum to estimate isotopic concentrations for plutonium and uranium**
- **Shielding estimates are made as part of the analysis process**
 - This supplemental information is an essential part of the analysis approach
- **Plutonium is classified as metallic or oxide based on the manifold of weak gamma rays that are emitted by alpha interactions on oxygen**
- **Plutonium age estimates are used to compute the initial isotopic composition**
- **The analysis is suitable for SNM that is shielded by unknown materials, provided that characteristic gamma rays are detectable**
- **The only input is the spectrum for a characterized HPGe detector and the source-to-detector distance**