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U/Pu Analysis with Gamma-Ray Spectroscopy

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Categories of Uranium

- Only three isotopes of uranium are found in nature
 - ^{238}U (99.27%)
 - ^{235}U (0.720%)
 - ^{234}U (0.006%)
- Categories of Enrichment ($E = \% \text{ of } ^{235}\text{U}$)
 - 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) $E \geq 20.0 \%$

Definition of ^{235}U Enrichment

Mass Fraction

$$E_{\text{Mass}\%} = 100 \times \frac{{}^{235}\text{U Mass}}{\text{Total U Mass}}$$

Atom Fraction

$$E_{\text{at}\%} = 100 \times \frac{\text{Number of } {}^{235}\text{U atoms}}{\text{Number of U atoms}}$$

**Approximate
relationship
between
expressions**

$$E_{\text{Mass}\%} = \frac{(235/238) \times E_{\text{at}\%}}{1 - (3/238) \times E_{\text{at}\%}} \approx \frac{235}{238} \times E_{\text{at}\%}$$

Uranium Gamma-Ray Spectrum (HPGe)

50.5% enrichment (HEU)

Pu Isotopes Accumulate with Increasing Reactor Burnup

As neutrons in the reactor create and interact with Pu, the Pu isotopic distribution changes.

Pu Isotopic Composition Range

	Low Burnup ————→ High Burnup			
^{238}Pu	0.02	0.3	0.8	1.6
^{239}Pu	93.5	82.5	73.3	57.3
^{240}Pu	6.2	13.8	18.3	25.0
$^{241}\text{Pu} + ^{241}\text{Am}$	0.3	3.8	6.6	11.7
^{242}Pu	0.04	0.8	2.1	5.6

IAEA

Definitions:

- Weapons-Grade $<7\% ^{240}\text{Pu}$ or $>93\% ^{239}\text{Pu}$

- Reactor-Grade $>7\% ^{240}\text{Pu}$ or $<93\% ^{239}\text{Pu}$

Plutonium γ -ray Spectrum

Shielding can change this picture dramatically!

Peak Intensity Expression

$$C_i(E_n) = \lambda_i N_i BR_{i,n} \varepsilon(E_n)$$

- $C_i(E_n)$ – the intensity (area) of the peak at energy E_n for isotope i
- λ_i – the decay constant of isotope i $\lambda_i = \ln 2 / \tau_i$
- N_i – the quantity of isotope i (in atoms)
- $BR_{i,n}$ – the branching ratio for isotope i of the peak at energy E_n
- $\varepsilon(E_n)$ – total efficiency for photopeak detection of a gamma ray with energy E_n . This includes detector efficiency, geometry (solid angle), item self-absorption, and attenuation in packaging and materials between item and detector.

Next: solve equation for N and then take ratio for two isotopes N_i/N_k

Relative Isotopic Ratios

$$f_k^i = \frac{N_i}{N_k} = \frac{C_i(E_n)}{C_k(E_m)} \frac{t_{1/2i}}{t_{1/2k}} \frac{BR_{k,m}}{BR_{i,n}} \frac{RE(E_m)}{RE(E_n)}$$

Desired
Answer

Peak
Analysis

Nuclear
References

Nuclear
References

C/BR
Analysis

All these values are obtained from the spectrum itself or from standard nuclear references; no calibration is necessary!

Determination of Counts

- $C_i(E_n)/C_k(E_m)$ is determined using standard spectral analysis techniques
 - $C(E)$ is a net area - proper background subtraction is critical
 - Peaks are often unresolved or coenergetic; peak fitting methods are required
- The two energies chosen to measure the two isotopes should be as close as possible

High Resolution is Essential

Note: this technique was originally developed for Pu isotopics measurements.

Compton Continuum

Gamma-Ray Peak Fitting

Overlapping Peaks

Uranium X-ray Region

The low energy analysis region of both Pu and U spectrum is very complex with many overlapping X-ray and gamma ray peaks.

Plutonium X-ray Region

Advanced algorithms are used to deconvolve regions where peaks are overlapping.

The net area under each red peak is the counts from a gamma-ray from a specific isotope at that energy. (The $C(E)$ values)

Relative Isotopic Ratios

We now know
how to
determine this

How do we
get this?

$$f_k^i = \frac{N_i}{N_k} = \frac{C_i(E_n)}{C_k(E_m)} \frac{t_{1/2i}}{t_{1/2k}} \frac{BR_{k,m}}{BR_{i,n}} \frac{RE(E_m)}{RE(E_n)}$$

Desired
Answer

Peak
Analysis

Nuclear
References

Nuclear
References

C/BR
Analysis

Relative Efficiency Determination

- $RE(E)$ is determined from the spectrum itself
- A Relative Efficiency curve can be created if you have gamma rays from the same isotope spread over a wide energy range
- If the isotopes are homogeneous, you can combine data from multiple isotopes to make a more complete Relative Efficiency curve.

Efficiency

Definition:
$$\text{Efficiency} = \frac{\gamma\text{-rays detected}}{\gamma\text{-rays emitted in item}}$$

General expression for peak area count rate

$$C = N \lambda R_b \varepsilon$$

C = count rate in peak

N = number of radioactive atoms

λ = radioactive decay constant
 $= \ln 2 / T^{1/2}$

Rearrange:

$$\varepsilon = \frac{1}{N \lambda} \frac{C}{R_b}$$

R_b = branching ratio

ε = efficiency

Relative efficiency is proportional to count rate/branching ratio

$$\varepsilon \propto \frac{C}{R_b}$$

The Efficiency Curve

- Efficiency is a function of γ -ray energy
- Imagine spectrum produced by 100% efficient detector of an isotope with 6 γ -rays with equivalent branching ratios
- In reality the relative sizes of the observed γ -ray peaks would differ

The Relative Efficiency Curve

- The Relative Efficiency Curve is determined by
 - Detector absolute efficiency over the energy range
 - The effect of U/Pu self absorption
 - The effect of container shielding
 - Detector filtering
- Since the formula we are using corrects for the Relative Efficiency curve, none of these factors will affect our results
- Ratio-based programs calculate the C/BR ratios and fit that data to a Relative Efficiency Curve model

The Relative Efficiency Curve

- The fitted curve is used to calculate the RE values for the gamma rays used to calculate the isotopic ratios
- A good Relative Efficiency curve fit is essential to the Ratio Based measurement technique
- The data and the models can be used to draw conclusions about the measurement configuration
 - U/Pu density
 - Absorber thicknesses

Relative Efficiency Curve Components

Relative Efficiency Depends on Detector

Relative efficiency for Sample U0446 (200g), no absorber

Curves normalized so that maxima above the K-edge of Uranium are unity.

Relative Efficiency Depends on External Attenuation

Relative Efficiency Depends on Item Characteristics

- = Low E γ Rays
- = High E γ Rays

Independence of Analysis

- Using **relative** efficiency and isotope **ratios** makes analysis independent of:
 - **Item Characteristics**
 - Size
 - Shape
 - Physical and Chemical Composition
 - Packaging
 - Filtering
 - **Data Acquisition Limitations**
 - Pulse Pile-up
 - Deadtime

NO CALIBRATION IS NEEDED!

Isotopic Homogeneity

- The isotopic composition must be uniform throughout the item, even if it is chemically or physically non-uniform.
 - One exception is that FRAM can analyze physically uniform, but isotopically heterogeneous, matrices of plutonium and ^{241}Am .

Isotopic Homogeneity:

Metal 80% Pu239 15% Pu240
Powder 80% Pu239 15% Pu240

Isotopic Heterogeneity:

Powder 80% Pu239 15% Pu240
Powder 50% Pu239 45% Pu240

The Exception: Pu-metal in Am241 salt

Peak Ratios to Isotopic Composition

Let's define the isotopic fraction for isotope i as

$$F_i = \frac{N_i}{N_T}$$

where

N_i – the number of atoms of isotope i

N_T – total number of atoms for all isotopes of that atomic number.

The sum of the isotopic fractions must be 1:

$$\sum F_i = 1$$

Uranium Enrichment

For the U isotopes of interest:

$$1 = F_{234} + F_{235} + F_{238}$$

Divide by F_{238} :

$$\frac{1}{F_{238}} = \frac{F_{234}}{F_{238}} + \frac{F_{235}}{F_{238}} + 1 = f_{238}^{234} + f_{238}^{235} + 1$$

Invert:

$$F_{238} = \frac{1}{f_{238}^{234} + f_{238}^{235} + 1}$$

Solve for F_{234} and F_{235} :

$$F_{234} = f_{238}^{234} F_{238} \quad F_{235} = f_{238}^{235} F_{238}$$

Uranium Gamma-Ray Spectrum

Performance

- Typical x-ray region precision: ^{235}U to $\pm 2\%$ in 300 s.
- For MOX, use plutonium isotopic composition analysis
- Container wall thickness must be less than about 10 mm steel for x-ray region analysis
- For thicker steel walls analyse using higher energy gamma rays
- ^{238}U - $^{234\text{m}}\text{Pa}$ equilibrium required – uranium must be at least 6 months old.

Plutonium Isotopic Composition

For the five Pu isotopes of interest:

$$1 = F_{238} + F_{239} + F_{240} + F_{241} + F_{242}$$

Divide by F_{239} :

$$\frac{1}{F_{239}} = \frac{F_{238}}{F_{239}} + 1 + \frac{F_{240}}{F_{239}} + \frac{F_{241}}{F_{239}} + \frac{F_{242}}{F_{239}} = f_{239}^{238} + 1 + f_{239}^{240} + f_{239}^{241} + f_{239}^{242}$$

Invert:

$$F_{239} = \frac{1}{f_{239}^{238} + 1 + f_{239}^{240} + f_{239}^{241} + f_{239}^{242}}$$

Plutonium Isotopic Composition

Next compute the other isotopic fractions:

$$F_{238} = f^{238} \cdot F_{239}$$

$$F_{240} = f^{240} \cdot F_{239}$$

$$F_{241} = f^{241} \cdot F_{239}$$

There are **no** measureable ^{242}Pu gamma-rays: for ^{242}Pu , use declared value or compute:

$$F_{242} = A \left[(F_{238})^b + (F_{239})^c + (F_{240})^d + (F_{241} + F_{241}^{\text{Am}})^e \right]$$

where A , b , c , d , and e are determined empirically.

Renormalize, if needed, so all isotopic fractions sum to unity. Iterate as necessary.

Plutonium Analysis Regions of FRAM

FRAM uses four overlapping analytical regions

Why Analyze the 94-104 keV Region

^{240}Pu Peak

Intensity:

Compare the intensity of the peak at 104 keV with that of the one at 160 keV

^{242}Pu Estimate by Correlation

- Determine relative content of ^{242}Pu from a correlation function based on the fractions of the other plutonium isotopes:

$$^{242}\text{Pu} = 53[^{240}\text{Pu}] \times [^{241}\text{Pu}] / [^{239}\text{Pu}]^2$$

Gunnink, Nucl. Matl. Mgmt. 9, (2), 83-93 (1980)

Typical form of correlation functions used is:

$$^{242}\text{Pu} = A \times [(^{238}\text{Pu})^B \times (^{239}\text{Pu})^C \times (^{240}\text{Pu})^D \times (^{241}\text{Pu} + ^{241}\text{Am})^E]$$

FRAM Isotopic Analysis Software

- FRAM is a powerful isotopic analysis code that utilizes the same underlying theory as the Multi-Group Analysis (MGA) code by LLNL.
- Fixed-energy Response-function Analysis with Multiple efficiencies (FRAM)
- Self-calibration using several gamma-ray peaks
- Analyze gamma ray data from 30keV to >1MeV of HPGe planar, HPGe Coaxial, or CdTe detector
- The key difference between FRAM and the MGA family of codes is the use of **user-editable analysis parameters** in FRAM.

Display Results

PC FRAM (5.1) Isotopic Analysis 2013-03-04 09:37:16
(Fixed energy Response function Analysis with Multiple efficiencies)
Operator ID:

spectrum source: C:\FRAMdata\Spectra\FRAM.chn
spectrum date: 1993-10-09 20:39:14
live time: 5714 s
true time: 7200 s
num channels: 8192

parameter set: Pu_Cx_120-460 (2011.05.11 18:25)
Pu Coax, Equilibrium, 120-460keV
Physical Efficiency, Gain 0.125 keV/ch, Offset 0 keV

comment:

diagnostics passed.

(By Corr) %Am241/Pu

	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
mass%	0.0116	93.3419	6.4562	0.1571	0.0332	0.1683
sigma	0.0004	0.0630	0.0631	0.0002	0.0007	0.0012
%RSD	3.55%	0.07%	0.98%	0.13%	2.06%	0.69%
%TotPwr	2.60	71.42	18.14	0.21	< .01	7.62
Specific Power (W/gPu):	(	2.5208 +/-	0.0053)e-003	(	0.21%)	
Effective Pu240 fraction:	(	6.5412 +/-	0.0631)e-002	(	0.97%)	
Time since chemical separation:	5558.8 +/-	27.3	days	(	0.49%)	
Relative mass (U235 / Pu):	1.519e-004	(	>99.99%)			
Relative mass (Np237 / Pu):	2.410e-005	(	2.02%)			
Relative mass (Am243 / Pu):	7.936e-010	(	>99.99%)

Display Efficiency curve

Display Fits of regions

Problems to Avoid when using the Ratio Method

- Resolution is critical in the x-ray region
 - Keep the dead time below 50%
 - Maintain a good pole zero
- Protect the detector from other materials
 - Shielding (collimator)
 - Distance (several meters at the least)
- Ensure the item is
 - Homogenous (same isotopes throughout)
 - For uranium, aged for 6 months (or proper separation date is entered in FRAM)
- For plutonium, ensure the correct amount of filter is used to sufficiently attenuate the 60 keV peak from ^{241}Am .