Radiochemistry Webinars
Nuclear Materials Analysis—Nondestructive Analysis

In Cooperation with our University Partners
Meet the Presenter...

Dr. Azaree Lintereur is an Assistant Professor for the Nuclear Engineering Program at the University of Utah, which she joined in August 2014. Prior to joining the university, Dr. Lintereur was a postdoctoral research associate at Pacific Northwest National Laboratory (PNNL) in the Radiation Detection and Nuclear Sciences group. Her work at PNNL emphasized radiation detector development for nuclear safeguards and homeland security applications. She specialized in $^3$He alternatives for neutron detection, with a focus on coincidence and multiplicity counter design. While at PNNL she was supported by the Next Generation Safeguards Initiative and completed the summer Intensive International Safeguards course. In her current position, she is developing a radiation detection laboratory and research group. Her group, which consists of both graduate and undergraduate students, is working on a variety of detection projects ranging from muon detection to dual modality calorimetry-gamma ray spectroscopy system design. These projects are supported by DOE and national laboratories. She also continues to consult on multiplicity counter development for PNNL. In addition to research, Dr. Lintereur teaches for the Nuclear Engineering Program. She has updated the graduate reactor physics course, developed a graduate nuclear instrumentation course, and co-taught the new graduate course in nuclear safeguards and nonproliferation. She has also formed an Institute of Nuclear Materials Management (INMM) student chapter, for which she serves as the faculty advisor.

Contact Information: azaree.lintereur@utah.edu
Introduction

• The measurement of nuclear materials is a key component in:
  – Nuclear nonproliferation
  – Nuclear security
  – Nuclear safeguards
  – Detection of nuclear material trafficking

• The measurement options include:
  – Nondestructive analysis
  – Destructive analysis
Forensics Process

Classification

Sampling → Categorization

Hypothesis → Analysis

Interpretation → Confidence
You are given an unmarked canister of unknown origin, do you:

A: Open the canister and take a sample for analysis

B: Analyze the canister without opening it and use that information to select the next set of analysis techniques

Starting with classification
Typically DA techniques

Starting with characterization
Typically NDA techniques
Nondestructive Analysis (NDA) vs. Destructive Analysis (DA)

**DA**
- Typically measures part of a sample
- Often slower
- Normally requires a fixed laboratory
- More accurate

**NDA**
- Can measure an entire sample
- Often portable
- Some measurements can be real-time
- Less accurate
DA Measurements

DA alters the sample during the measurement

DA includes techniques such as:

- Liquid scintillation counting
- Mass spectrometry
NDA Techniques – the sample is not altered
(not an exhaustive list)

Physical Properties
- Mass
- Temperature
- Density
- Pressure

Visual Inspection
- Counting
- Images
- Appearance

Nuclear Properties
- Gamma Rays
- Neutrons
- Passive
- Active
Nuclear NDA

- The quantitative or qualitative determination of the kinds and/or amount of nuclear material in a sample without alteration or invasion of the sample

- Includes
  - Gamma ray spectroscopy
  - Neutron detection
  - Coincidence or multiplicity counting
  - Calorimetry
  - Densitometry
  - X-ray fluorescence
Nuclear NDA Signatures

- Gamma rays*
- Alpha particles
- Beta particles
- Neutrons
  - Single events
  - Coincidence events
  - Triples and higher
- Fissionable

*In some cases the signature that we will be discussing will technically be x-rays, the slides will note when x-rays are the signature of interest.
Challenges

- Uniformity
- Sampling
- Matrix
- Background
- Measurement environments
- Reliable equipment
- Sensitive
- Accurate
- Versatile
- Time
NDA Information

**Gamma Ray**
- Energy
- Isotopic composition
- Electromagnetic radiation absorption
- Challenges with dense and non-uniform samples

**Neutron**
- Counting
- Temporally correlated events
- Active assay
- Minimal absorption even in large or non-uniform samples
- Hydrogenous materials affect the results

**Heat**
- Calorimetry
Spectral Features - *what are you looking for?*

If the energy of the gamma ray emission(s) are accurately recorded the isotope(s) present can be identified.

Source: [Knoll, 2010]
Spectral Features - *when reality hits*

A-Full-energy photopeak (photoelectric effect)

B-Compton continuum (Compton scattering)

C-Compton edge (maximum Compton scatter energy deposition)

D-Compton valley (multiple Compton scatters)

E-Backscatter peak

F-Excess energy region (high energy background gammas)

G-Low energy rise (electronic noise)

*For photons above 1022 keV, there may be single and double escape peaks as well*
Gamma Ray Detector Parameters of Interest

• What are some of the critical detector properties to maximize the information that can be obtained?

  – **Efficiency**
    • Absolute efficiency:
      \[
      \text{Efficiency} = \frac{\text{number of pulses recorded}}{\text{number of quanta emitted by the source}}
      \]
    • Intrinsic efficiency:
      \[
      \text{Intrinsic Efficiency} = \frac{\text{number of pulses recorded}}{\text{number of quanta incident on the detector}}
      \]

  – **Interaction probabilities**
    • Photoelectric effect \( \sim Z^{4-5} \)
    • Compton scattering \( \sim Z \)
    • Pair production \( \sim Z^2 \)

  – **Energy Resolution**
    • Full Width at Half Maximum (FWHM) of the peak centroid
      \( \text{FWHM} = 2.35 \sigma \)
Basic Gamma Ray Detector Categories

Scintillators
- Organic
- Inorganic

Semiconductors
- Cooled
- Wide Band Gap

[Source: kicp.uchicago.edu/research/projects/cogent.html]
[Source: M. Bliss, PNNL]
[Source: www.Beijing-energy.com]
[Source: www.kromek.com]
Scintillation Detectors

- Scintillators produce light in response to ionizing radiation interactions
  - Solids, liquids, or gases

- Basics:
  - Energy deposited by incident radiation produces transitions in molecule energy levels or electron states
  - After de-excitation the scintillator emits a photon in the visible light range
  - The emitted light is guided to a photomultiplier tube (PMT) where it interacts with the photocathode and releases electrons
  - The electrons move through the PMT as the result of an electric field
  - The electrons interact with the PMT dynodes (which emit secondary electrons)
  - The secondary electrons released by the dynodes are propagated towards subsequent dynodes, amplifying the original signal
  - The final amplification is on the order of $10^6$
Scintillation Detectors

- Gamma ray
- NaI
- Cathode
- Light tight enclosure/reflector
- Scintillation light
- Photocathode
- Focusing electrodes
- Dynodes
- Secondary electrons
- Primary electrons
- Optical coupling
- Anode
Semiconductor Detectors

- Ionizing radiation interactions produce charges which are collected to generate a signal

- Basics
  - A charged particle (secondary or primary) passing through a semiconductor generates electron-hole pairs (energy required order of eV/ion pair)
  - Charges move under the influence of an applied electric field
  - The charges are “collected”
  - Associated circuitry can be configured to operate in current or pulse mode

- Leakage current can be an issue (due to low energy required for charge pair generation)
  - Some semiconductors need to be cooled (77 K) to reduce leakage
  - Wide band gap materials (i.e., CZT) can be operated at room temperature

- Fewer signal conversions which improves energy resolution
Semiconductor Detectors

Diagram:
- Semiconductor
- Primary radiation
- e (electron)
- h (hole)
- n and p (regions)
- HV (High Voltage)
- C (capacitor)
- R (resistor)
- Ground
- Signal output
Things Aren’t Always Clear

The detector you use will influence the information obtained

[Source: Walt Hensley, PNNL, from PNNL-SA-86636]
## Gamma Ray Emissions

<table>
<thead>
<tr>
<th>Region</th>
<th>$^{238}\text{Pu}$</th>
<th>$^{239}\text{Pu}$</th>
<th>$^{240}\text{Pu}$</th>
<th>$^{241}\text{Pu}$</th>
<th>$^{241}\text{Am}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>keV</td>
<td>keV</td>
<td>γ/s*g</td>
<td>keV</td>
<td>γ/s*g</td>
<td>keV</td>
</tr>
<tr>
<td>40-60</td>
<td>43.5</td>
<td>2.5x10^8</td>
<td>51.6</td>
<td>6.2x10^5</td>
<td>45.2</td>
</tr>
<tr>
<td>90-105</td>
<td>99.9</td>
<td>4.6x10^7</td>
<td>98.8</td>
<td>2.8x10^4</td>
<td>104.2</td>
</tr>
<tr>
<td>120-450</td>
<td>152.7</td>
<td>6.1x10^6</td>
<td>129.3</td>
<td>1.4x10^5</td>
<td>160.3</td>
</tr>
<tr>
<td></td>
<td>203.5</td>
<td>1.3x10^4</td>
<td></td>
<td></td>
<td>164.6*</td>
</tr>
<tr>
<td></td>
<td>345.0</td>
<td>1.3x10^4</td>
<td></td>
<td></td>
<td>208.0*</td>
</tr>
<tr>
<td></td>
<td>375.0</td>
<td>3.6x10^4</td>
<td></td>
<td></td>
<td>332.4*</td>
</tr>
<tr>
<td></td>
<td>413.7</td>
<td>3.4x10^4</td>
<td></td>
<td></td>
<td>370.9*</td>
</tr>
<tr>
<td>450-800</td>
<td>766.4</td>
<td>1.4x10^5</td>
<td>646.0</td>
<td>3.4x10^2</td>
<td>642.5</td>
</tr>
<tr>
<td></td>
<td>717.7</td>
<td>6.3x10^1</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Resolving these peaks is not trivial

Data from PANDA Chapter 8
Isotopic Information

Gamma ray spectrum (Ge detector), 530 g Pu as PuO$_2$ (weight percent $^{238}\text{Pu}$ 0.202%, $^{239}\text{Pu}$ 82.49%, $^{240}\text{Pu}$ 13.75%, $^{241}\text{Pu}$ 2.69%, $^{242}\text{Pu}$ 0.76%, $^{241}\text{Am}$ 11800 µg/g Pu)
Information Extraction

• Identification Algorithms
  – Peak Fitting
    • Requires a characterized detector
    • Background subtraction
    • Typically relies on algorithms to identify peaks
      • Derivative peak searches
      • Cross-correlation
    • Low resolution detectors can make peak identification challenging
  – Spectral Fitting – Library Directed
    • Requires a library of source templates to which spectra can be compared
    • Libraries typically don’t have all options
Challenges

• Detector parameters

• Time
  – Often trying to make a measurement in seconds, not hours or days

• Distance
  – Not only will the signal be weakened with distance, but geographical features can further affect results

• Shielding
  – Can produce non-linear effects
  – Non-uniform samples

[Source: Doyle]
Gamma Ray Spectroscopy Instrumentation

- Battery-powered
- Computer-based
- Data acquisition
- Data analysis
- Often very specialized

[Source: Safeguards Techniques and Equipment]
[Source: Zendel]
[Source: Chichester]
NDA Information

**Gamma Ray**
- Energy
- Isotopic composition
- Electromagnetic radiation absorption
- Challenges with dense and non-uniform samples

**Neutron**
- Counting
- Temporally correlated events
- Active assay
- Minimal absorption even in large or non-uniform samples
- Hydrogenous materials affect the results

**Heat**
- Calorimetry
What About Neutrons?

• High emissions (per unit mass)
  – Can detect small quantities

• Minimal background

• There is a lot of information in neutron distributions

• Highly penetrating (appropriate for large or dense samples)

• All you have to do is count....
  – “simple” detection process

• Candidates for imaging because they scatter in certain materials

• Spectroscopy is possible, but very challenging
  – Contrast to gamma ray detection, information from neutrons is (typically) generated by counting, not energy
Neutron Interactions

- **Scattering**
  - Elastic: (n,n)
  - Inelastic: (n,n')

- **Absorption**
  - Electro-Magnetic: (n,γ)
  - Neutral: (n,*n) *=1, 2, ...
  - Charged: (n,p), (n,α) etc.
  - Fission: (n,f)
The Basic (Capture) Options

<table>
<thead>
<tr>
<th></th>
<th>(^3)He</th>
<th>(^{10})B</th>
<th>(^6)Li</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cross Section (b)</td>
<td>5330</td>
<td>3840</td>
<td>940</td>
</tr>
<tr>
<td>Q Value (MeV)</td>
<td>0.76</td>
<td>2.31</td>
<td>4.78</td>
</tr>
<tr>
<td>Gamma Ray</td>
<td>&gt;10(^{-6})</td>
<td>&gt;10(^{-6})</td>
<td>&gt;10(^{-6}) (can depend on PSD)</td>
</tr>
<tr>
<td>Rejection</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Other</td>
<td>Safe</td>
<td>BF(_3) Corrosive/Toxic</td>
<td>Typically in a matrix (i.e., (^6)LiF) for stability</td>
</tr>
</tbody>
</table>

[Source: PNNL-SA-111028]
Is Counting Neutrons Enough?

Reality Strikes – nuclear materials emit neutrons from 3 different processes:

- **Spontaneous Fission**
- **Induced Fission (Multiplication)**
- **\((\alpha, n)\) Reactions (with low-Z elements e.g., oxygen)**

Challenge: How do you deconvolve mass, multiplication and \((\alpha, n)\) production?
The Neutron Assay Goal

Determine the parameters of an unknown SNM sample by characterizing its neutron emissions

The goal is 1% RSD in less than 1000 seconds

[Source: D. Mcavoy, LLNL]
Detector Characteristics

- High Efficiency
- Effective discrimination against gamma rays:

\[
\text{Gamma-Ray Rejection Efficiency} = \frac{\text{False Neutron Counts}}{\text{Incident Gamma Ray}}
\]
Variables That Affect Neutron Counting

- Fission rate
- Multiplication (induced fission)
- \((\alpha,n)\) reactions
- Neutron energy spectrum
- Counter nonuniformity
- Neutron capture in the sample
- Neutron “die-away” time
Counter Fundamentals

- Neutron detection media: $^3\text{He}$
- Polyethylene moderator

- Performance parameters
  - Efficiency
  - Die-away time
  - Uniformity
  - Dead Time
Remember Those Variables?

- Fission rate
- Multiplication (induced fission)
- $(\alpha, n)$ reactions
  - Neutron energy spectrum
  - Counter non-uniformity
  - Neutron capture in the sample
  - Neutron “die-away” time
What is the Impact of These Parameters?

(α,n) Reactions

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Spontaneous fission yield (n/s-g)</th>
<th>α yield (α/s-g)</th>
<th>(α,n) yield oxide (n/s-g)</th>
<th>(α,n) yield UF₆/PuF₄ (n/s-g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>²³⁵U</td>
<td>3.0 x 10⁻⁴</td>
<td>7.9 x 10⁴</td>
<td>7.1 x 10⁻⁴</td>
<td>0.08</td>
</tr>
<tr>
<td>²³⁶U</td>
<td>5.5 x 10⁻⁴</td>
<td>2.3 x 10⁶</td>
<td>2.4 x 10⁻²</td>
<td>2.9</td>
</tr>
<tr>
<td>²³⁸U</td>
<td>1.4 x 10⁻²</td>
<td>1.2 x 10⁴</td>
<td>8.3 x 10⁻⁵</td>
<td>0.028</td>
</tr>
<tr>
<td>²³⁸Pu</td>
<td>2.6 x 10³</td>
<td>6.4 x 10¹¹</td>
<td>1.34 x 10⁴</td>
<td>2.2 x 10⁶</td>
</tr>
<tr>
<td>²³⁹Pu</td>
<td>2.2 x 10⁻²</td>
<td>2.3 x 10⁹</td>
<td>3.81 x 10¹</td>
<td>5.6 x 10³</td>
</tr>
<tr>
<td>²⁴⁰Pu</td>
<td>1.0 x 10³</td>
<td>8.4 x 10⁹</td>
<td>1.41 x 10²</td>
<td>2.1 x 10⁴</td>
</tr>
<tr>
<td>²⁴¹Pu</td>
<td>5.0 x 10⁻²</td>
<td>9.4 x 10⁷</td>
<td>1.3</td>
<td>1.7 x 10²</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>1.7 x 10³</td>
<td>1.4 x 10⁸</td>
<td>2.0</td>
<td>2.7 x 10²</td>
</tr>
</tbody>
</table>

Data from: [LA-13422-M]
Multiplication

Multiplication will also affect the total number of neutrons available for counting, so single dimension counting will not accurately characterize the sample.

For example, if a sample has a multiplication of 1.50 the total number of neutrons will increase with each generation.

<table>
<thead>
<tr>
<th>Generation</th>
<th>Source</th>
<th>Multiplication</th>
<th>Total Neutrons</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>100</td>
<td>0</td>
<td>100</td>
</tr>
<tr>
<td>1</td>
<td>100</td>
<td>50</td>
<td>150</td>
</tr>
<tr>
<td>2</td>
<td>100</td>
<td>50+25</td>
<td>175</td>
</tr>
<tr>
<td>3</td>
<td>100</td>
<td>50+25+13</td>
<td>188</td>
</tr>
<tr>
<td>4</td>
<td>100</td>
<td>50+25+13+6</td>
<td>194</td>
</tr>
</tbody>
</table>

The multiplicity distribution will shift in a sample with multiplication.
Multiplication

Data from: [E. Morse, Analytical Methods for Nonproliferation]

<table>
<thead>
<tr>
<th>P(ν)</th>
<th>$^{240}$Pu</th>
<th>$^{239}$Pu (th)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.066</td>
<td>0.011</td>
</tr>
<tr>
<td>1</td>
<td>0.232</td>
<td>0.099</td>
</tr>
<tr>
<td>2</td>
<td>0.329</td>
<td>0.275</td>
</tr>
<tr>
<td>3</td>
<td>0.251</td>
<td>0.327</td>
</tr>
<tr>
<td>4</td>
<td>0.102</td>
<td>0.205</td>
</tr>
<tr>
<td>5</td>
<td>0.018</td>
<td>0.073</td>
</tr>
<tr>
<td>6</td>
<td>0.002</td>
<td>0.010</td>
</tr>
<tr>
<td>7</td>
<td>-</td>
<td>0.001</td>
</tr>
<tr>
<td>8</td>
<td>-</td>
<td>0.002</td>
</tr>
</tbody>
</table>
Neutron Counting

- Counting doesn’t have to be one-dimensional
  - Single events
    - Are neutrons present?
  - Two time correlated neutrons
    - “Coincidence Counting”
    - Up to two sample parameters
  - Three time correlated neutrons
    - “Multiplicity Counting”
    - Up to three sample parameters

<table>
<thead>
<tr>
<th>Example Parameters</th>
<th>Variables</th>
</tr>
</thead>
<tbody>
<tr>
<td>mass</td>
<td>singles</td>
</tr>
<tr>
<td>sample multiplication</td>
<td>doubles</td>
</tr>
<tr>
<td>(α,n) production</td>
<td>triples</td>
</tr>
</tbody>
</table>

Data from Boldeman 1985
"I think you should be more explicit here in Step Two."

Sidney Harris
Counter Classifications

Coincidence Counters

• Measure singles (overall neutron count rate)
• Measure doubles (rate at which two neutrons are detected simultaneously)

Multiplicity Counters

• Extension of coincidence counting
• Measure singles, doubles, and triples

Active Well Coincidence Counter (AWCC)  [Source: PNNL-21090]

High-Efficiency Neutron Counter (HENC)  [Source: LA-UR-07-1402]
Extracting Information

Example: $F, M$ and $\alpha$

\[
S = F \varepsilon M \nu_{s1} (1 + \alpha)
\]

\[
D = \frac{F \varepsilon^2 M^2 f_d}{2} \left[ \nu_{s2} + \left( \frac{M - 1}{\nu_{i1} - 1} \right) \nu_{s1} (1 + \alpha) \nu_{i2} \right]
\]

\[
T = \frac{F \varepsilon^3 f_t M^3}{6} \left[ \nu_{s3} + \left( \frac{M - 1}{\nu_{i1} - 1} \right) (3 \nu_{s2} \nu_{i2} + \nu_{s1} (1 + \alpha) \nu_{i3}) \right]
\]

$\nu_{s1}, \nu_{s2}, \nu_{s3} =$ factorial moments of the spontaneous fission distribution

$\nu_{i1}, \nu_{i2}, \nu_{i3} =$ factorial moments of the induced fission distribution

$\varepsilon =$ detection efficiency

$F =$ fission rate

$f_d =$ doubles gate fraction

$M =$ sample self-multiplication

$\alpha =$ the $(\alpha,n)$ reaction rate

$f_t =$ triples gate fraction
Mass

• Put it all together:

\[ m_{240} = \frac{F}{473 \text{ fissions} \cdot s \cdot g} \]

• And if more than one isotope is present?

\[ ^{240}\text{Pu}_{\text{eff}} = 2.52 \cdot ^{238}\text{Pu} + ^{240}\text{Pu} + 1.68 \cdot ^{242}\text{Pu} \]

• With the isotopic ratios you can determine mass – remember gamma ray spectroscopy?
## Coincidence vs. Multiplicity Counters

<table>
<thead>
<tr>
<th>Strengths</th>
<th>Singles and Doubles</th>
<th>Singles, Doubles and Triples</th>
</tr>
</thead>
</table>
| **Strengths** | • Appropriate for 2 unknown variables  
• Shorter acquisition times than multiplicity counting (~200-300 s)  
• Cheaper (lower performance requirements)  
• Workhorse of the IAEA | • Better accuracy than with coincidence counting  
• Appropriate for samples where there are more than 2 (i.e., 3) unknowns (mixed oxide fuel)  
• Calibration for unknown sample parameter(s) isn’t required  
• Appropriate when prior knowledge is unreliable (inspections, verifications) |
| **Weaknesses** | • Prior knowledge of the sample may be required  
• Counter calibration with known samples is required | • Longer measurements times (~1000 sec)  
• Cost  
• Complexity |
# Fission Neutrons

- **Passive and active assay**
  - Passive detection → plutonium
  - Active interrogation → uranium

## What are you looking for?
“SIGNIFICANT QUANTITIES”

<table>
<thead>
<tr>
<th>SNM</th>
<th>Mass</th>
<th>Emission Rate</th>
<th>Neutron Flux at 3 m</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu</td>
<td>8 kg</td>
<td>$2 \times 10^6$ n/s</td>
<td>2 n/(s·cm$^2$)</td>
</tr>
<tr>
<td>HEU</td>
<td>25 kg</td>
<td>100 n/s</td>
<td>$10^{-4}$ n/(s·cm$^2$)</td>
</tr>
</tbody>
</table>

[Source: Kouzes et al, doi:10.1016/j.nima.2007.10.026]
Active Neutron Coincidence Counters

- Counts correlated neutrons from induced fissions (not spontaneous fissions)
- Requires an external (to the sample) neutron source
  - The external neutron source used must not produce any correlated neutrons (e.g., an AmLi source)
    - Used predominately for uranium assays due to its low spontaneous fission rate
    - Odd mass fissile isotopes ($^{239}$Pu, $^{235}$U, $^{233}$U, etc.)
    - High background compared to passive counters
  - Corrections are required for the multiplication between spontaneous and induced fission multiplication

[Source: PNNL-SA-86637]
Neutron Interactions

- Scattering
  - Elastic
    - (n,n)
  - Inelastic
    - (n,n')

- Absorption
  - Electro-Magnetic
    - (n,γ)
  - Neutral
    - (n,*n) * = 1, 2, ...
  - Charged
    - (n,p), (n,α) etc.
  - Fission
    - (n,f)
Fast Neutron Detection

- Elastic scattering of neutrons off of a target nuclei
  - Liquid scintillators
    - Organic scintillant in a hydrogen containing solvent
  - Plastics
    - Organic scintillant in polymerized hydrocarbon
  - Gas recoil proportional counters
    - Low Z gas

- Neutrons don’t need to be thermalized for detection, so the time to detect a signal is much faster than with capture reactions

- PSD may be necessary for gamma ray discrimination
Applications

• Faster die-away time for coincidence and multiplicity counting because neutrons don’t have to be thermalized
• High count rates
• Timing is preserved
• Some energy information available

[Source: LLNL-CONF-643807] [Source: INL/EXT-12-27619] [Source: DHS Stilbene]
Combined Analysis

• Full sample characterization may require more than one analysis technique:
  – Fission rate and isotopes
  – Gamma ray to neutron ratios
  – Gamma ray spectroscopy and neutron counting

[Source: Safeguards Techniques and Equipment]
NDA Information

**Gamma Ray**
- Energy
- Isotopic composition
- Electromagnetic radiation absorption
- Challenges with dense and non-uniform samples

**Neutron**
- Counting
- Temporally correlated events
- Active assay
- Minimal absorption even in large or non-uniform samples
- Hydrogenous materials affect the results

**Heat**
- Calorimetry
Calorimetry

- Physical and chemical processes result in heat production/consumption

- Calorimetry is the quantitative measurement of the heat exchanged

- Heat flux measurements are independent of the material and matrix

- Most precise method for plutonium mass determination available (requires isotopic information)
Calorimetry Basics

- Alpha particles, recoil progeny nuclei, attenuated within the sample cell which produces heat
- Nearly all beta-particle energy converted to heat (neutrinos escape)
- Rate of energy emission by ionizing radiation per unit mass of a single radionuclide = Specific Power

Source: [“Principles and Applications of Calorimetric Assay” Bracken and Rudy]
Specific Power

- Can be used with samples that contain multiple radioactive isotopes
- The effective specific power is:
  \[ P_{\text{eff}} = \sum_i R_i(t) \times P_i \]
  - \( R_i \) is the abundance of the \( i^{\text{th}} \) radionuclide (gamma ray spectroscopy)
  - \( P_i \) is the specific power of the \( i^{\text{th}} \) radionuclide

- Calorimetry can determine the mass of a radionuclide:
  \[ m_i = \frac{R_i(t) \times W}{P_{\text{eff}}} \]
  - \( m_i \) is the mass of the \( i^{\text{th}} \) radionuclide
Calorimetry Summary

- **Advantages**
  - Precise, nearly bias free, method of quantifying SNM
  - Can be used with different physical forms
  - The entire sample placed in the sample chamber is assayed

- **Disadvantages**
  - Effective power uncertainty can affect the accuracy for inhomogeneous samples
  - Long measurement times currently required (hours)
  - Heat from chemical reactions is indistinguishable from radioactive decay heat (though can normally be identified by time dependence)
NDA Information

**Gamma Ray**

- Energy
- Isotopic composition
- Electromagnetic radiation absorption
- Challenges with dense and non-uniform samples

**Neutron**

- Counting
- Temporally correlated events
- Active assay
- Minimal absorption even in large or non-uniform samples
- Hydrogenous materials affect the results

**Heat**

- Calorimetry
Densitometry

- Density measurement with electromagnetic radiation
  - Element (not isotope!) specific

- Fundamental relationship (for a specific energy):
  \[ I = I_o e^{(-\mu \rho x)} \]

- Radiation source can be man-made or natural

- Can use multiple energies for compound mixtures
X-Ray Fluorescence

- Compliments densitometry
- Measures the characteristic x-rays emitted by the atoms ionized by photons that interact with the sample (element specific)
- Appropriate for chemical analyses (limit is approximately a few ppm)
- Can be used with different sample forms with minimal (or no) sample preparation
- Surface measurements

Source: [Amptek XR-100CR detector with PX5 pulse processor and power supply]
NDA Goals

• **Looking for:**
  – Sample parameters
  – Material isotopics
  – Mass of each isotope

• **System parameters of interest:**
  – Efficiency
  – Energy and time resolution
  – Die-away time (coincidence and multiplicity counting)
  – Geometric uniformity

• **Combined methods:**
  – Neutron coincidence/multiplicity or calorimetry and isotopic information

• **Uncertainty:**
  – Precision and accuracy need to be quantified
The Limits of Measurement Systems

• Even if you could eliminate uncertainty from:
  – Operator influence
  – The measurement procedure

• There would still be uncertainty present

• Statistical uncertainty is here to stay...
  – But it has a minimum value, which should be targeted in all NDA measurements
The Overview

- **Gamma ray spectroscopy**
  - Isotopic identification
  - Scintillators, semiconductors
  - Peak identification can be challenging

- **Neutron “counting”**
  - Multi-dimensional (mass quantification)
  - Capture and scatter based detectors
  - There is a need for high performing $^3$He replacements

- **Calorimetry**
  - Mass quantification
  - Long measurement times

- **Densitometry**
  - Density or thickness determination
  - Challenges with compounds

- **X-ray fluorescence**
  - Chemical analysis
  - Surface or subsurface measurements
References

D. Chichester, “Introduction to Nuclear Safeguards: Nondestructive Analysis,” INL/CON-09-16307
D. Davidson and R. McElroy, “Comparison of Coincidence and Multiplicity Counting Techniques for Safeguards,”
INMM Proc. Japan Chapter Annual Meeting, 1995
Dissertation, 2013
L. Nakae et al., “Recent Developments in Neutron Detection and Multiplicity Counting with Liquid Scintillator,”
LLNL-CONF-422505, 2010
International Nuclear Verification Series No.1 Rev. 2, Safeguards Techniques and Equipment, 2011
M. Zendel, IAEA Safeguards Equipment, JAEA-IAEA Workshop, 2007
Upcoming Webinars

• Nuclear Materials Analysis — Mass Spectroscopy
• Chronometry
• Nuclear Materials Analysis — Radioanalytical Methods (Destructive)

NAMP website: www.wipp.energy.gov/namp