

Advanced Fuel Cycle Safeguards



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Outline

- **Introduction and Current Technical Basis**
- **Reprocessing**
- **Fuel Fabrication**
- **Fast Reactor**





Introduction and Current Technical Basis

- **Comparison of NRC, DOE, IAEA Definitions and Requirements**
- **Fundamentals of NDA**
- **Gamma-ray Based Instruments**
- **Neutron Based Instruments**
- **Calorimetry**





Comparison of NRC, DOE, IAEA Definitions and Requirements





Safeguard Regimes

■ Three Safeguard Regimes

- NRC: Regulates commercial nuclear facilities
- DOE: Regulates defense nuclear facilities
- IAEA: Applies safeguards to Nonproliferation Treaty signatories

■ Purpose of Safeguard Regimes

- Domestic (NRC and DOE)
 - *Protect against sabotage, theft and other threats of direct concern to the government and operators of the facility*
- International (IAEA)
 - *Goal is to detect diversion of significant amounts of fissile nuclear materials by the State/facility operator through verification of the State's declaration*





Types of Safeguards Measures

■ Nuclear Material Accounting

- Establishing the quantities of nuclear material present within defined areas and the changes in those quantities within defined periods.

■ Containment and Surveillance

- Ensures previously measured material in containers remains in place and unaltered or moves along declared and authorized paths
- Examples: video and seals

■ Process Monitoring

- The use of process data (flow rate, temperature, pressure, density, etc.) to draw frequent conclusions on plant configurations, operations and material flows/inventories
- Necessary for In-process, interim inventories





Safeguards Goal of Material Balance Areas (MBAs)

- **Fully measured material balance**
 - All materials accounted for
 - All materials have measured values





DOE Definition of MBA

- **Subsidiary account of facility**
- **Geographical area with defined boundaries**
- **Identify location and quantity of nuclear materials**





NRC Definition of MBA

- **Identifiable physical area**
- **Quantify nuclear material with measured values**





IAEA Definition of MBA

- Quantity of nuclear material in transfers can be determined
- Physical inventory can be determined
- Establish a material balance





Views of the MBA - Accountant

- Start with a “known” listing of material
- Record receipts and shipments
- The result is the ending, or “book” inventory:
$$BI + TI - TO = \text{Book Inventory}$$
- Verify that book inventory = physical inventory

$$\text{MB} = \text{Book inventory} - \text{Physical Inventory}$$





Views of the MBA - Scientist

- MBA is the “closed system” for which mass is conserved
- Determine the inventory through measurement at end of period
- Measure all the terms and determine the difference

$$\text{Mass Balance} = \text{BI} - \text{EI} + \text{TI} - \text{TO}$$





Views of the MBA – Physical Protection

- What is the target?
- Where is the target located?

An area containing nuclear material that:

- Must have controls
- Must be protected





Summary of MBA definitions

- **Accounting unit of facility**
- **Physical location**
- **Measure materials**
- **Establish a material balance; loss detection**





Material Accountancy

■ Inventory Types

- Long-term, Shut down Inventory
 - *Every 6-12 months the facility must cease processing and determine the amount of material contained within the facility*
- Interim Inventory, Shut down not required
 - *Every 1-2 months the facility must determine the amount of material within the facility, but shut-down is not required*
 - *In-process inventory is normally performed*
 - More difficult than shut down inventory
 - Larger errors due to estimates of material quantities





Material Accountancy

■ Detection requirements

- Inventory Difference (Material Balance, Material Unaccounted For) must be determined within certain error limits
- Those error limits determine how well the material must be measured
- Each regime has different error limits





Inventory Difference

■ Inventory Difference (ID) =

Beginning physical inventory

+ Sum of increases to inventory (receipts)

- Sum of decreases from inventory (shipments)

- Ending physical inventory

■ Sigma ID

- The uncertainty in the ID
- A statistical combination of the uncertainty of each measurement that makes up the ID
- Determines what losses you can detect





Sigma ID Requirements

Based on the Current, Most Stringent, Category I Requirements

Agency	Goal/ Requirement Terms	Sigma ID	Frequency of Long-Term, Shutdown Inventory	Frequency of Interim Inventory, Shutdown Not Required
IAEA	Material Unaccounted For (MUF): 8kgs Pu abrupt in one month and 8kgs protracted in one year	Sigma ID \leq 2.42 kg of Pu	Annual	Monthly
NRC	Standard Error of Inventory Difference (SEID)	Sigma ID \leq 0.1% of active inventory	Semi-Annual	Monthly
DOE	Limit of Error (LOE)	Sigma ID \leq 1% of active inventory or Category II of material	At least Annually	Bi-monthly





Rough Estimate of Sigma ID for a Commercial Reprocessing Facility

- **IAEA goal for Sigma ID**
 - Absolute number
 - Independent of the facility's throughput
 - More challenging for high throughput facilities.
- **NRC and DOE requirements for Sigma ID**
 - Percentages of the active inventory
 - Change with the throughput
 - Current NRC requirement is the most challenging of the three, even for this high throughput facility.

LWR Spent Fuel Processed per year	800 MTHM
Pu processed/yr (1% of spent fuel)	8,000 kg
Pu processed per month (assumed to be active inventory at time of shut down inventory)	667 kg
NRC's Sigma ID is 0.1% of active inventory	667 g
DOE's Sigma ID is 1% of active inventory	6.67 kg
IAEA's goal for Sigma ID	2.42 kg Pu





Improving Safeguards Effectiveness

■ Reduce sigma ID

- Better measurements
- Less material
 - *Smaller material balance areas*
 - *More frequent inventories*

■ Additional Measures

- C/S
- Processing Monitoring
- Access
- Inspector Presence
- But, quantification needed





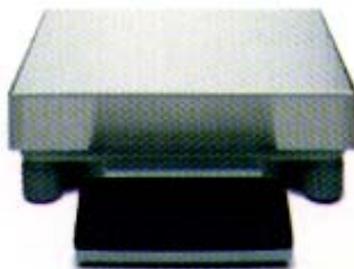
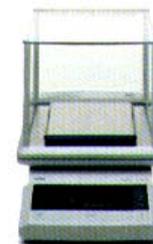
Fundamentals of NDA





The Fully Measured Material Balance

- Strive for highest possible accuracy
- Minimize uncertainty in Material Unaccounted For (MUF)
- Preferred measurement techniques
 - weighing, combined with
 - destructive analysis of samples





Some SNM items cannot, in practice, be measured by DA



waste, scrap, sealed product, unknown items, etc.





SNM Sample Containers



SNM comes not only in a variety of physical and chemical forms, but also in a variety of containers. Each container has an effect on the absorption of the emitted radiation and the measurement result.





Nondestructive Assay (NDA)

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





Basic Measurement Principle

Since each gram of a given isotope decays at a specified rate, the amount of radiation per second is proportional to the number of grams of the SNM in question.

The measured radiation is proportional to the amount of SNM in the sample IF...

- all of the radiation produced in the sample is emitted, and
- all of the emitted radiation is detected

These assumptions are hardly ever met, so data analysis must correct for the “missing” radiation. This is the main challenge in developing SNM measurement techniques and instruments.





Role of NDA

- Rapid verification
- Measurements of poorly-characterized items
- Assay or verification of sealed items
 - items in storage
 - temporarily sealed items (from cleanout, scrap, ...)
 - fuel rods and assemblies
- Measurement of holdup (**ducts, pipes, gloveboxes**)
- Support of audits & inspections
 - **by outside agencies** (IAEA, Euratom, ABBACC, GAN,...)
 - **by in-plant inspectors for internal assessments**





Uses for NDA

- Ore location
- Process Control
- Quality control
- Health & safety
- Criticality control
- Material control
- Materials Accounting
- Waste characterization/verification/screening
- Nuclear Inspections (**nonproliferation, arms control, regulation**)





SNM Attributes

- Emits alpha & beta radiation
- Emits gamma (γ) radiation
- Emits infra-red (heat) radiation
 - **[from α emissions into the matrix]**
- Emits singles neutrons
 - **[from (α, n) matrix interactions, delayed neutrons]**
- Emits coincidence neutron radiation
 - **[from spontaneous and induced fissions in the sample]**
- Is fissionable - can be induced to fission with neutrons.





Passive vs. Active NDA

Passive NDA: Measure the radiation emitted spontaneously by the sample.

Active NDA: Irradiate the sample and measure its response to the radiation.





Examples of Confirmatory Measurements



A Geiger counter is used to survey ductwork in a reprocessing plant to locate radioactive deposits. Quantitative holdup measurements then focus on these locations to determine the in-plant holdup.





Examples of Verification Measurements

Quantitative measurements — not used to replace accountability values.

- **Sampling of SNM inventory during inspection**
- **Mass measurements during some emergency inventories**





Gamma-ray Based Instruments





Basic Gamma-Ray Assay

Fundamental Measurement Principle

- Specific gamma rays (identified by energy) indicate the isotopic composition of the sample.
- Intensity of the gamma rays indicate the amount of the isotope.





Generic γ -Ray Assay Equation

$$M_{\text{SNM}} = \frac{R_{\text{Rad}} \cdot \text{CF}}{K}$$

- M_{SNM}** = Mass of Special Nuclear Material (SNM)
- R_{Rad}** = measured radiation rate (counts per second) from SNM sample
- CF** = correction for losses due to:
- Sample self-absorption
 - Container absorption
 - Measurement system electronics
- K** = Calibration constant (corrected response/gram SNM)

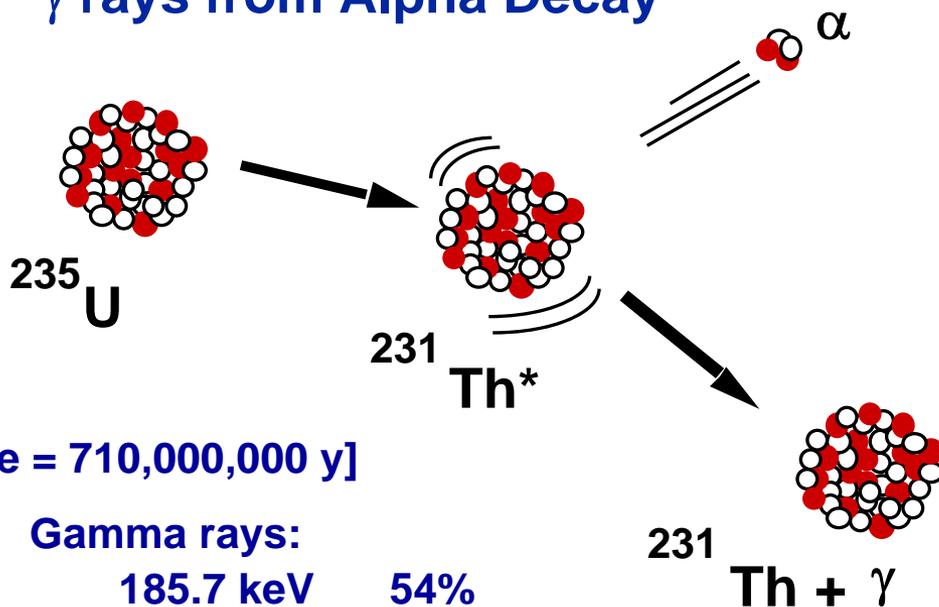




Gamma-Ray Signature

γ rays from Alpha Decay

alpha emission
(^4He)



[Half-life = 710,000,000 y]

Gamma rays:

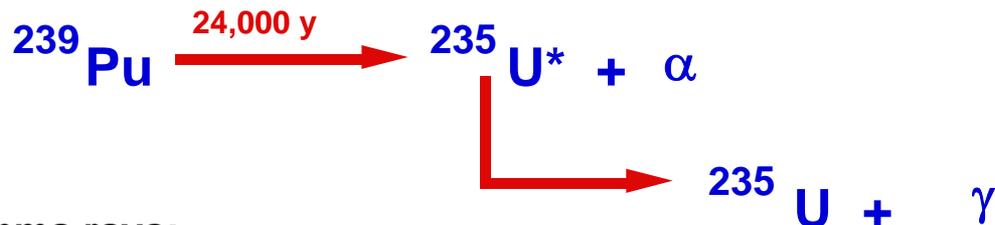
185.7 keV	54%
143.8 keV	11%
163.4 keV	5%
205.3 keV	3%

Gamma rays





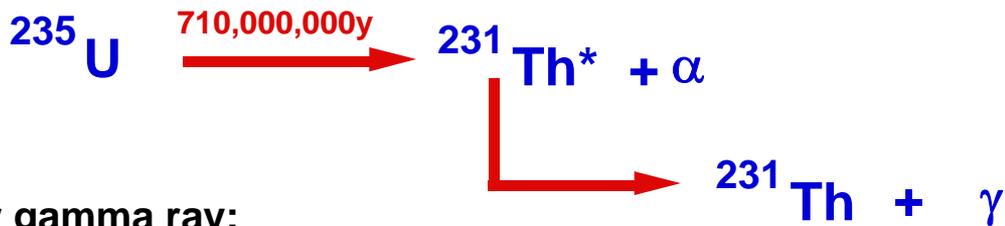
Major γ -Ray Signatures



Major gamma rays:

129.28 keV Intensity 140,000 gammas/(g-sec)

413.69 keV Intensity 34,000 gammas/(g-sec)



Major gamma ray:

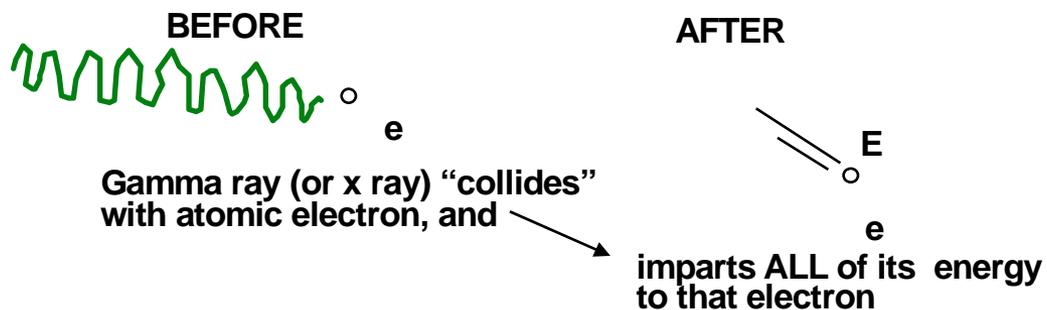
185.72 keV Intensity 43,000 gammas/(g-sec)



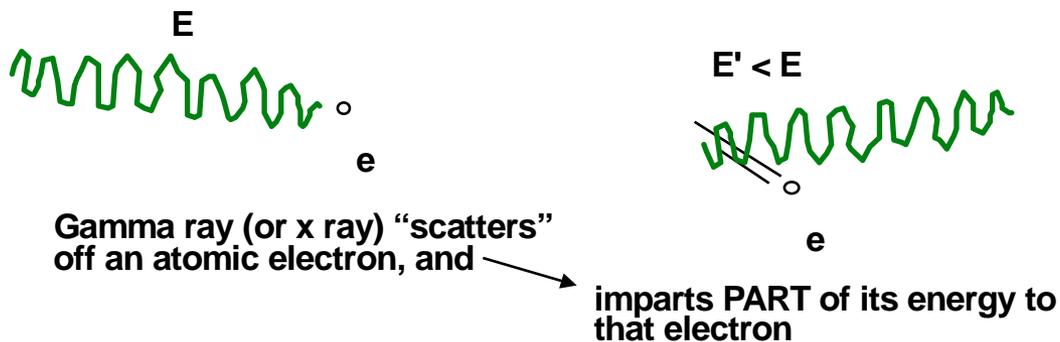


Interaction of Gamma Rays with Matter

PHOTOELECTRIC Effect

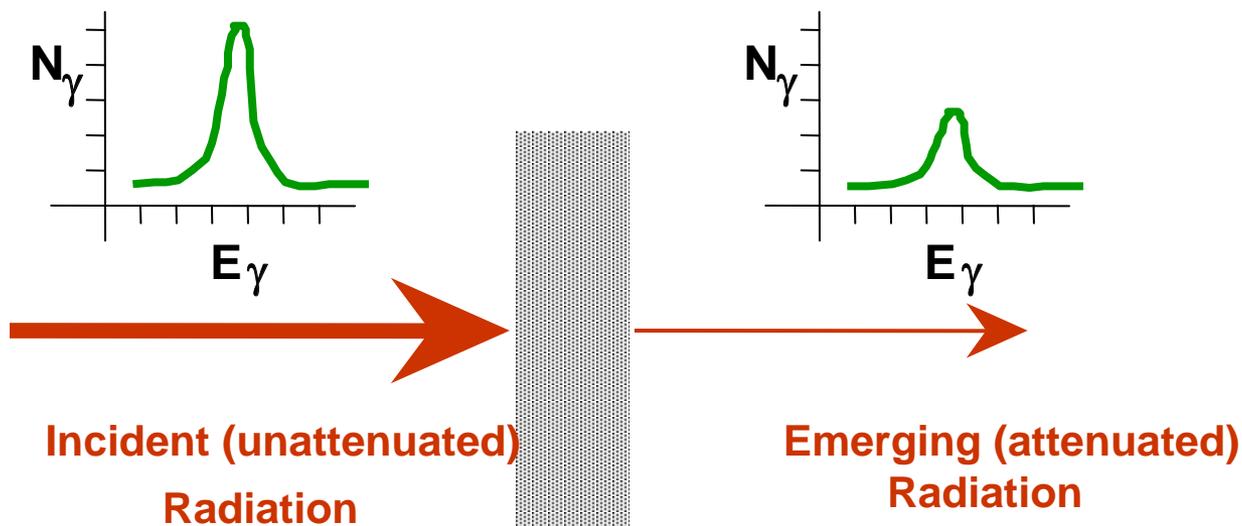


COMPTON Effect





Absorption of γ -Rays



$$I = I_0 e^{-\mu \rho x}$$

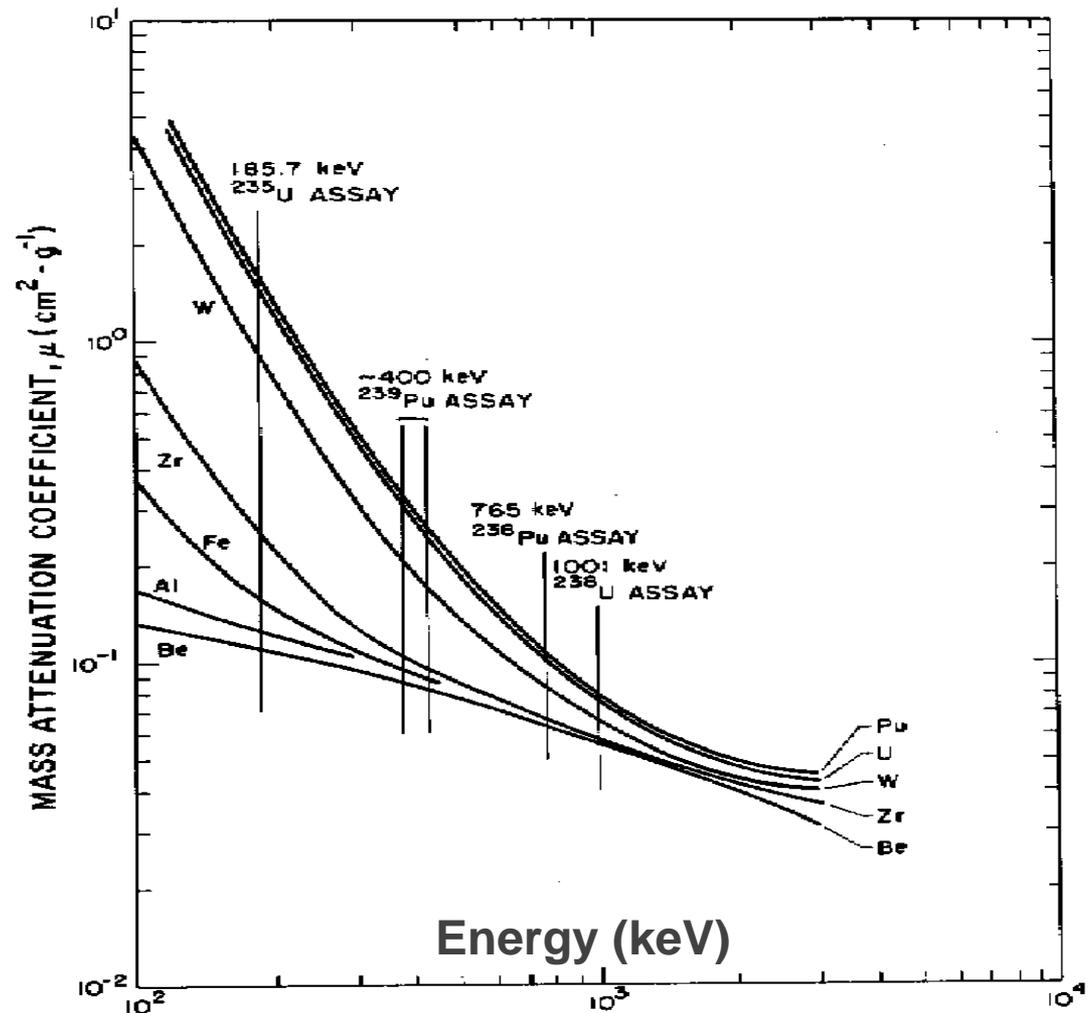
$$\text{Transmission } , T = I/I_0 = e^{-\mu \rho x}$$

- ρ = density (g/cm³)
- μ = mass absorption coefficient (cm²/g)
- x = thickness (cm)





Mass Absorption Coefficient

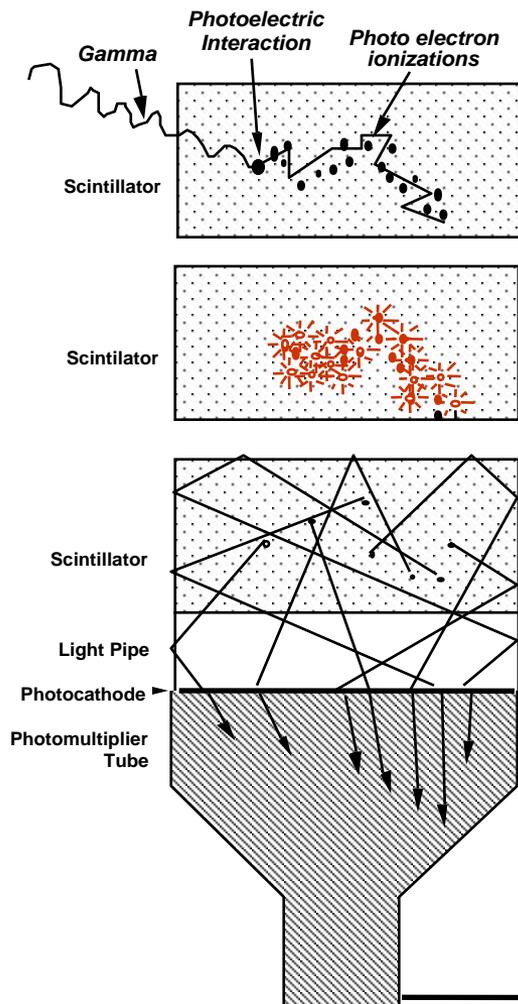


NRC Seminar June 10, 2008





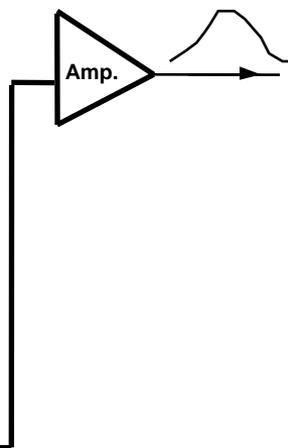
Nal Scintillation Detector



Gamma ray transfers all of its energy to an electron.

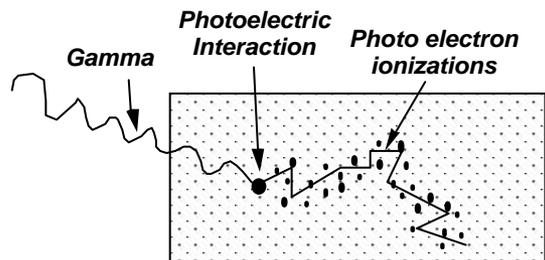
Photoelectron ionizes atoms which relax by emitting light whose intensity is proportional to the energy lost by the gamma ray.

This light travels to the photocathode of a photomultiplier tube, which emits electrons that multiply as they interact with the dynode string and are finally captured by the anode.

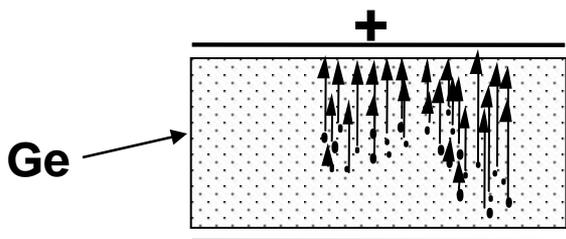




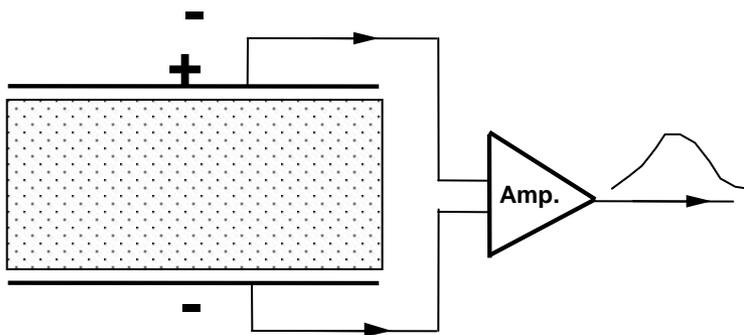
HPGe Solid-State Detector



Photoelectron ionizes Ge atoms liberating charge that is proportional to the energy lost by the γ ray



Electrons move toward the positive electrode and "holes" toward the negative one causing a charge pulse

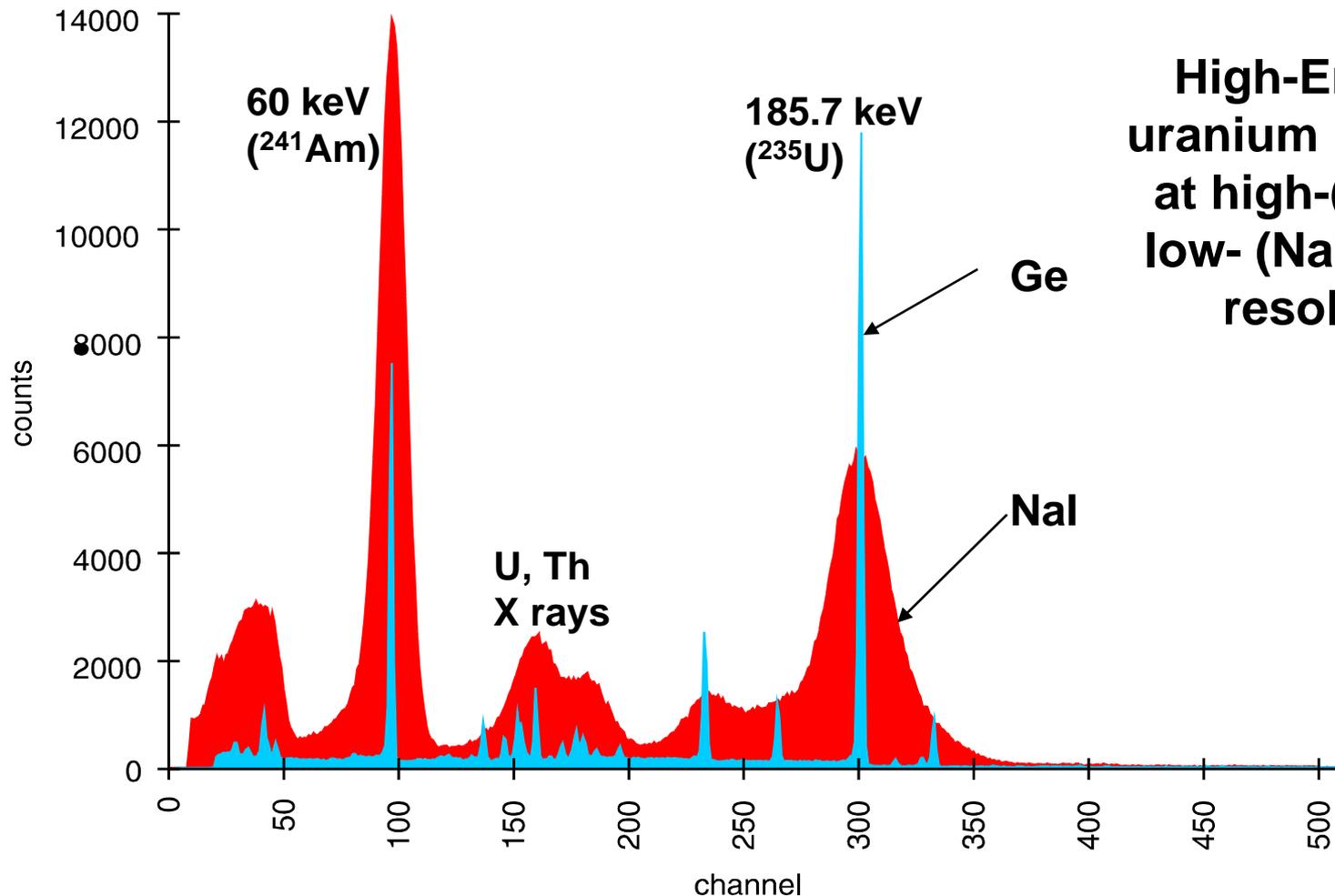


The preamplifier converts the charge pulse to a voltage pulse that is proportional to the energy of the γ ray





Uranium Spectral Features

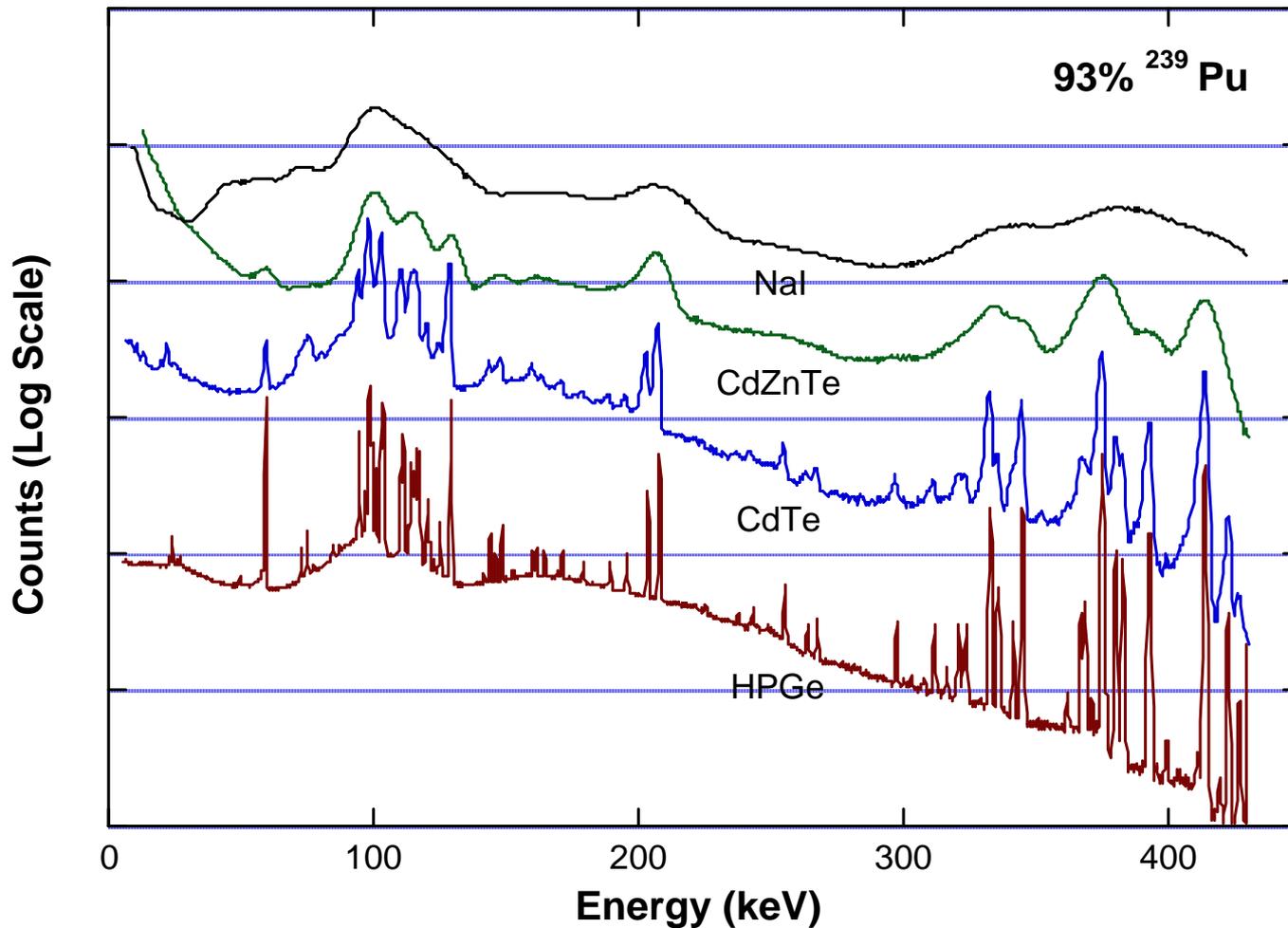


High-Enriched uranium spectrum at high-(Ge) and low-(NaI) energy resolution



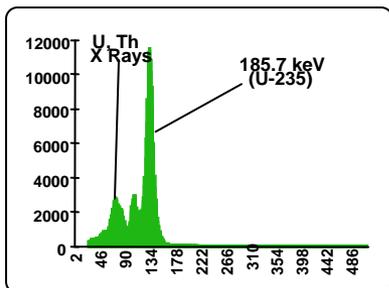


Plutonium Spectra



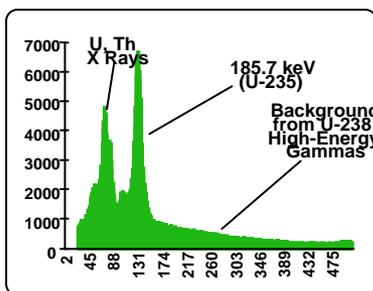


The Isotopic “Fingerprint”



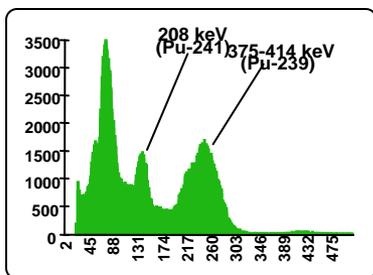
High-Enriched Uranium:

- 93% ^{235}U , very little ^{238}U
- 186-keV gamma-ray peak is strong compared to other radiation, such as x-rays
- little background above 186 keV from ^{238}U



Low-Enriched Uranium

- 3% ^{235}U , mostly ^{238}U
- 186-keV gamma-ray peak is weak compared to other radiation
- significant background above 186 keV from ^{238}U



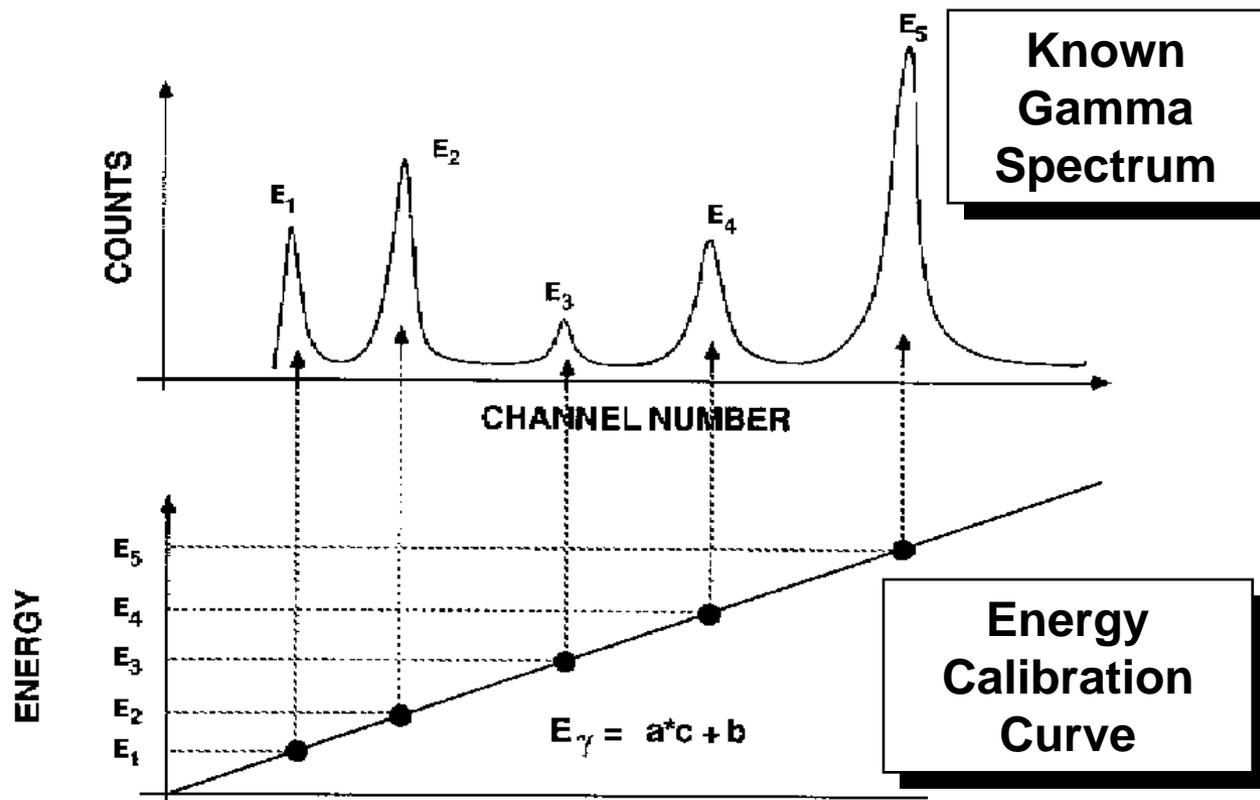
Plutonium:

- 93% ^{239}Pu , some ^{241}Pu
- 414-keV gamma-ray peak from ^{239}Pu evident, as is 208-keV from ^{241}Pu





Energy Calibration



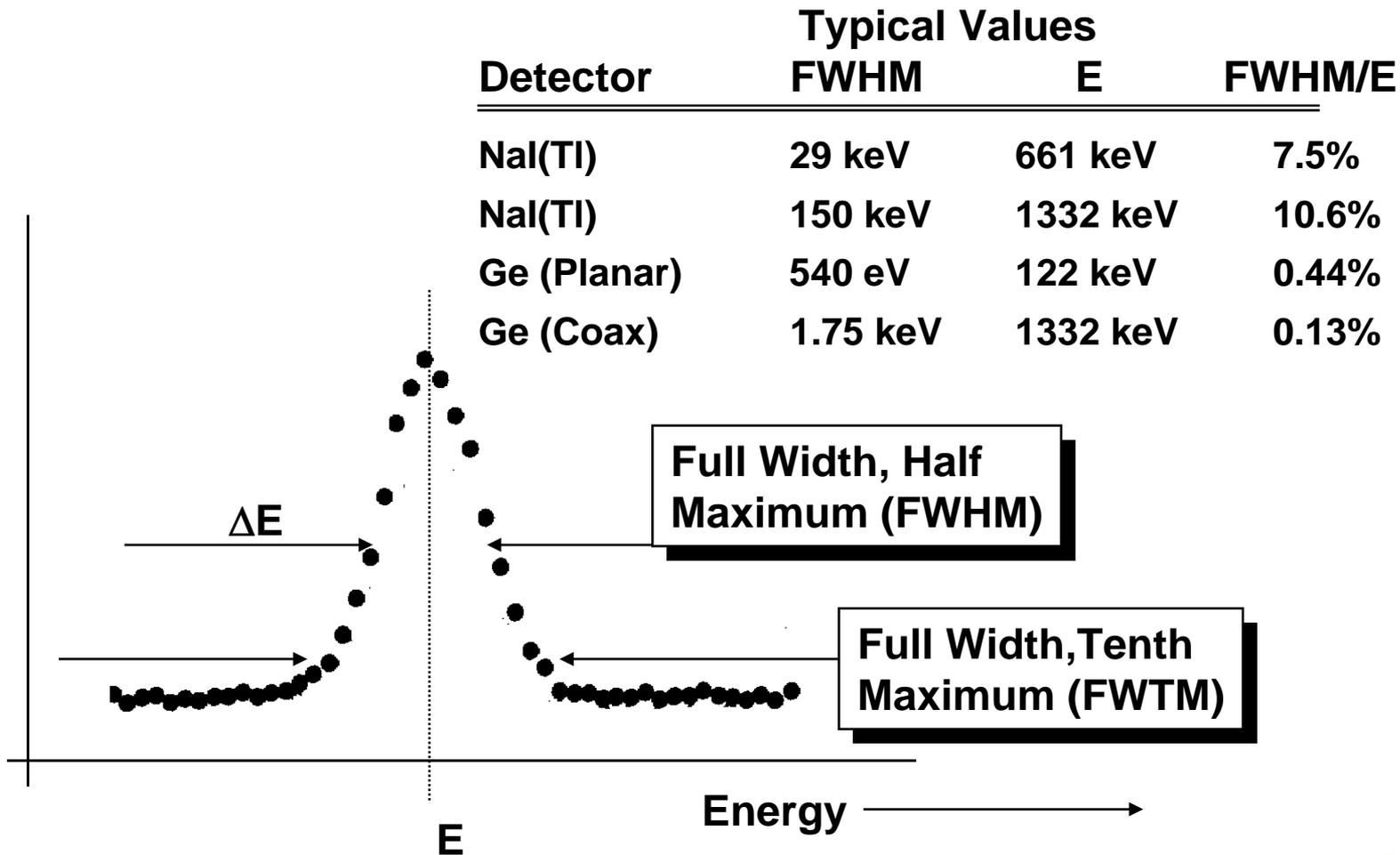
$$E_\gamma (\text{keV}) = A \cdot \text{Channel\#} + B$$

Energy accurate to about 0.1 keV



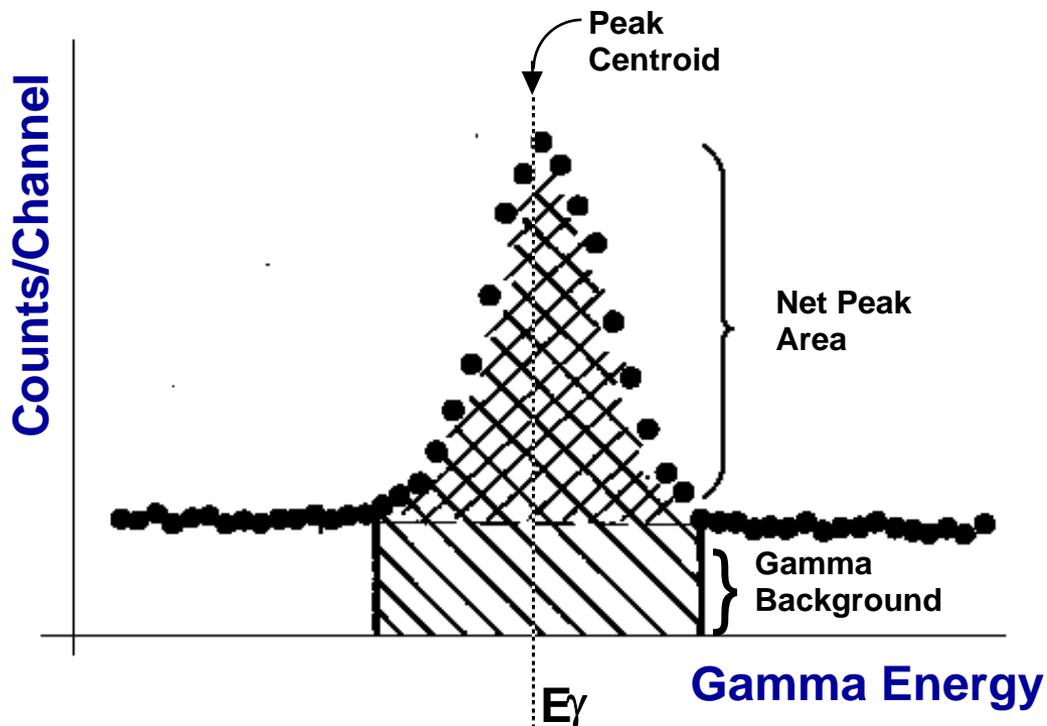


Energy Resolution





γ -Ray Photopeak Area Analysis

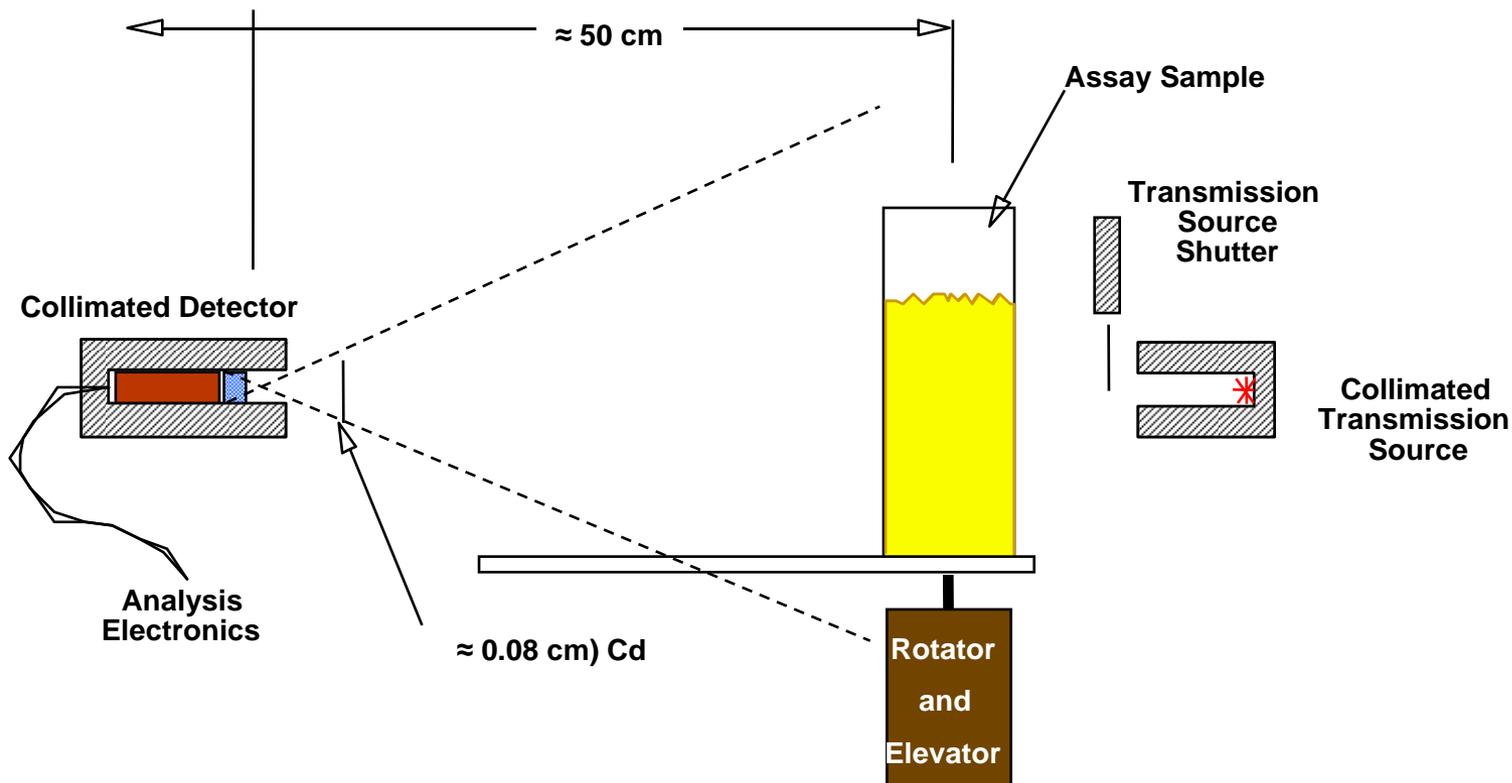


γ ray background is evaluated from the Compton continuum on either side of the peak





Basic Gamma-Ray NDA Setup

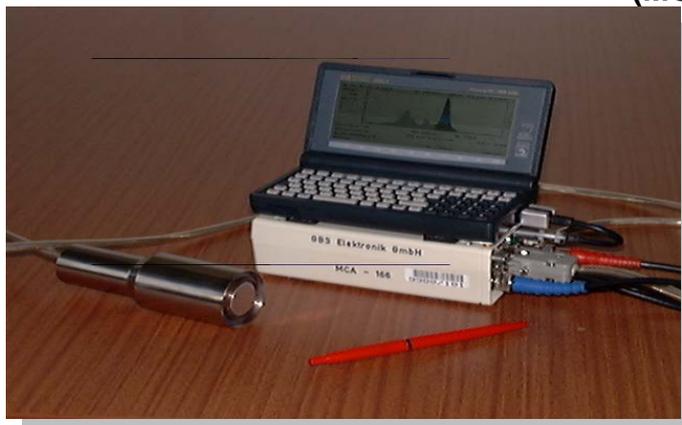
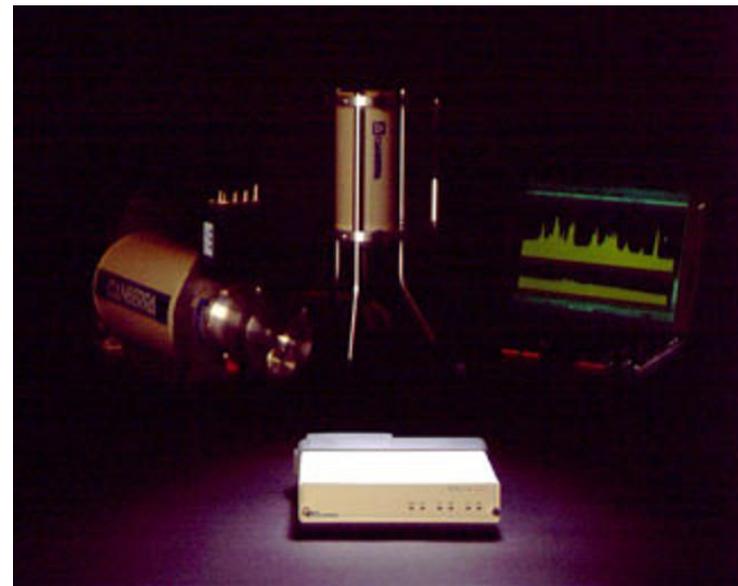
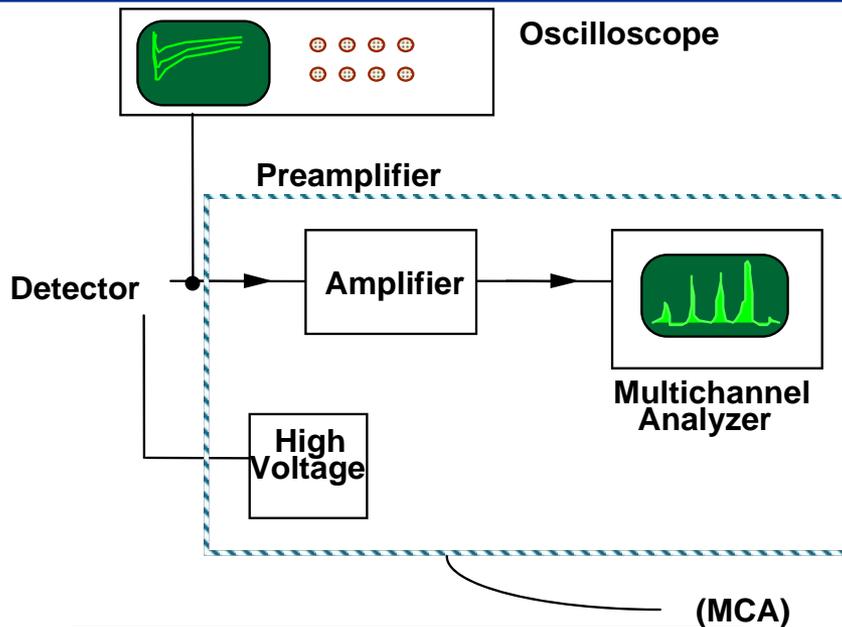


- Transmission source placed on far side of sample from detector.
- Sample mounted on rotating platform, at a distance from the detector.
- Detector views entire sample at one time.





Gamma Spectroscopy Instrumentation



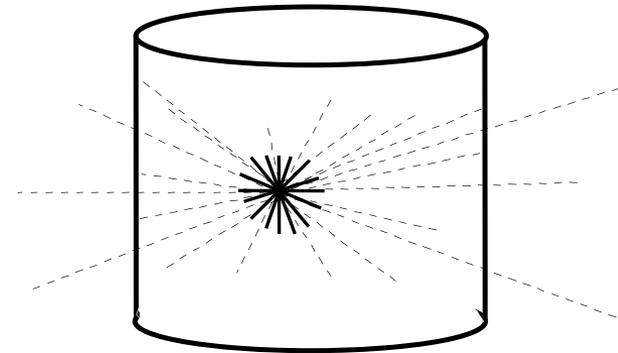
- **Battery-powered**
- **Computer-based**
 - **data acquisition**
 - **data analysis**
- **Commercially available**





Sample Characteristics for Successful Gamma-Ray Assay

- **Medium to low density**
- **Medium to small size**
- **Homogeneous (uniform) material distribution**



Dense, heterogeneous materials are inappropriate for gamma-ray assay, because they have too much absorption. Such samples are better suited for neutron assay techniques.





Basic Calibration Procedure

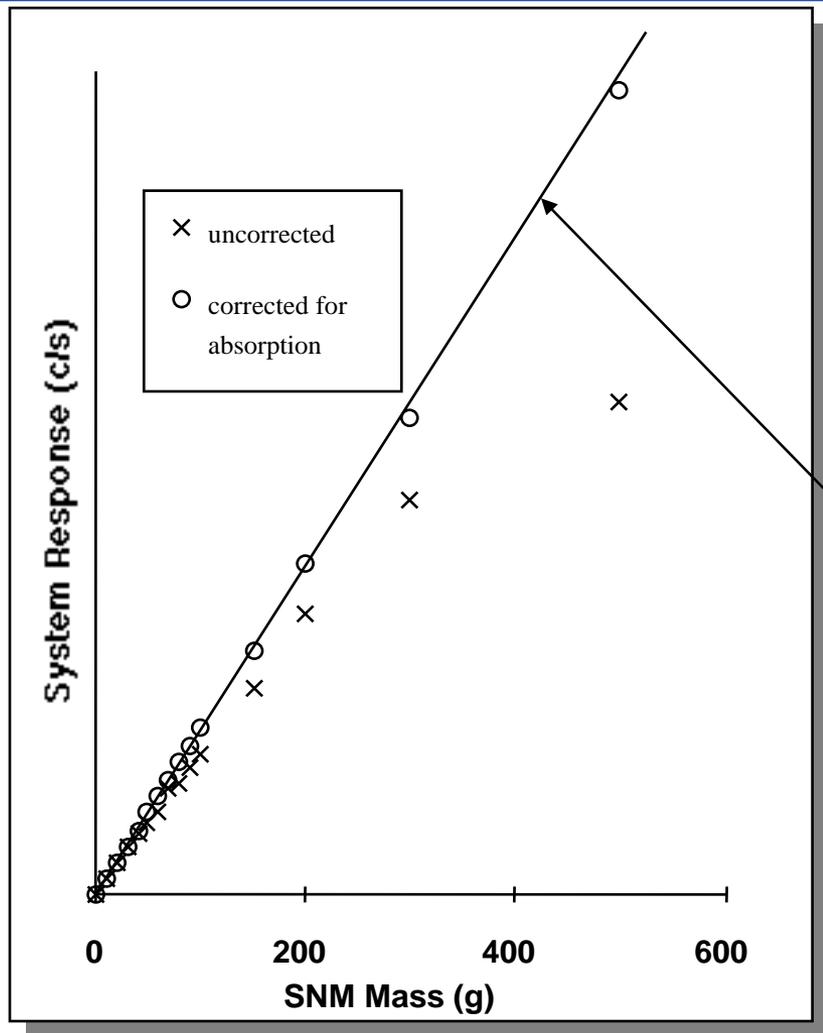
Purpose: to establish the relationship between the response of the detector system and the amount of SNM in the sample.

- Set up measurement apparatus for assay campaign.
- Establish data acquisition & analysis procedures.
- Measure **known samples** (calibration standards) using these procedures and this setup....





Example Gamma Assay Calibration Curve



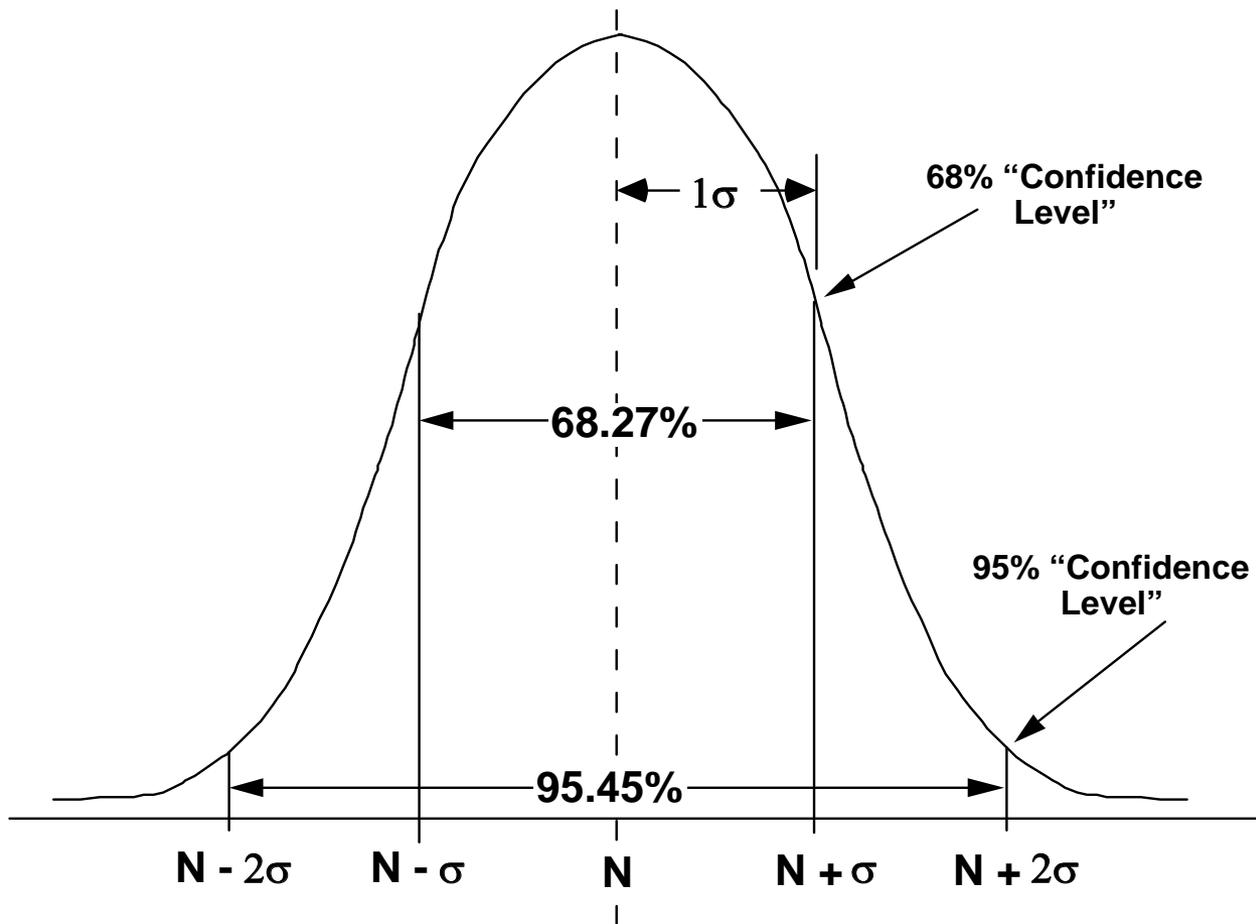
Note that absorption by the sample is increasingly important at higher SNM masses.

Application of the self-absorption correction (using the external source) removes the effect of this absorption.





Normal (Gaussian) Distribution





Uncertainty in a Nuclear Count

For a measured number of counts, N , from a nuclear counting process, the standard deviation in N is:

$$\sigma(N) = \sqrt{N}$$

The RELATIVE STANDARD DEVIATION (RSD or σ_R) of the measurement is:

$$\sigma_R(N) = \frac{\sigma(N)}{N} = \frac{1}{\sqrt{N}}$$

To improve the RSD, increase N (i.e., “take more data”):

- Count longer or repeat measurement (equivalent)
- Use “hotter” source
- Move sample and detector closer to one another





Measurement Control

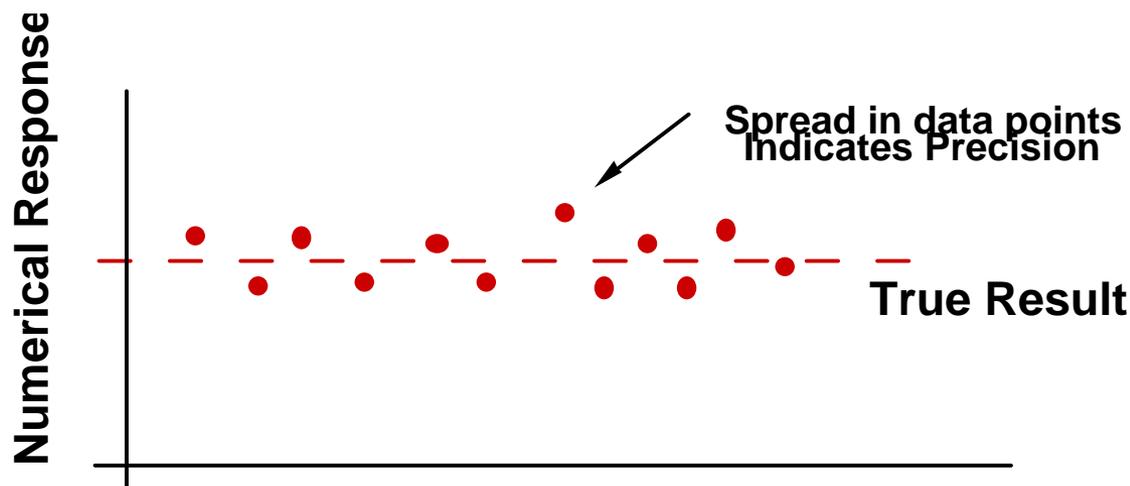
- Goals:
 - To control the quality of the measurement process
 - To verify the continued stability of the measurement system since the last calibration

- Procedures
 - Assay measurement control standards periodically
 - Measure room radiation background periodically
 - Measure data quality (**appearance of spectrum, etc.**)
 - *electronics gain*
 - *detector resolution*
 - Note visual appearance of instrument

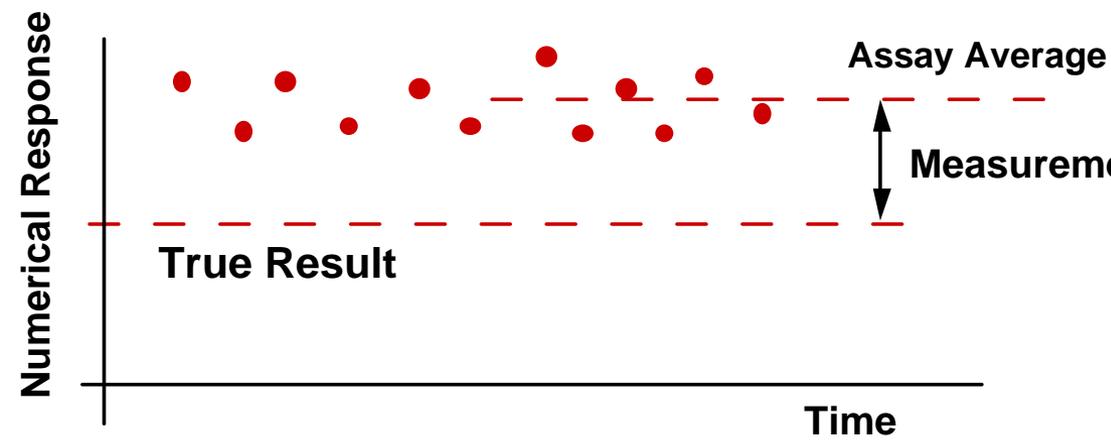




Measurement Precision and Bias



Measurement Precision:
The degree to which repeated measurements of the same sample give the same results.



Measurement Bias:
The amount by which the average of many results on a standard differs from the true value.





Methods to Improve Measurement Precision

- Count for longer time
- Operator Errors
 - **Provide explicit assay procedures**
 - **Provide careful training for assay procedures**
 - **use checklists to verify execution of procedures**
- Background Variations
 - **Shield detector from extraneous signals**
 - **administratively control source traffic in counting area**
 - **perform regular background measurements, diagnostics**





Dealing with Measurement Bias

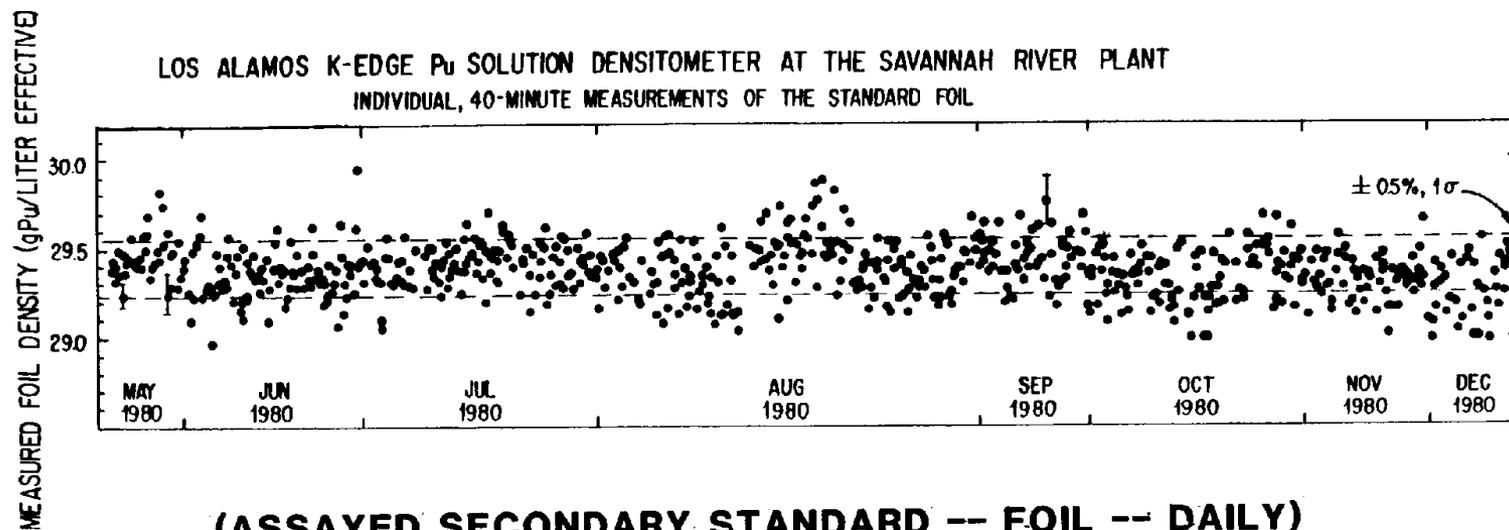
- Sources of Measurement Bias
 - **Statistical errors in calibration**
 - **Shifts in shielding, absorbers, collimators**
 - **Changes in geometry of setup (detector, sample stand,...)**
 - **Differences between sample container and container for calibration standards (diameter, height, wall thickness, ...)**
 - **Lumps in sample (error in absorption correction)**

- Strategies to Reduce Measurement Bias
 - **Calibrate often and make calibration measurement precise**
 - **Anchor shielding and collimators to measurement system**
 - **Anchor detector and sample stations**
 - **Calibrate for several container sizes; develop geometry corrections to calibration; control sample containers to match standard containers.**
 - **Assure uniform samples; apply lump corrections, if possible.**





Control Charts



Features

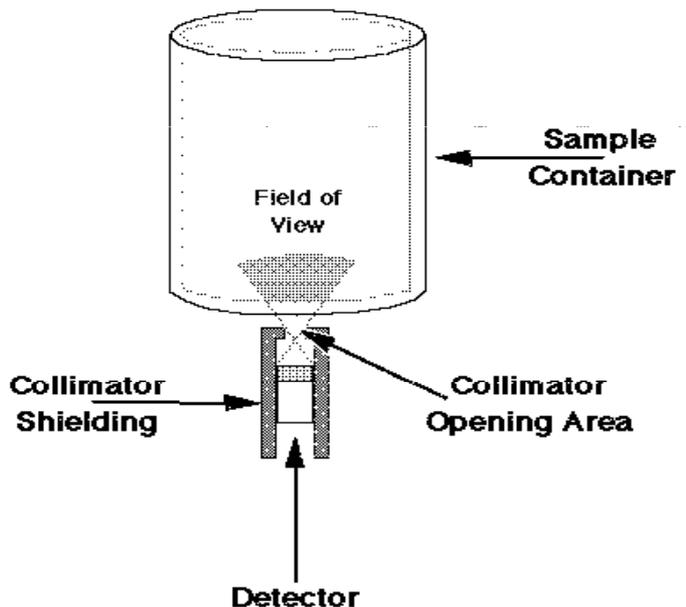
- Assayed standard Pu foil, built into instrument.
- Generally normal, quite stable performance
- Some excursions, in May, August, 1980 [Temperature problems when air conditioning fails]
- Slight downward drift in winter, 1980





Infinite-Thickness Enrichment Assay

Schematic of Enrichment Measurement



Detector views the sample through a collimator.

Visible volume determined by collimator and the absorption coefficient of U.

The 186-keV γ -ray intensity is proportional to the ^{235}U in the visible volume.

Los Alamos





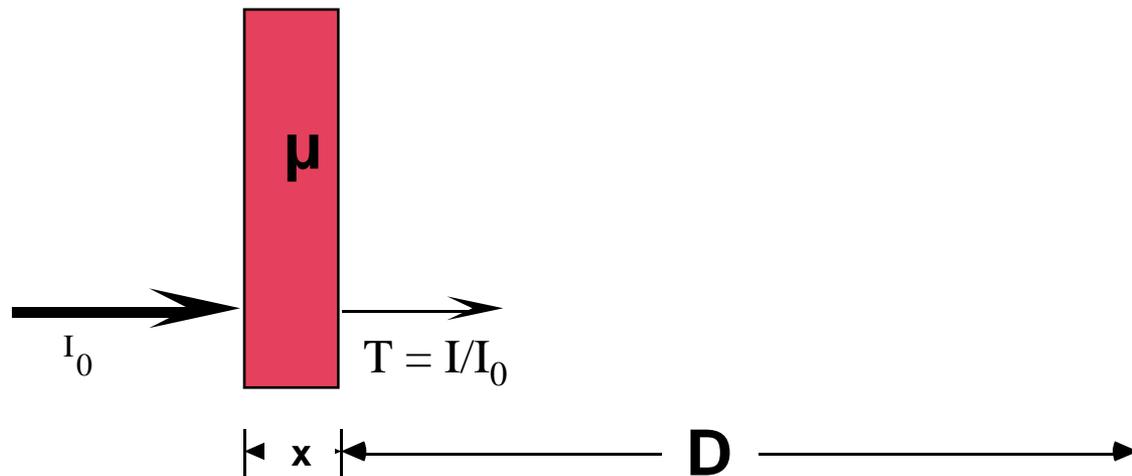
Infinite Thickness for 186-keV γ Rays in Uranium Materials

<u>Material</u>	<u>Density (ρ)</u> <u>(g/cm³)</u>	<u>Mean Free Path</u> <u>(cm)</u>	<u>“Infinite” Thickness</u> <u>(cm)</u>
Metal	18.7	0.04	0.26
UF ₆ solid	4.7	0.20	1.43
UO ₂ (sintered)	10.9	0.07	0.49
UO ₂ (powder)	2.0	0.39	2.75
U ₃ O ₈ (powder)	7.3	0.11	0.74
Uranyl nitrate	2.8	0.43	3.01





SLAB CORRECTION FACTOR (Far-Field) CF_{atten}



DETECTOR

Assumes that detector is far enough away from source that parallel γ -ray “beams” reach the detector.

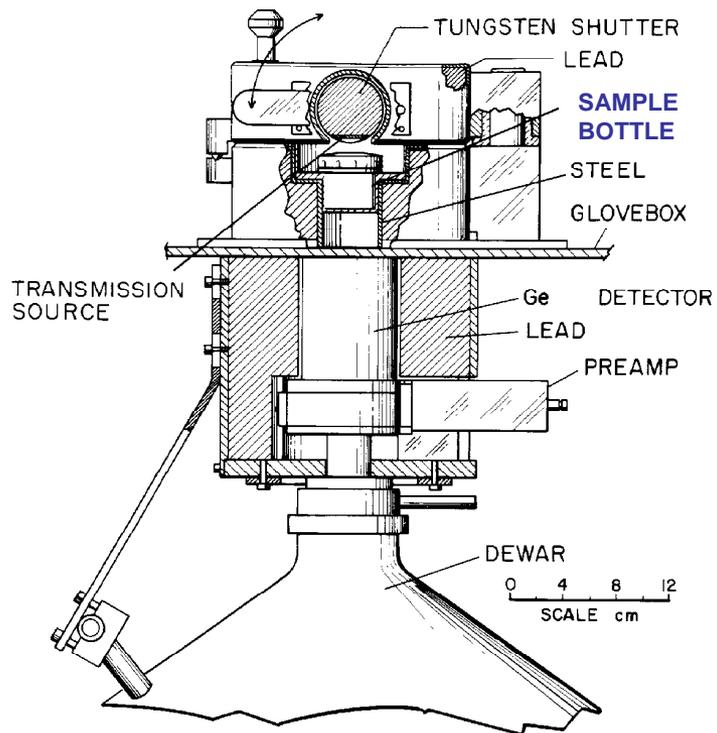
FOR $D \gg x$:

$$CF_{\text{atten.}} = \frac{\mu \rho x}{1 - e^{-\mu \rho x}} = \frac{-\ln(T)}{1 - T}$$

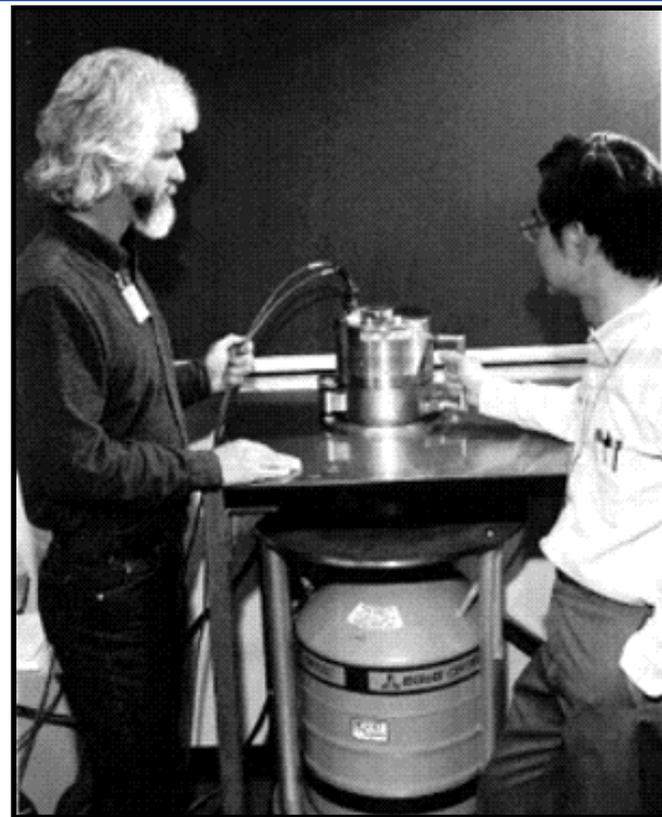




Solution Assays by Gamma-Ray Spectroscopy



PLUTONIUM SOLUTION ASSAY INSTRUMENT



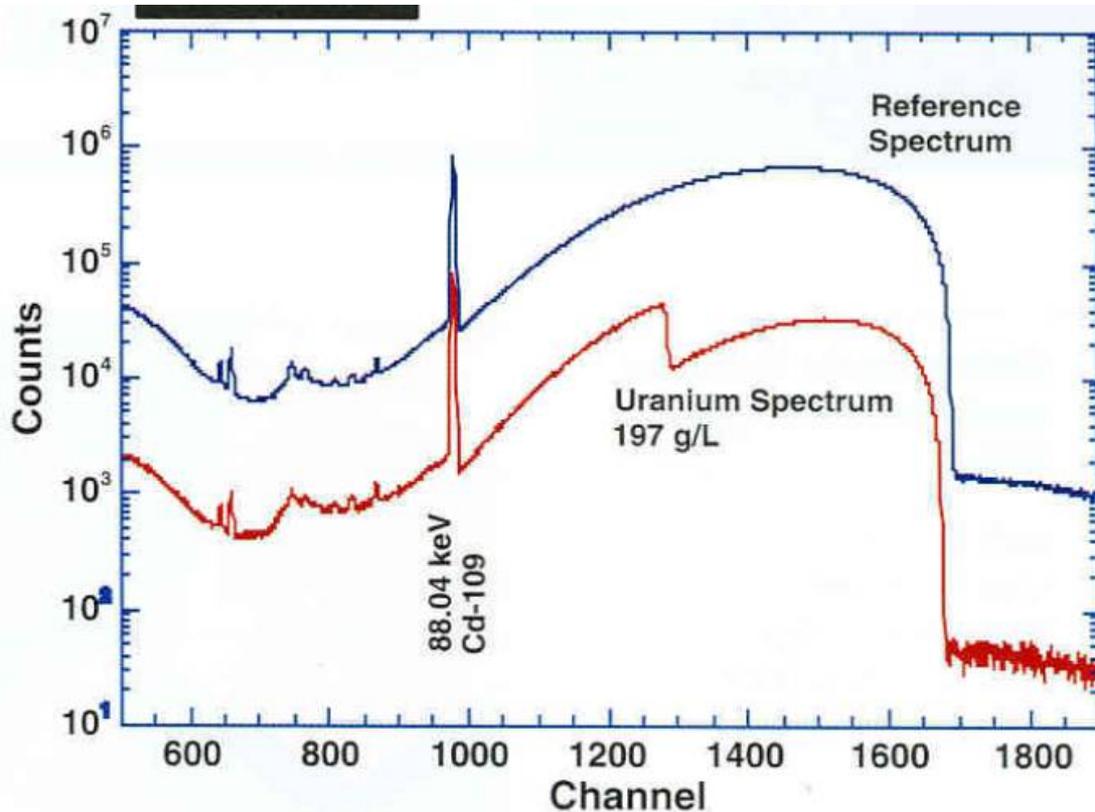
- Absorption-corrected passive gamma-ray assay
- High-resolution γ detector measures spectrum through floor of glovebox.
- Gives grams isotope in a 0.001 to 400 gram SNM/liter sample, with 0.2% to 1% accuracy in 1000 sec.



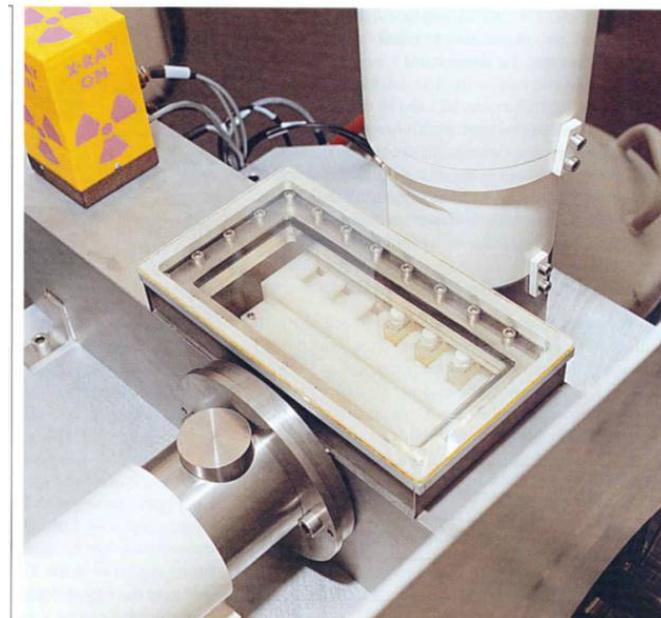


Absorption edge densitometry, x-ray fluorescence

K-Edge densitometer



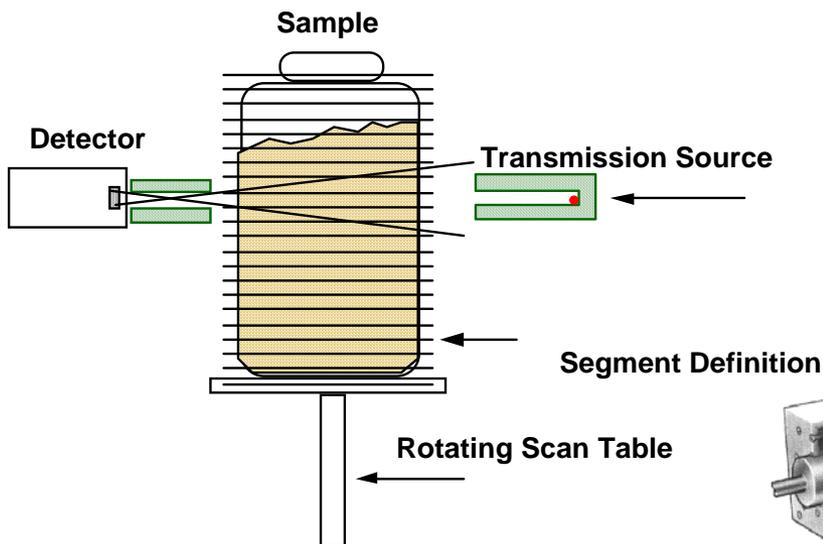
Hybrid K-Edge XRF





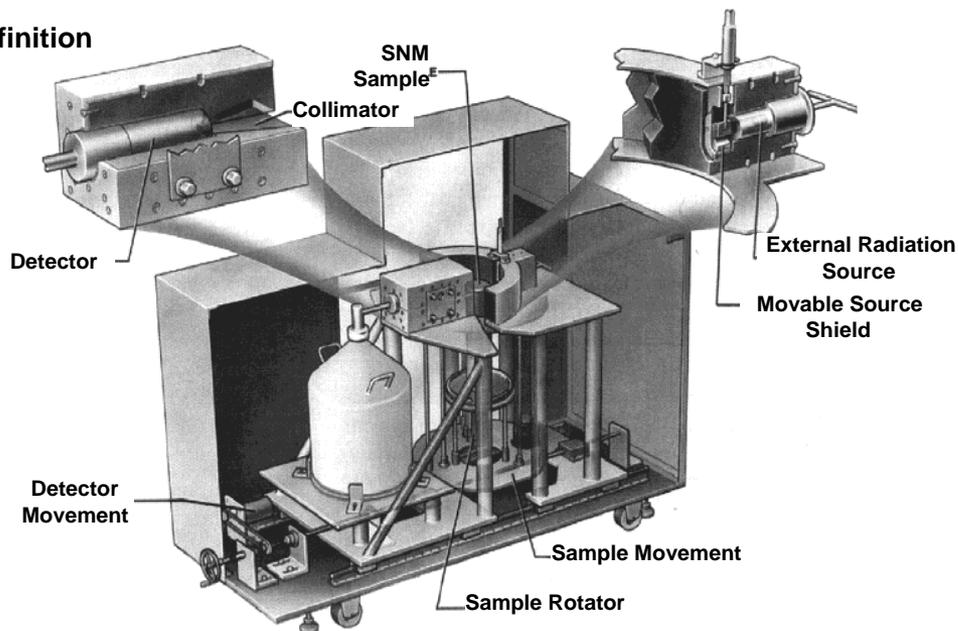
Other Transmission-Corrected Gamma Assay Systems

Segmented Gamma Scanner (SGS)



- **Passive, absorption-corrected gamma-ray assay of SNM**
 - samples of final product
 - low-density U waste
- **Sample is assayed one segment at a time**

- **Appropriate samples: small (<2 liters) or low density waste**
- **Accuracy possible in 1000 sec: 2% to 10% on 1 to 200 grams U**





Gamma-Ray Tomography

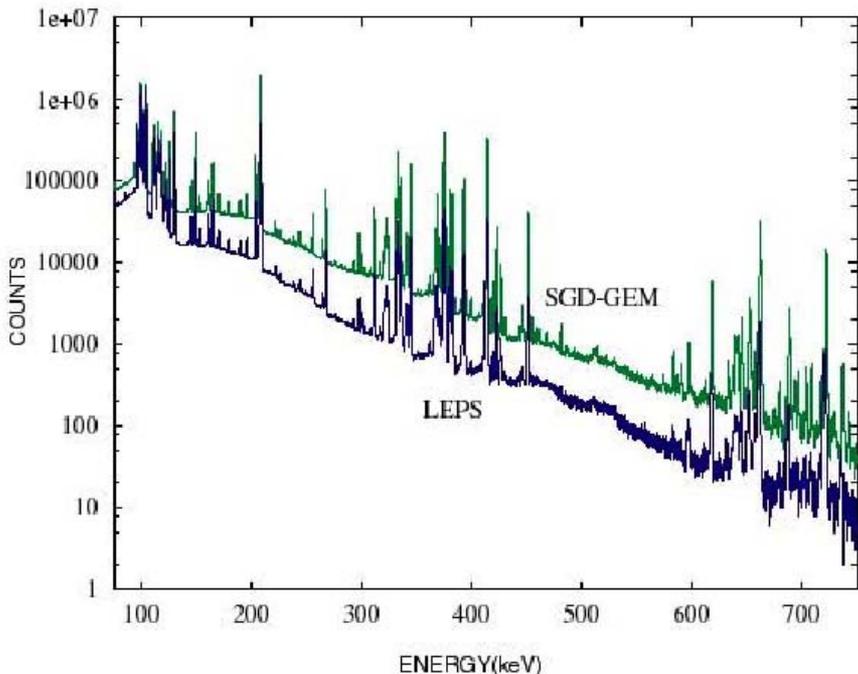


- Segmentation is replaced by collimation that divides the sample into “voxels.”
- Measuring the transmission, in each voxel, gives a 3-D “density map” of the sample.
- The measured density and γ -ray rate from each voxel provide a 3-D image of the SNM content.

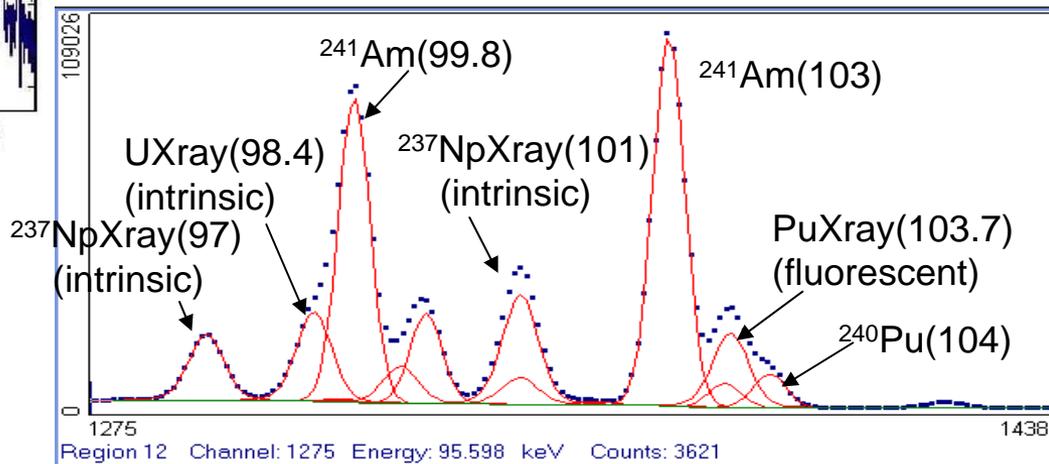




Passive Gamma-Ray NDA Applications: Plutonium Isotopic Composition



A high-resolution spectrum provides gamma-ray intensities from the isotopes in the sample. Isotope ratios are computed from peak ratios. Since a relative measurement is performed, standards are not needed, and measurement geometry does not need to be controlled. Relative detector efficiency, as a function of gamma-ray energy, is obtained from the spectrum.





Portable In-Plant Holdup Measurements by Gamma-Ray Spectroscopy



- Portable multichannel analyzer with gamma detector and data analysis electronics.
- Operator can position a shielded detector in a variety of locations to meet variety of measurement requirements.
- Holdup measured at 5% to 50% accuracy on 0.1 to 10 grams SNM; accuracy very dependent on measurement geometry.





Gamma NDA Summary

- Gamma NDA Technique:
 - **Count gamma rays from sample**
 - **Gamma-ray absorption correction very important**
 - **Appropriate samples: small to medium density/size, uniform**
- Accuracies Possible:
 - **0.2% to 50%, depending on sample uniformity and technique**
 - **Best performance on solutions**
 - **Improved performance for high-resolution over low-resolution detectors.**
 - **Worst cases: Large, dense, heterogeneous samples [often assayable by neutron techniques]**



Advanced Fuel Cycle Safeguards



Michael Miller
GNEP Safeguards Campaign Director
Los Alamos National Laboratory

U.S. Nuclear Regulatory Commission
Rockville, MD
June 10, 2008



Outline

- Introduction and Current Technical Basis
- Reprocessing
- Fuel Fabrication
- Fast Reactor





Introduction and Current Technical Basis

- **Comparison of NRC, DOE, IAEA Definitions and Requirements**
- **Fundamentals of NDA**
- **Gamma-ray Based Instruments**
- **Neutron Based Instruments**
- **Calorimetry**



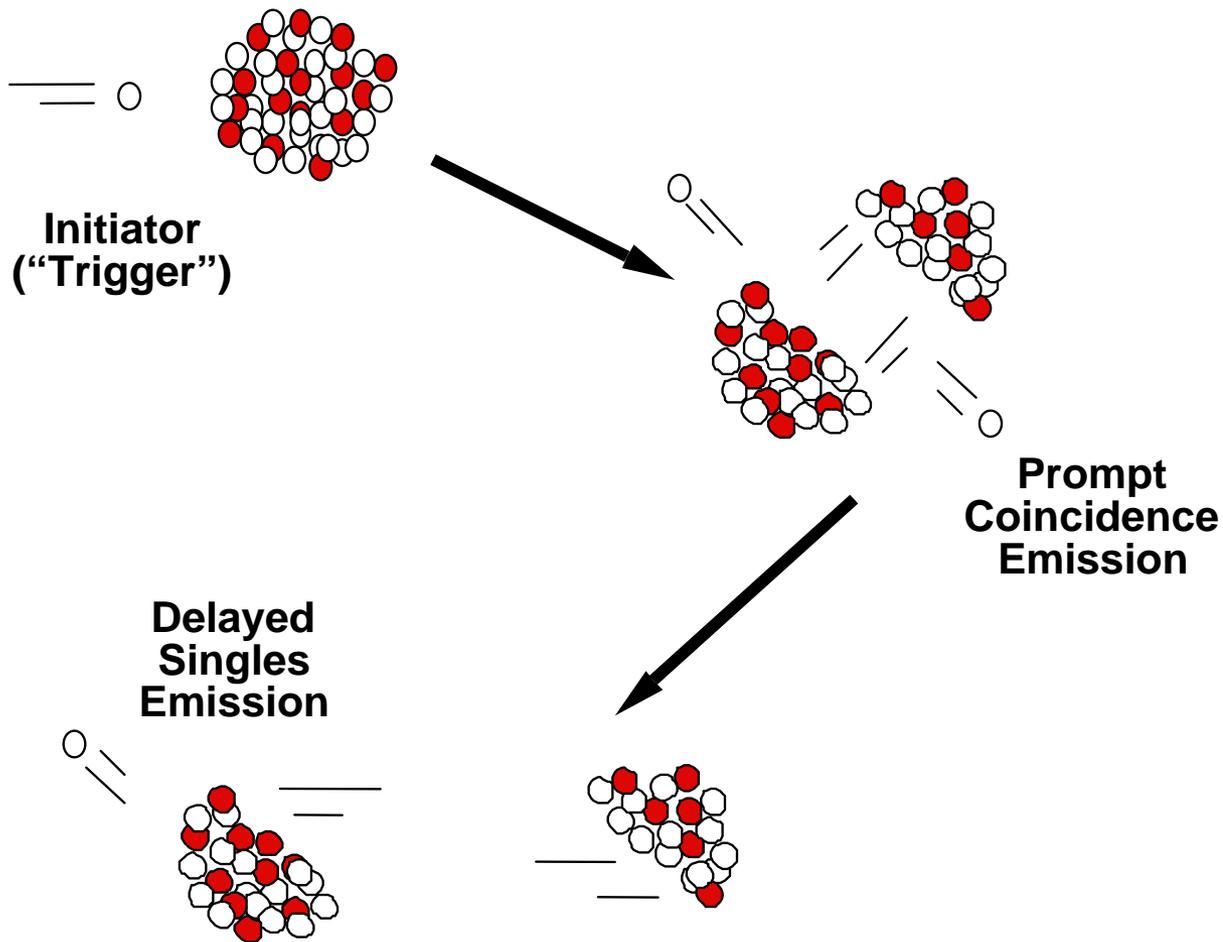


Neutron Based Instruments





The Neutron's Role in Fission





Introduction: Why Neutron Counting?

- Neutrons have great **penetrability**. Thus neutron signatures are sometimes the only way to rapidly assay large, dense samples.
- Neutron Count Data can be obtained **rapidly**. (But isotopic information is needed to interpret the data correctly and this adds more time to the assay.)
- Spontaneous Neutron Emission is a **primary** signature of the even isotopes of plutonium.
- Induced Fission Neutrons are a signature for both fissile plutonium and uranium.





History of Neutron Counting

- The first neutron assay instruments attempted to use the **total** neutron rate to deduce assay information -- however, accurate assays can be obtained only for a very few types of SNM.
- The next development was **neutron coincidence counting**. This technique has had wide application for international safeguards. It has had more limited application to US domestic safeguards because large errors can occur if the technique is not applied correctly to impure materials.
- **Neutron Multiplicity Counting** is an extension of neutron coincidence counting. It improves neutron assay accuracy dramatically by adding more measured information.





Multiplicity Counting Vs. Coincidence Counting

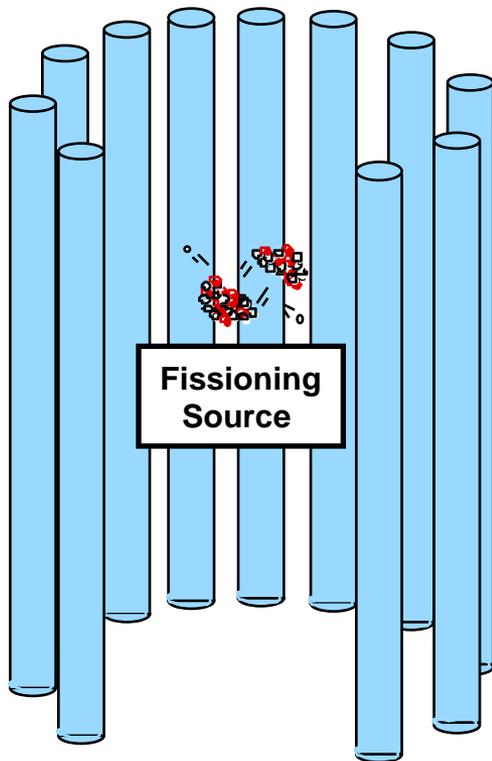
- In standard neutron coincidence counting, a priori information must be used to obtain an assay. Does not work well for materials that contain variable, low-Z impurities.
- In neutron multiplicity counting, the distribution of coincident multiplets in the neutron pulse stream is used to obtain a third measured quantity: the rate of coincident “triples”.
- With careful detector design to reduce the variables to three, neutron multiplicity assays are possible without sample dependent calibrations.





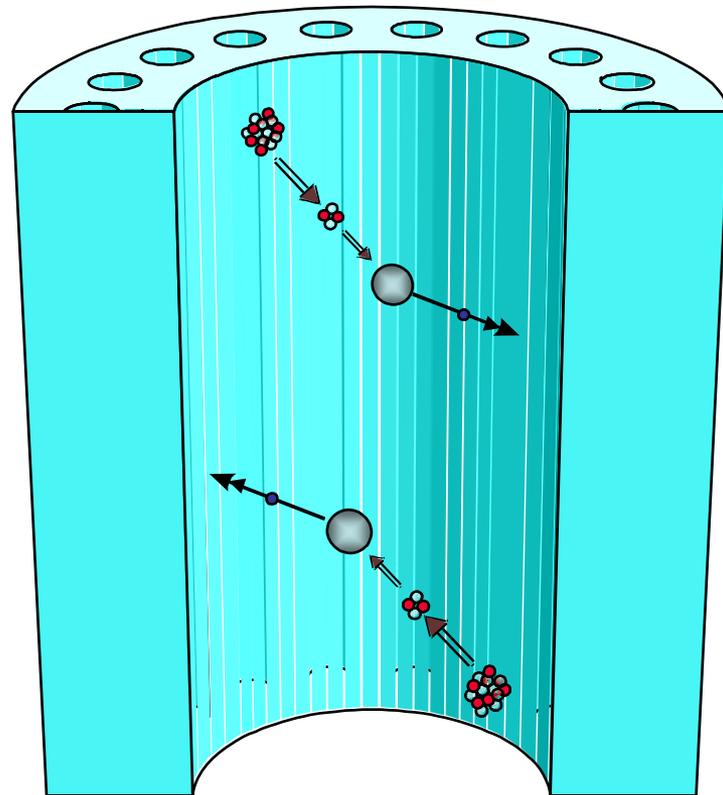
Neutron Detection - "Well Counter"

^3He neutron detectors



Detector electronics registers the coincidence and total count rates

polyethylene moderator





Moderation

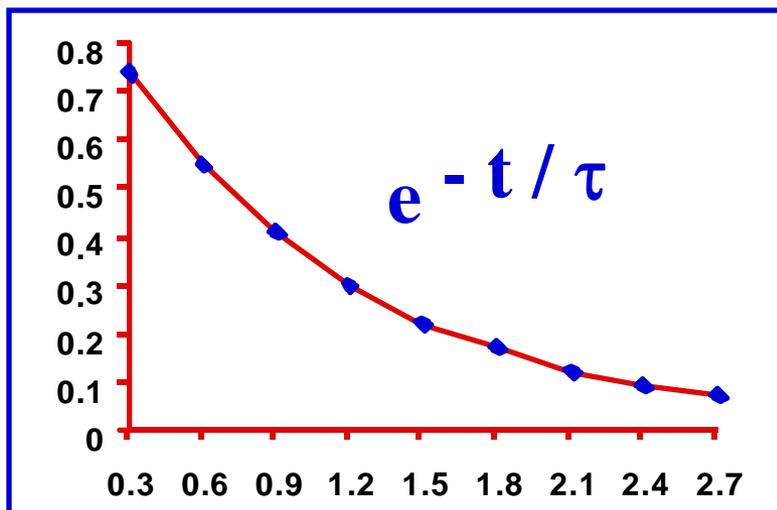
- Moderation is the process by which a neutron collides with matter and **loses energy**.
 - i.e. 2 MeV to 0.025 eV.
- The **probability of neutron detection in ^3He is largest** when the neutrons have energies near **thermal**.
- Most energy lost (best moderation) when neutron collides with nuclei of similar mass. {i.e. hydrogen (protons)}.
 - Water
 - Polyethylene
- Moderation usually takes many collisions (~27 for a 2MeV neutron in polyethylene).





Neutron Detector Die-Away

- After moderation, neutrons are lost in the detector by several processes:
 - Diffusing out of detector.
 - Diffusing to a ^3He detector tube and being absorbed.
 - Absorption by hydrogen or cadmium.
- Hydrogen both moderates and absorbs the neutrons.



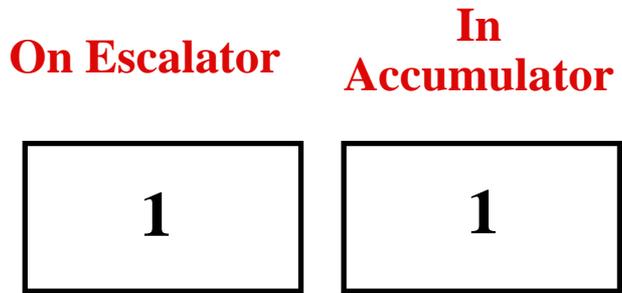
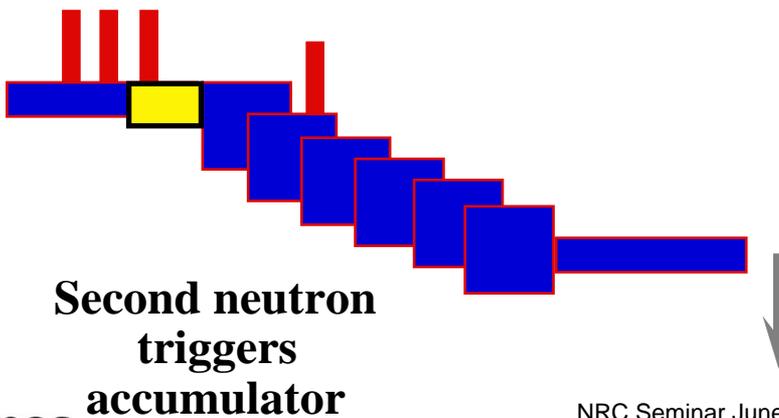
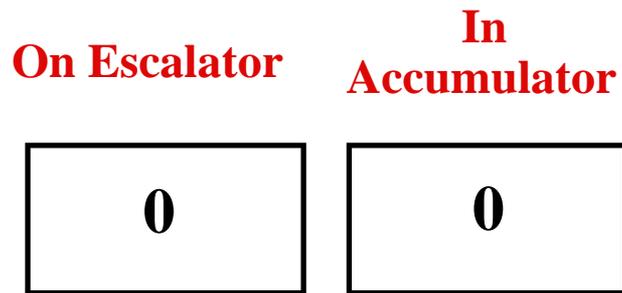
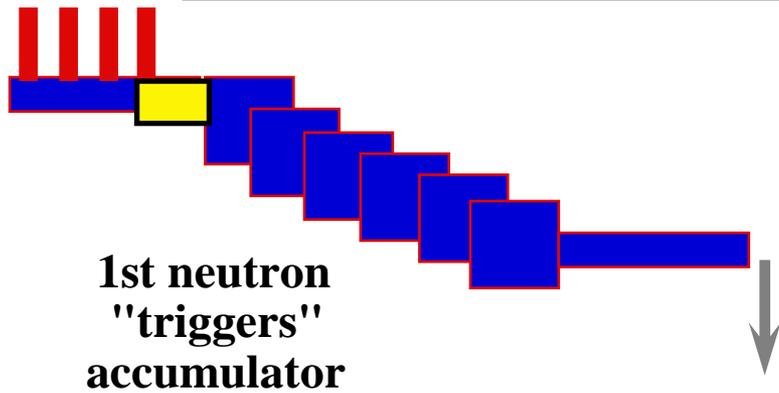
In most thermal detectors the neutron population decreases nearly exponentially in time. The time constant is called the die-away time.





The Shift Register - An Example

Four Pulses "Approach" the Escalator

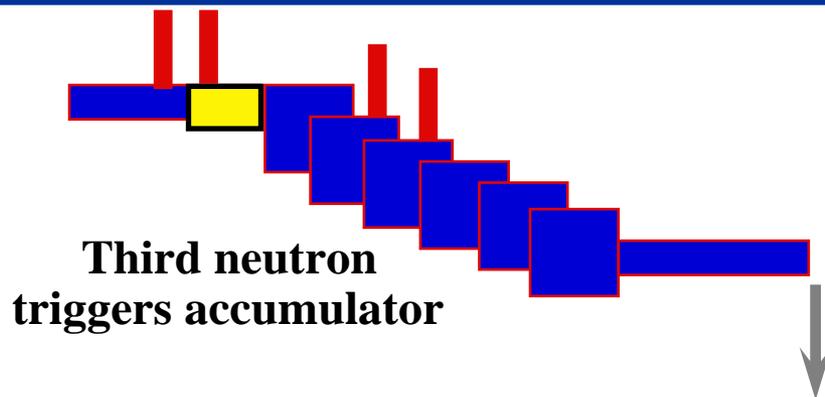


Add 1 to Accumulator





The Shift Register Example Continued



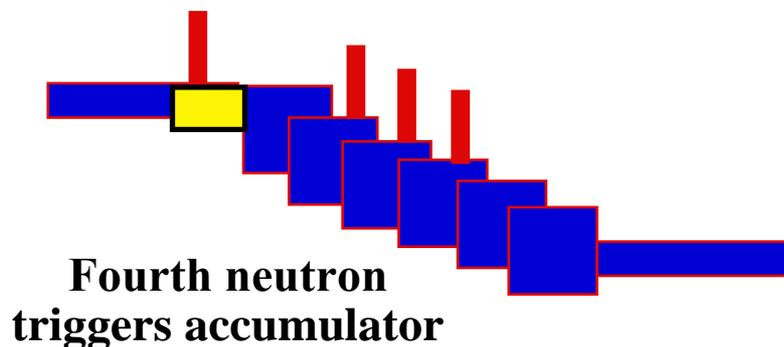
On Escalator

2

In Accumulator

3

Add 2 to Accumulator



On Escalator

3

In Accumulator

6

Add 3 to Accumulator

Four Pulses Yield 6 Coincidences





What Do We Really Count?

- The Shift Register really counts coincidences.
- The Rossi-Alpha distribution is based on counting all possible coincidences.
- The formula for all possible coincidences is:

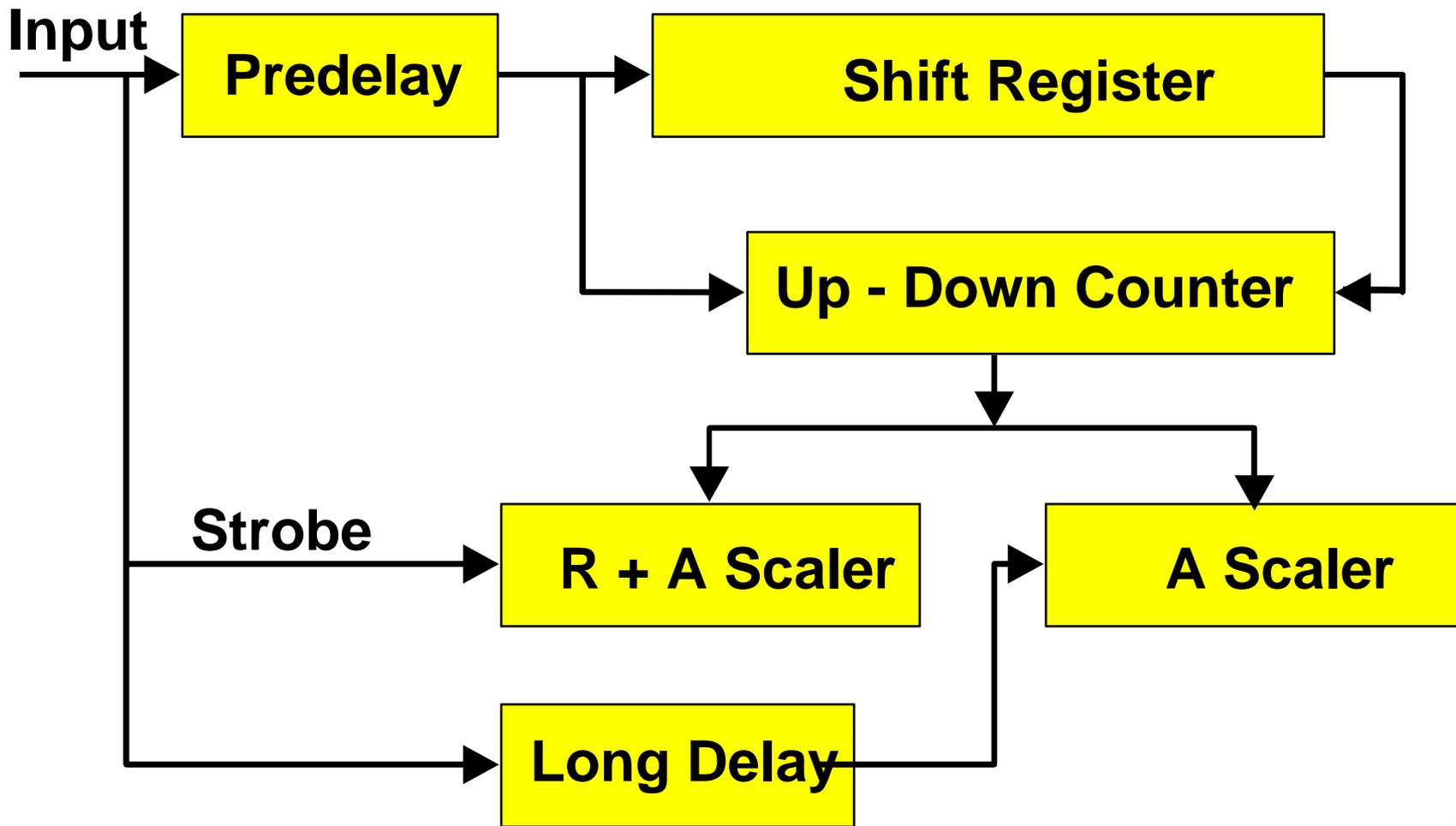
$$\frac{n(n - 1)}{2}$$

- Where n is the number of pulses.



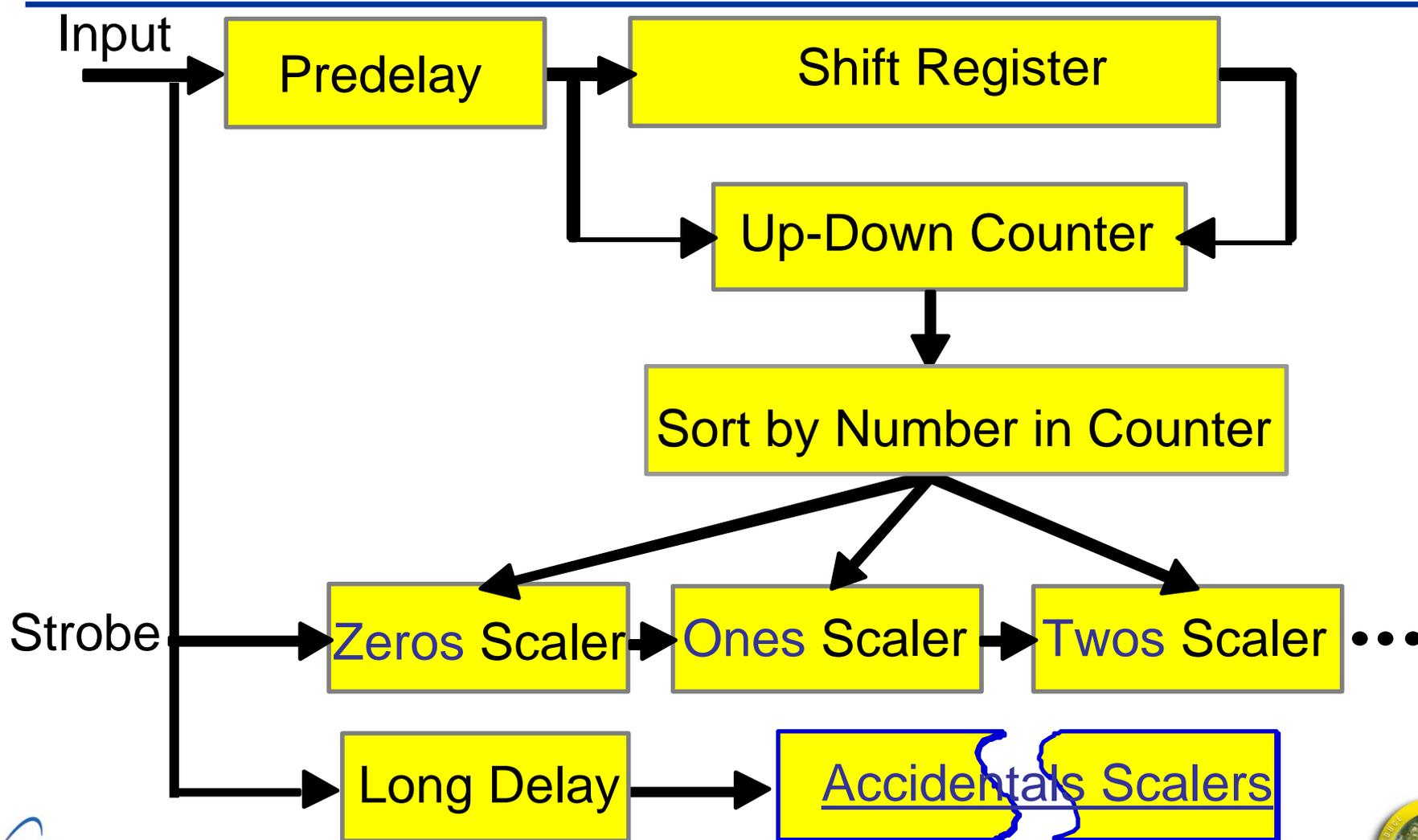


The Shift Register Circuit



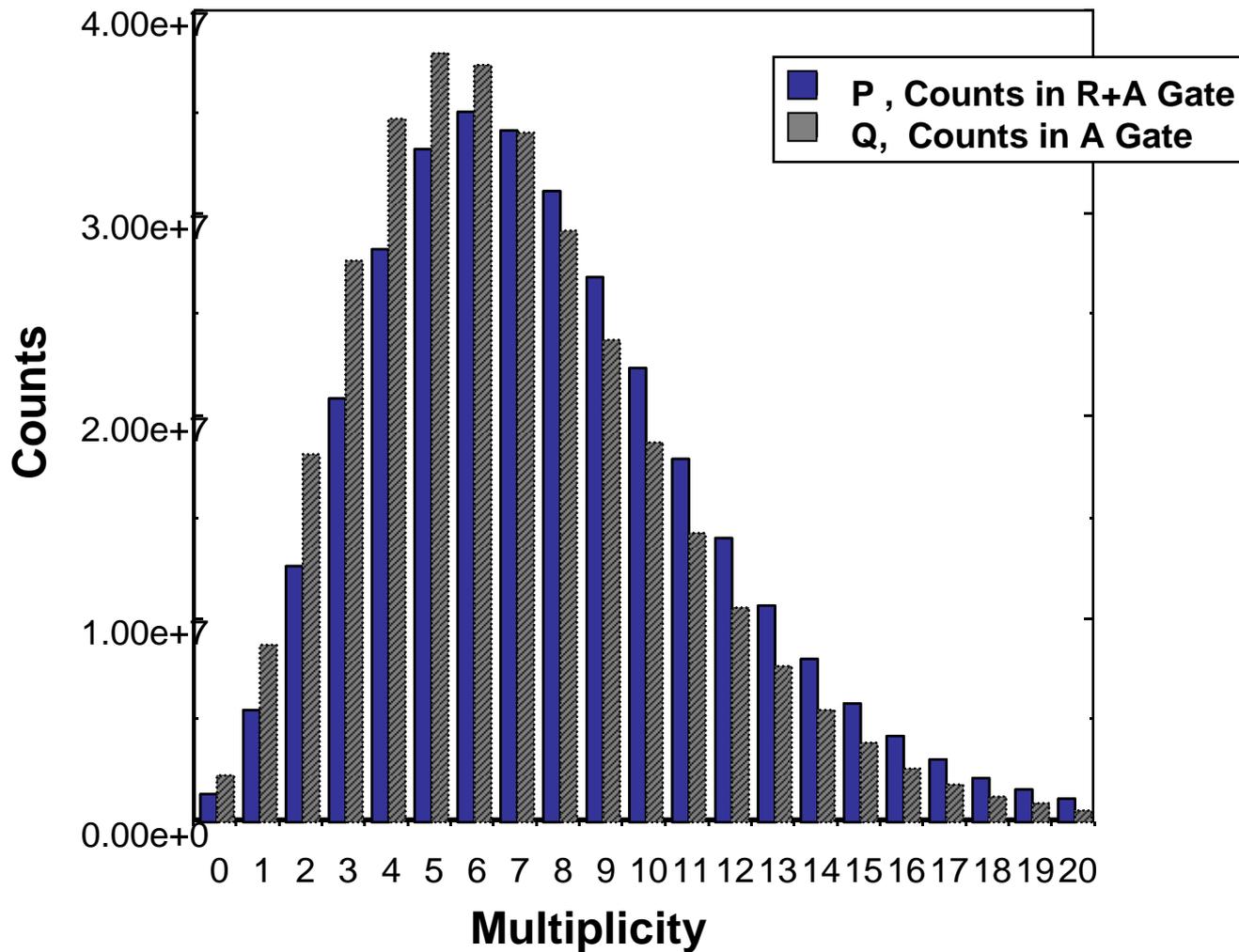


Multiplicity Shift Register



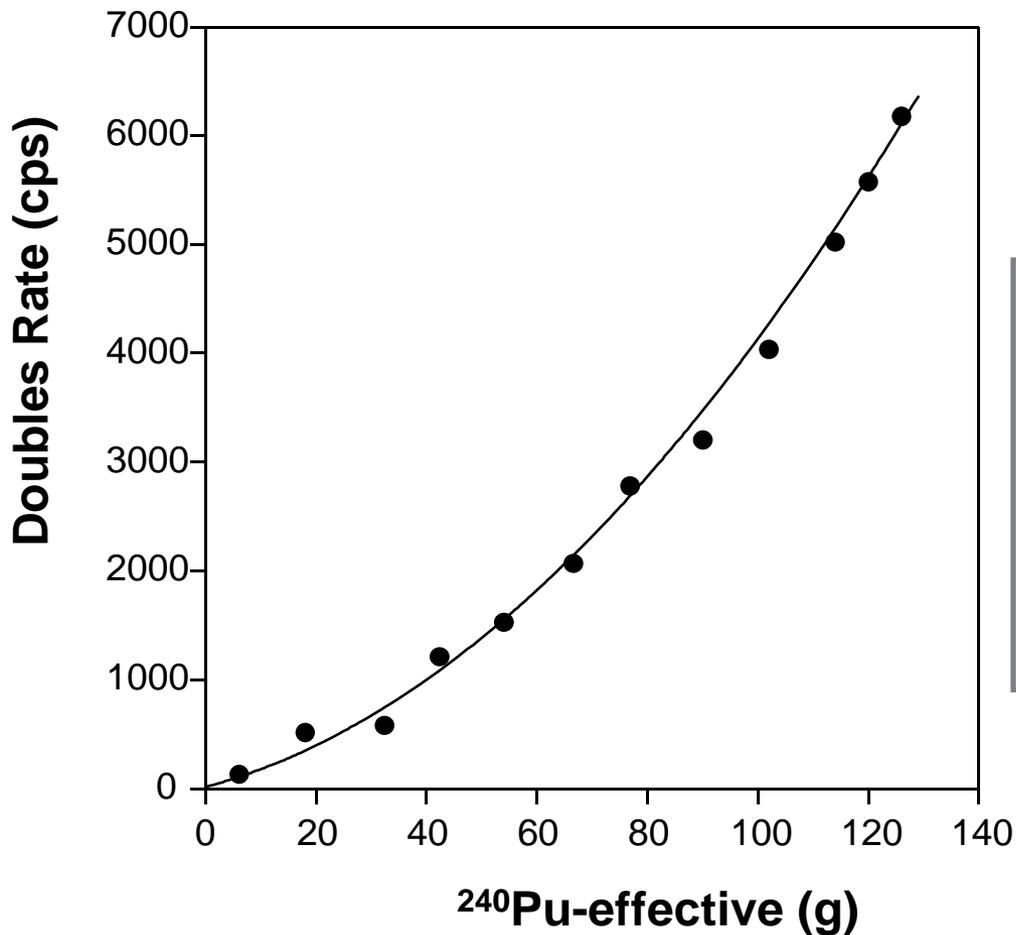


Multiplicity Distribution – 3.8 kg Pu metal





Calibration Curve Method

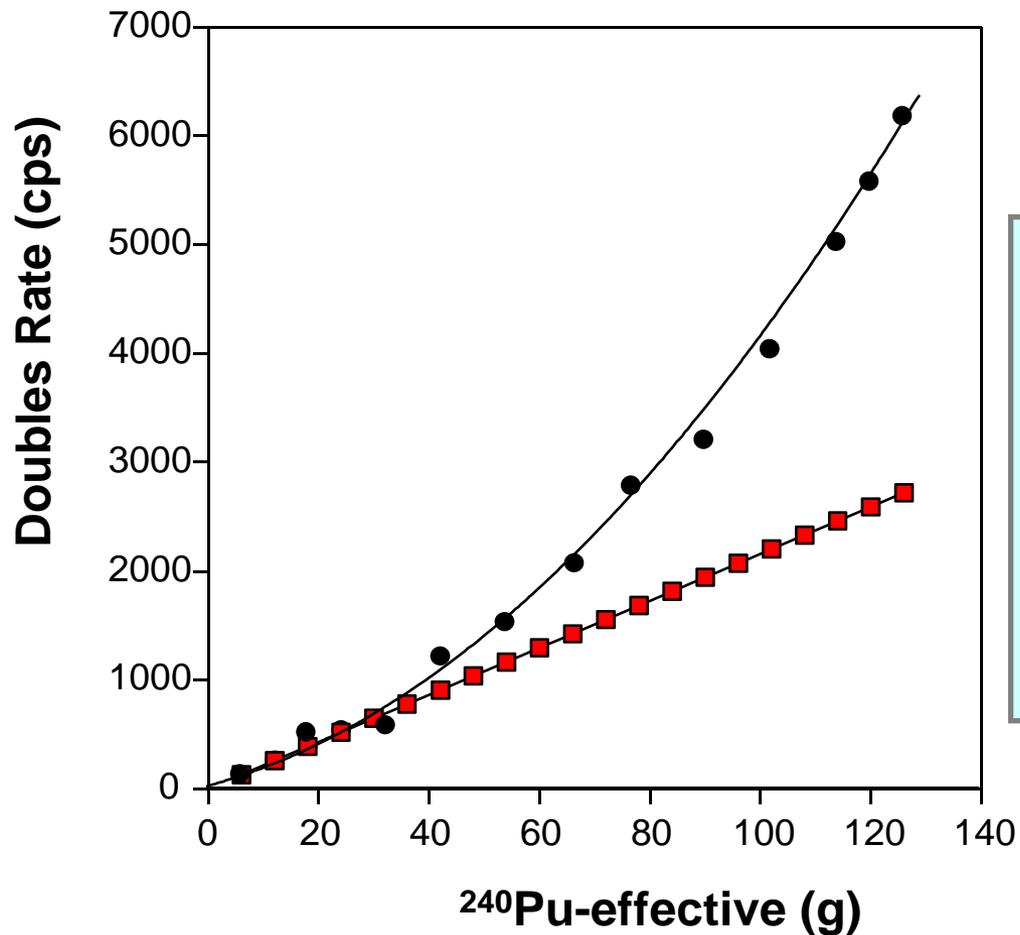


Need representative standards for each sample type and careful calibration measurements.





Known Alpha



Use singles and doubles to deduce a “multiplication” correction that linearizes the calibration. Works well for pure oxides, metals, and fluorides. Still need standards.





The Problem with Standard Coincidence Counting

There are **3 principal unknowns** in neutron counting:

^{240}Pu -effective mass, a , and M .

Standard Coincidence Counting provides only **2** pieces of measured information, singles and doubles (or totals and coincidences) To obtain an accurate assay, **one must know a lot about the sample.**

If the assumed information is incorrect, **large errors** can occur.

In Neutron Multiplicity Counting, **3** pieces of measured information are used with a **mathematical model** to deduce an assay that is far superior for most impure materials.





The Point Model Equations

$$S = F \epsilon M v_{s1} (1 + \alpha)$$

$$D = F (f_d / 2) (\epsilon M)^2 \{ v_{s2} + [(M-1)/(v_{i1} - 1)] v_{s1} (1 + \alpha) v_{i2} \}$$

$$T = F (f_t / 6) (\epsilon M)^3 \{ v_{s3} + [(M-1)/(v_{i1} - 1)] [3 v_{s2} v_{i2} + v_{s1} (1 + \alpha) v_{i3}] + 3 [(M-1)/(v_{i1} - 1)]^2 v_{s1} (1 + \alpha) v_{i2}^2 \}$$

where:

ϵ = detection efficiency

f_d = fraction of doubles in the coincidence gate

f_t = fraction of triples in the coincidence gate

F = spontaneous fission rate = 473.5 n/s/g * effective PU-240 mass

v_{n1} = average number of neutrons produced per fission event
(n=s -- spontaneous fission, n=i -- induced fission)

v_{n2} = average number of neutron pairs produced per fission event

v_{n3} = average number of neutron "triplets" produced per fission event

α = ratio of (alpha, n) neutron rate to the spontaneous fission rate

M = fission multiplication





Neutron NDA Techniques

- **Passive** coincidence counting
- **Active interrogation** with coincidence counting
- **Active** interrogation with counting of delayed neutrons





The High-Level Neutron Coincidence Counter (HLNC)

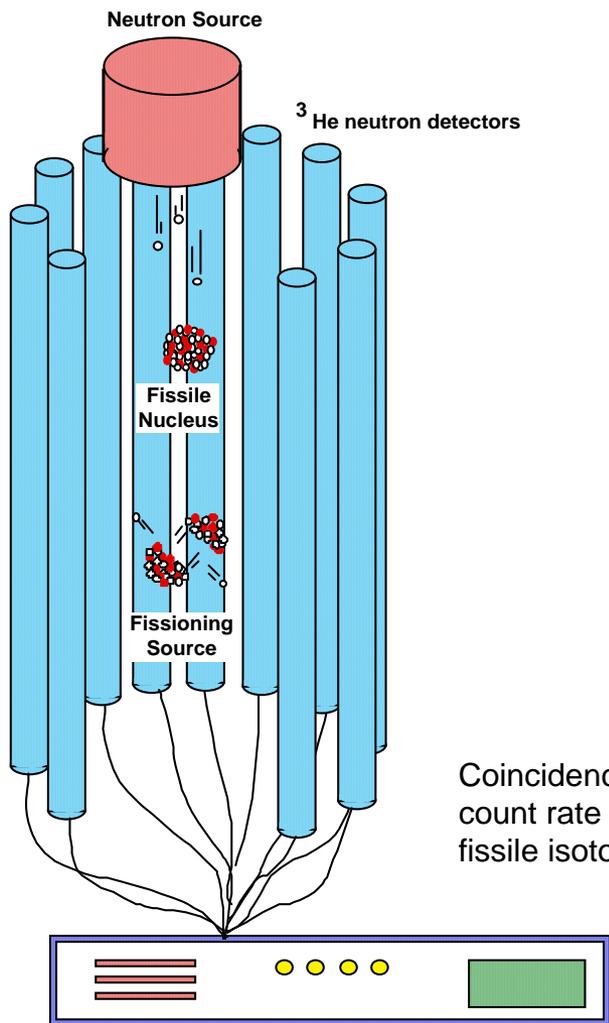


The cylindrical counter (HLNC) surrounds the sample with ^3He detectors. The data acquisition and analysis electronics are on the bench. This counter is commercially available.





Active Neutron Coincidence Counting



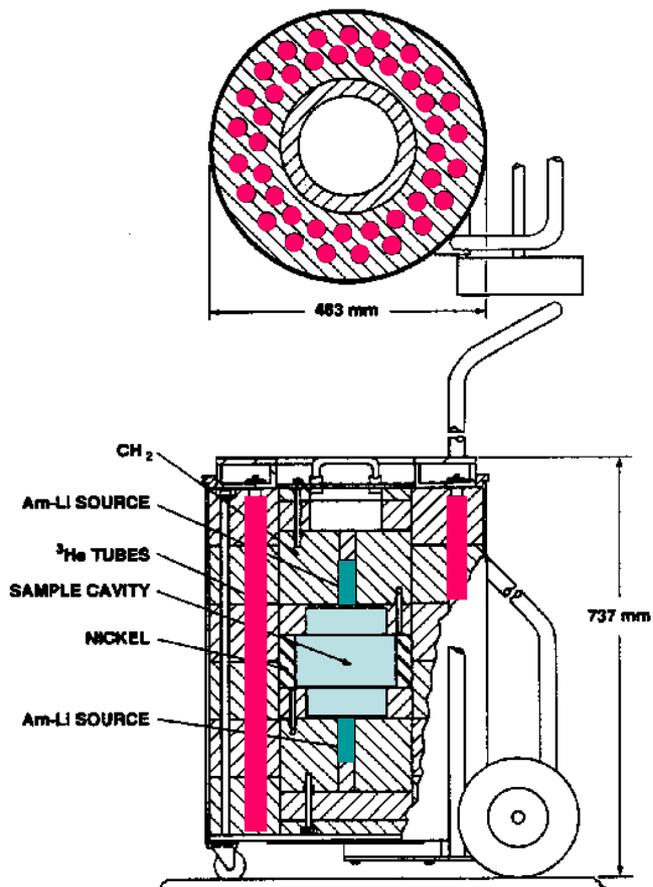
Coincidence electronics registers the coincidence count rate which is proportional to the mass of fissile isotopes

- Random (AmLi) neutron source induces fissions in fissile nuclei.
- High background, (reduced sensitivity) compared to passive counters (due to AmLi source).
- Fissioning source surrounded by neutron detectors.
- Prompt multiple neutron emission from induced fission detected as coincidence neutron events.
- Designed primarily for ^{235}U assay.





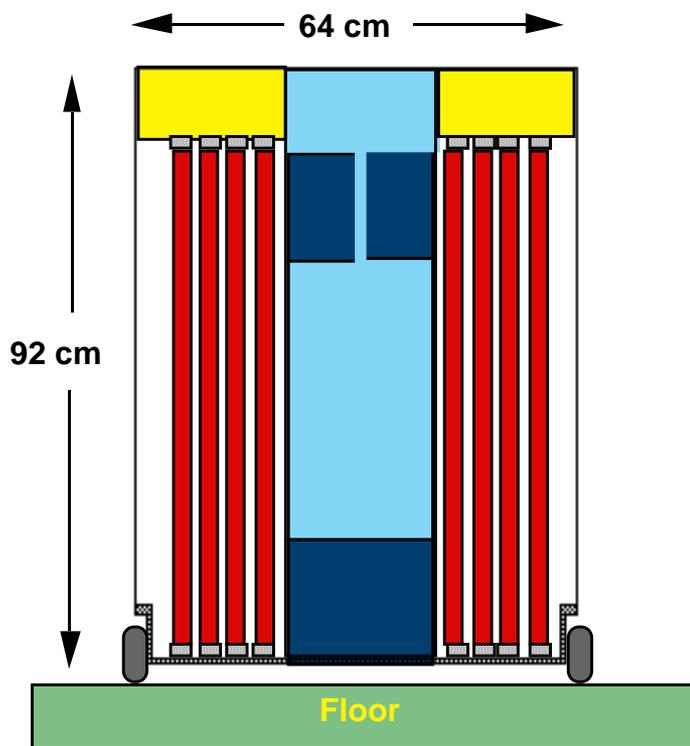
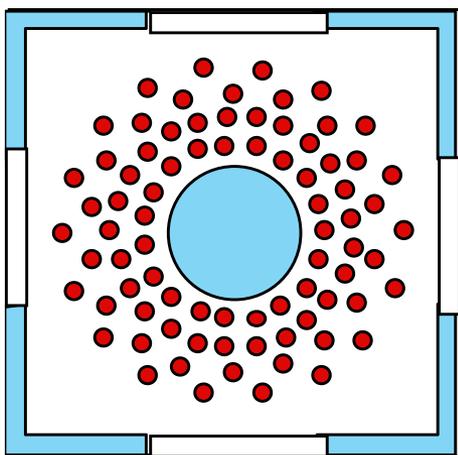
Active Neutron Coincidence Counter (AWCC)





Plutonium Scrap NMC

-  Polyethylene
-  Graphite
-  Helium-3 Tubes
-  Air
-  Junction Box

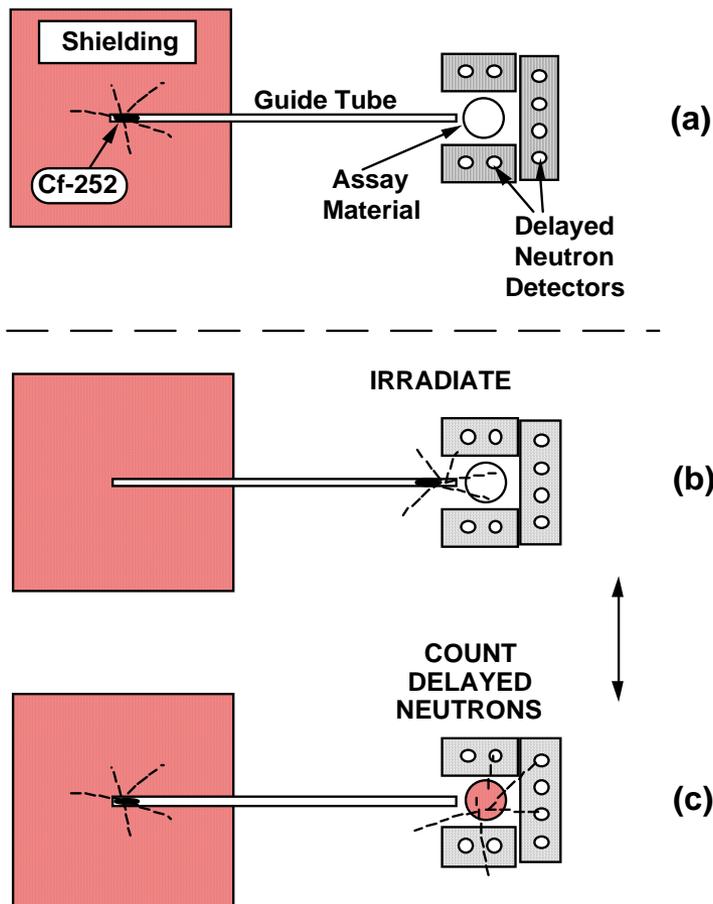


Sample Cavity is 20 cm x 41 cm
and is Cadmium Lined





The Shuffler Assay Principle



The interrogating source is stored in heavy shielding.

The interrogating source quickly moves to a position near the SNM. The source remains near the sample for a few seconds, inducing fissions.

The interrogating source is quickly moved back to its shielded position; the detectors then count delayed neutrons.

The "irradiation-count" cycle is repeated many times to acquire adequate statistical precision. The periodic motion of the interrogating source, shuffling in and out of its shielded storage, gives the "Shuffler" its name.





^{252}Cf Shuffler





Sample Characteristics for Neutron Assays

- Uniformity still important, but not always critical
- High-density
- Large size
- Heterogeneous

In addition, most samples assayable with gamma rays are also assayable with neutrons

Special Cases for Neutrons

- Low or high-density waste (small amount of SNM)
- High-gamma-yield material (e.g., spent fuel)





Problems for Neutron Assays

- Hydrogenous materials (Protons!) [**increased system response**]
 - **plastics**
 - **moisture**
 - **any other moderator**
- Absorption by high-density SNM (fissile)
 - **n capture to induce fissions** (absorption)
 - **induced fissions** (multiplication)
- Absorption by neutron poisons [**good neutron absorbers**]





Appropriate Samples for Active Neutron Coincidence Counting and Delayed Neutron Counting

- All U samples [assays mass of ^{235}U]
- Pu samples [assays mass of $^{239,241}\text{Pu}$, passive signal may complicate]
- Mixed U/Pu samples (MOX) [need passive and active measurement to sort out components]
- Low-mass or high-gamma samples (waste, spent fuel) - [delayed neutron counting (Shufflers)]





Neutron NDA Calibration

- Philosophy the same as for gamma NDA
 - **Measure standards**
 - **Apply all relevant corrections**
- Best to calibrate over a range of expected sample characteristics





Neutron Calibration Standards

- Standards' characteristics:
 - Range of **SNM masses**
 - Range of **matrices**
 - Range of **material types**

- Establishes response variations from:
 - **Moderation**
 - **Multiplication**
 - **Absorption**





Measurement Precision in Neutron NDA

- Origins
 - Random decay processes (counting statistics)
 - Fluctuating neutron backgrounds
- Strategies to improve precision
 - Longer count times (improve statistics)
 - Background/source control
 - [Active] Longer count times for interrogation source
 - [Shuffler] Careful mechanical design and many count cycles minimize source placement problems.





Measurement Bias in Neutron NDA

- Origins
 - **Calibration uncertainty**
 - **Chronic sample mis-placement in measurement well**
 - **Altered neutron backgrounds (singles and coincidence)**
- Strategies to minimize bias
 - **Calibrate often and with high precision.**
 - **Background/source control**





Measurement Control for Neutron NDA

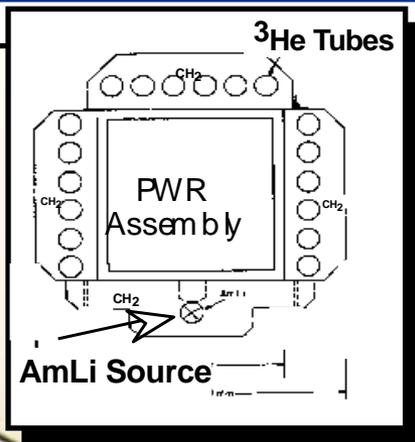
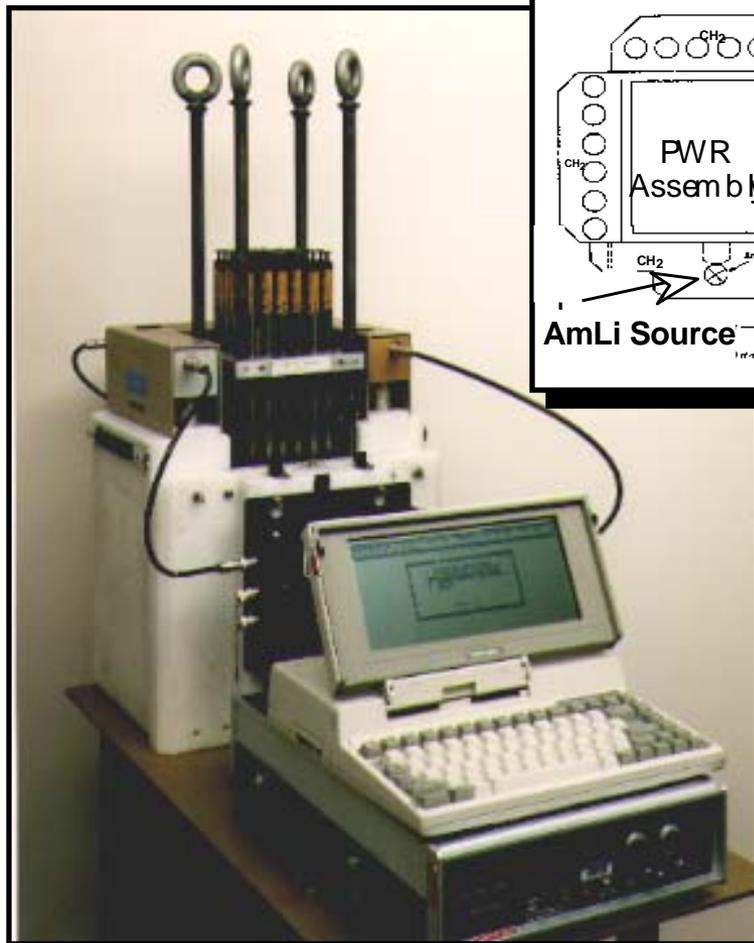
- Philosophy — still the same as for γ -ray measurements
 - Periodically check quality of data and instrument.
 - Periodically measure measurement control standard.
 - Check precision by repeated measurements of standard.
 - Check bias by verification of average standard value against declared value.

- Measurement Control standards for neutron NDA:
 - Stable SNM (encapsulated metal, sealed oxide, ...)
 - ^{252}Cf source (fission neutrons, decay rate known)
 - [AWCC] AmLi source built in + SNM sample
 - [Shuffler] Calibrated standard





Active Neutron Coincidence Collar



The active Neutron Coincidence Collar is shown measuring a LWR fuel assembly. The collar surrounds the assembly on 3 sides with ^3He tubes and functions like a conventional coincidence counter. On the 4th side of the assembly, an AmLi random neutron source is positioned to interrogate the sample. The collar operates like an AWCC, with the assay result giving fissile content per unit length along the fuel assembly. For MOX fuels, the collar can be operated in both active and passive mode. Also, the collar can also be used in a passive mode for ^{238}U .





Neutron NDA Summary

■ Neutron NDA Techniques:

- **Passive coincidence, active coincidence, delayed n**
- **Absorption, multiplication, moderation corrections can be important**
- **(α ,n) interactions can affect results strongly (background, bias)**
- **Appropriate samples: minimum hydrogen (moderator); passive - Pu, active - U; high-sensitivity (waste) - passive or delayed n.**
- **Calibration curves of families of sample types recommended**

■ Accuracies Possible:

- **1% to 50%, depending on sample composition and technique**
- **Best performance on dry, dilute samples (no moderation, multiplication)**
- **Worst cases: samples with varying moisture, moderation, matrix.**



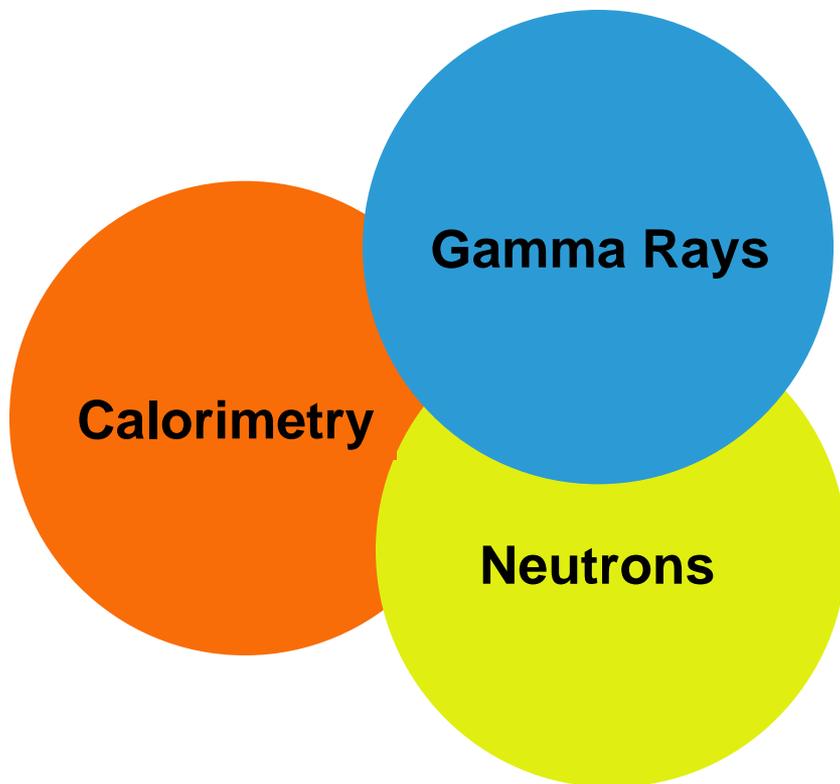


Calorimetry





Calorimetry



Measurement Principle:

Determination of Heat output of the sample, as an indication of the **amount of heat-producing isotopes** of SNM in the sample.





Why Calorimetry?

- The most **accurate** NDA measurement (<1% uncertainty) of nuclear material
- You **can't hide** the heat of the material
 - **No absorption, moderation, multiplication,...**
- Inherently matrix independent
 - **Assay result independent of material, matrix or container type**
 - **matrix can only affect duration of analysis**
 - **No physical SNM standards needed!**
- NIST-traceable technique
- Most DOE facilities are using calorimetry
 - **Hanford, PNL, RF, LANL, SRS, LLNL, ANL-W...**





Uses of Calorimetry

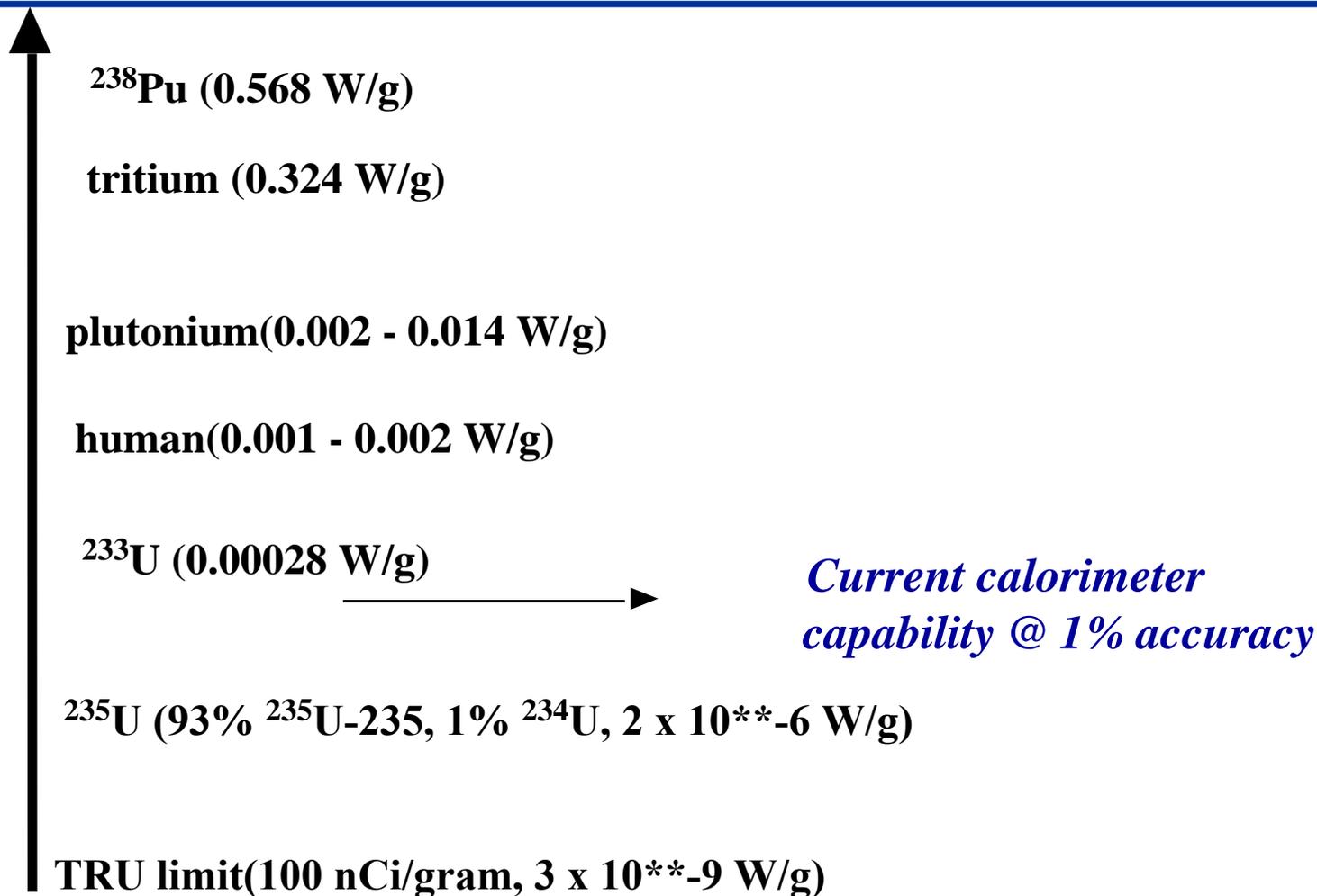
- **Accountability Measurements**
- **Verification measurements**
- **Shipper receiver measurements**
- **Calibration of NDA working standards**
- **Measurement of biases and precisions of other NDA techniques**
- **Resolution of assay “outliers” and problem samples**
- **Benchmarking NDA measurement campaigns**
- **Process control measurements**
- **Product acceptance measurements**

Considered the “gold standard” of NDA measurements throughout the complex





Relative Thermal Powers





Fundamental Measurement Characteristics

- Precise NDA measurements **based on heat output of sample**
 - ***measurement accuracy of 0.1% to 0.5% for most cases***
 - ***best suited for plutonium samples***
- Sealed containers
- Assay results independent of matrix material
- **Requires isotopics to convert power to grams**
 - ***isotopics from: Gamma ray (NDA) or Chemistry (DA)***
- Integrates over total sample volume
- Faster than DA, slower than NDA (γ or n)





Half-lives and Specific Powers

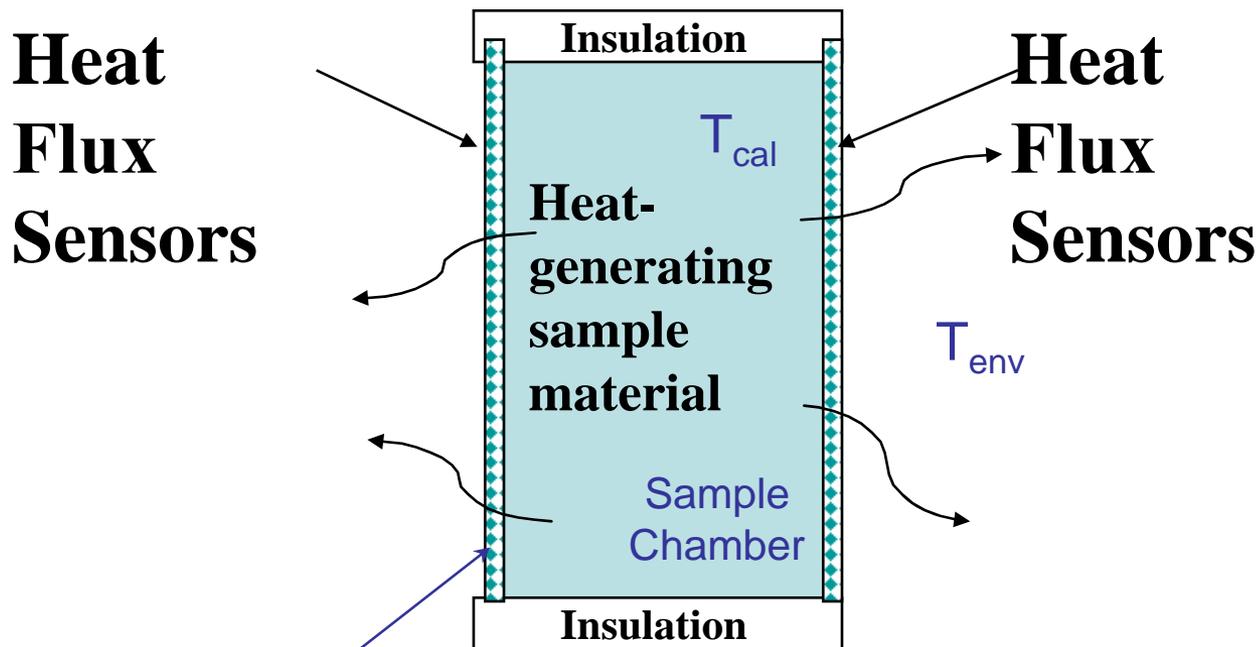
Isotope	Half-life (years)	Specific Power (Watts/gram)
Pu-238	87.74 ₀₄	0.56757 ₂₆
Pu-239	24119 ₂₆	0.0019288 ₀₃
Pu-240	6564 ₁₁	0.0070824 ₂₀
Pu-241	14.348 ₂₂	0.003412 ₀₂
Pu-242	376300 ₉₀₀	0.0001159 ₀₃
Am-241	433.6 ₁₄	0.1142 ₀₅
H-3	12.3232 ₄₃	0.3240 ₀₉





Calorimeters measure the flux through heat sensors of thermal energy generated by radioactive decay

$$dQ/dt = (T_{cal} - T_{env}) / R_{Th}$$

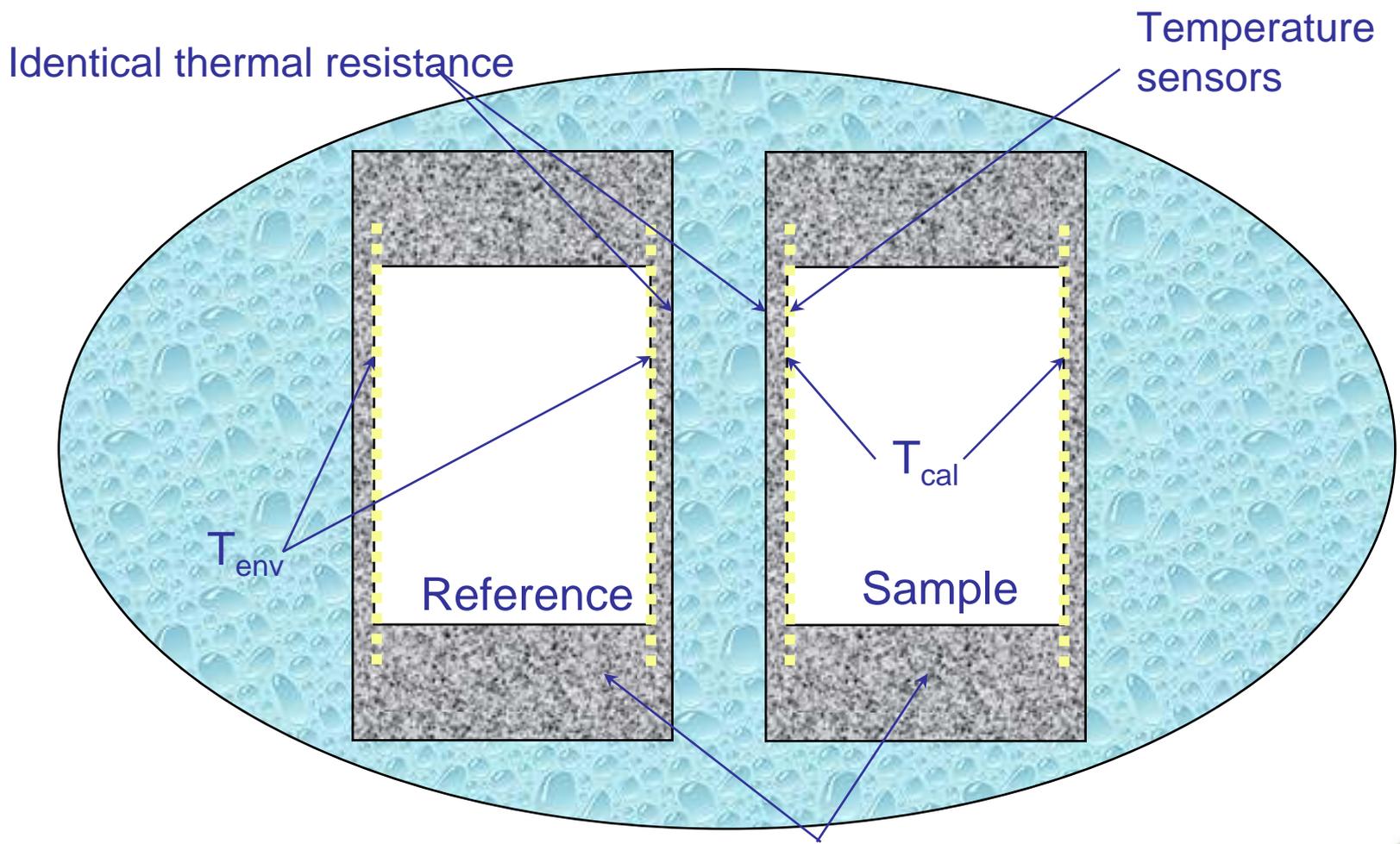


R_{Th} = Thermal Resistance





Twin Bridge Schematic



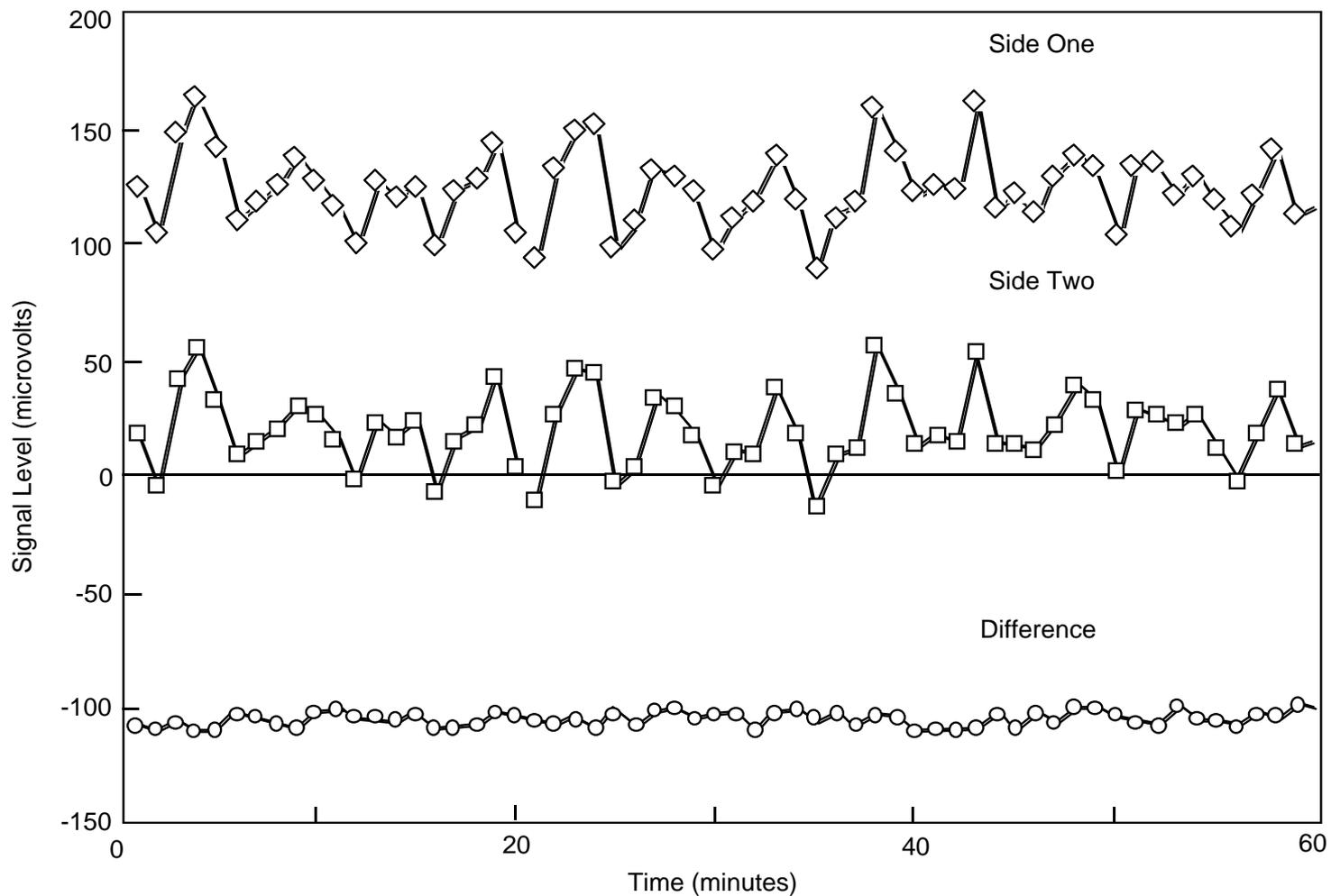
Thermal Resistance of ends increased relative to sides

NRC Seminar June 10, 2008



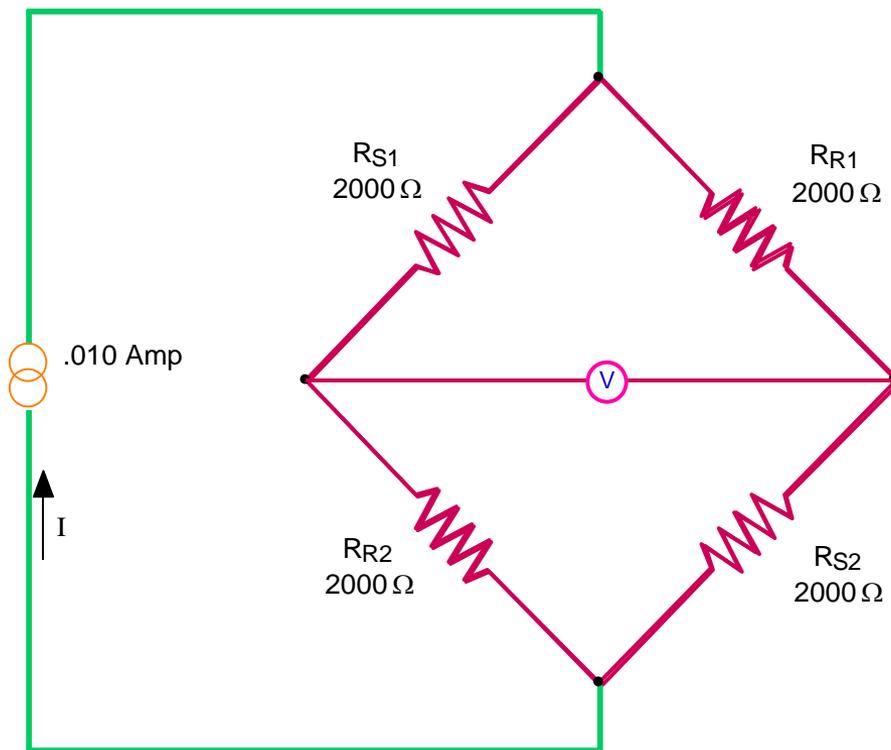


Why Twin Design?



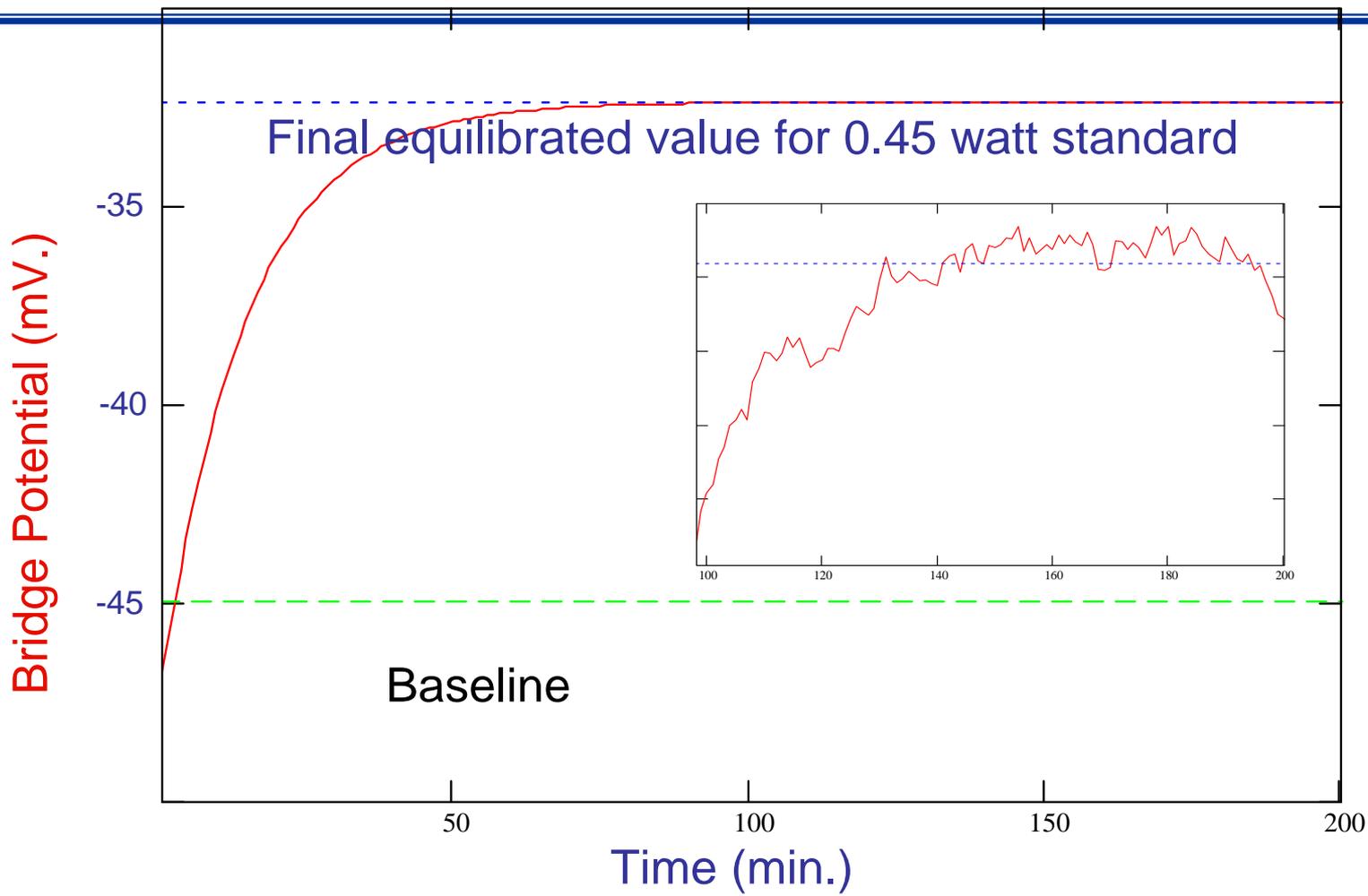


Resistance change measured by Wheatstone Bridge



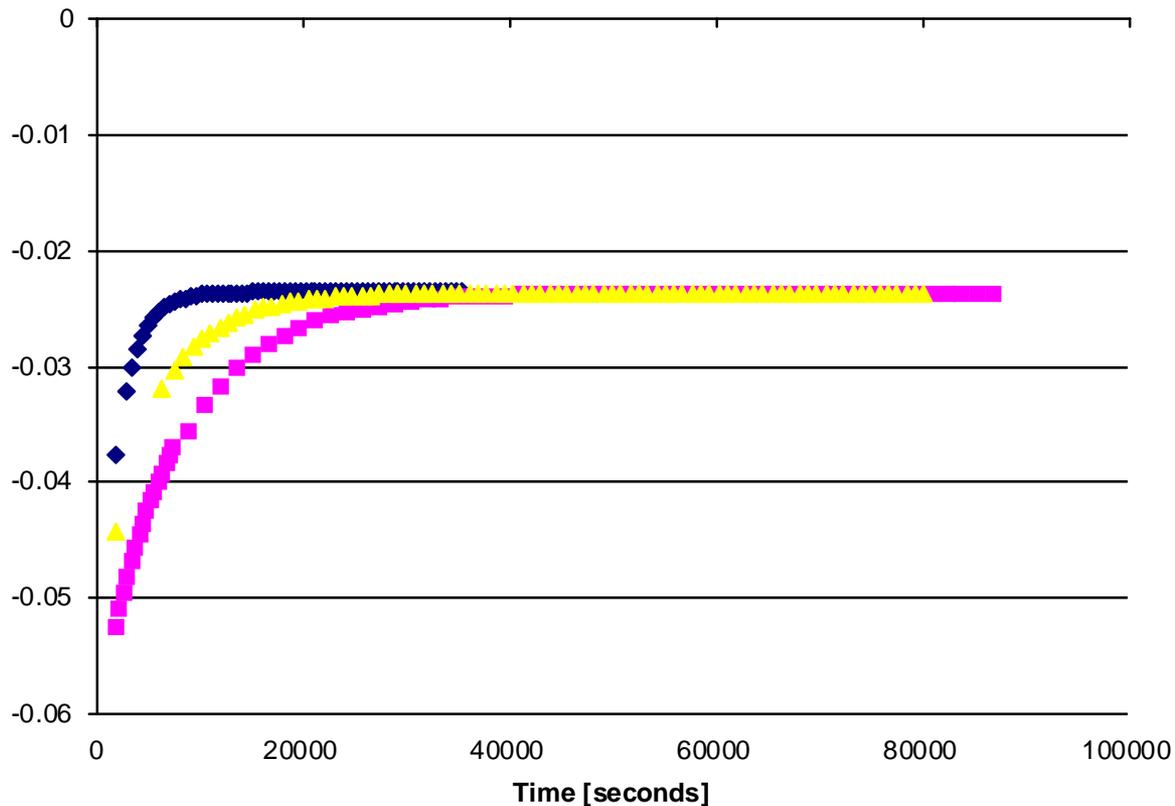


Wheatstone Bridge Calorimeter





Calorimeter sensor output independent of matrix



- ◆ 27Jun Al foil2 1.0wj
- 4Jul Cu shot & 1.0wj
- ▲ 8Jul salt & 1.0wj





Calorimetric Assay of SNM (mass)

- Equation for total SNM mass

$$Mass(grams) = \frac{Thermal\ Power\ (Watts)}{P_{eff}\ (Watts\ /\ g)}$$

- Measure isotopic composition of item (high resolution gamma spectroscopy or mass spectroscopy) and compute Effective Specific Power, P_{eff}

$$P_{eff}\ (Watts\ /\ g) = \sum P_i * f_i$$

P_i = Isotope specific power (Watts/g)

f_i = Isotopic fraction in sample, relative to plutonium

i = all isotopes present (i.e. ^{238}Pu , ^{239}Pu , ..., ^{242}Pu , ^{241}Am)





Effective Specific Power:

The rate of energy emission per unit mass of plutonium at the time of measurement.

$$P_{\text{eff}} = \sum_{i=1}^n R_i P_i$$

Where:

- there are n isotopes in the sample
- R_i is the mass ratio of each isotope present
- P_i is the specific power of each isotope

- Isotopic information not required for monoisotopic items (e.g. ^3H or ^{241}Am)

^{238}Pu and ^{241}Am can be important because of their very high specific heat values. ^{239}Pu can be important because of its high abundance in most samples.





Calibration and Standards

- No SNM standards representative of material type are needed
 - **Electrical standards**
 - **Heat standards**
- HEAT output is measured, and heat is not absorbed or otherwise lost, unlike neutron or gamma NDA methods.





Situations that degrade Ca/Iso measurement performance

- Distortions to gamma-ray isotopic assay
 - Separated Am-241 and Pu, each in different matrices
 - Gamma-ray interferences
 - Pu-241/ U-237 equilibrium
 - Inhomogeneous isotopic distribution
- Distortions to calorimetry
 - Chemical reactions
 - Power emitting isotopes with no gamma-ray [e.g., ^{90}Sr , ^3H]





Calorimeter Performance

- **Plutonium:**
 - **0.2% to 0.5% on power measurements**
 - **0.5% to 1.0% on specific power**
- **Bulk HEU (multi-Kg):**
 - **1 to 3% on power measurements**
 - **0.5 to 1.0% on specific power**
- **Measurement time:**
 - **20 min. preconditioned high conductivity sample**
 - **2-4 hours typical samples**
 - **8-12 hours insulated samples (e.g., salts)**





Appropriate Samples For Calorimetry

- Measurable heat outputs
 - **Pu samples**
 - **High-mass U samples**
 - **Tritium**
- Good heat-transfer characteristics
 - **Time to thermal equilibrium**
 - **Throughput considerations**





Calorimetry equipment



Calorimeter used for standards verification



LANL-customized calorimeter



Antech Inc. calorimeter





Calorimetry NDA Summary

■ Calorimetry NDA Technique

- **Measure heat output from the sample**
- **Measure specific power of the sample**
- **Appropriate samples: heat-emitting (Pu, large U, T)**

■ Accuracies Possible [rivals chemistry]

- **Power: 0.1% to 0.5% (1-8 hr assay times)**
- **Specific power (0.5% to 1%)**
- **Best performance on isotopically homogeneous samples**
- **Most useful on heterogeneous NM samples**



Advanced Fuel Cycle Safeguards



Michael Miller
GNEP Safeguards Campaign Director
Los Alamos National Laboratory

U.S. Nuclear Regulatory Commission
Rockville, MD
June 10, 2008



Outline

- **Introduction and Current Technical Basis**
- **Reprocessing**
- **Fuel Fabrication**
- **Fast Reactor**



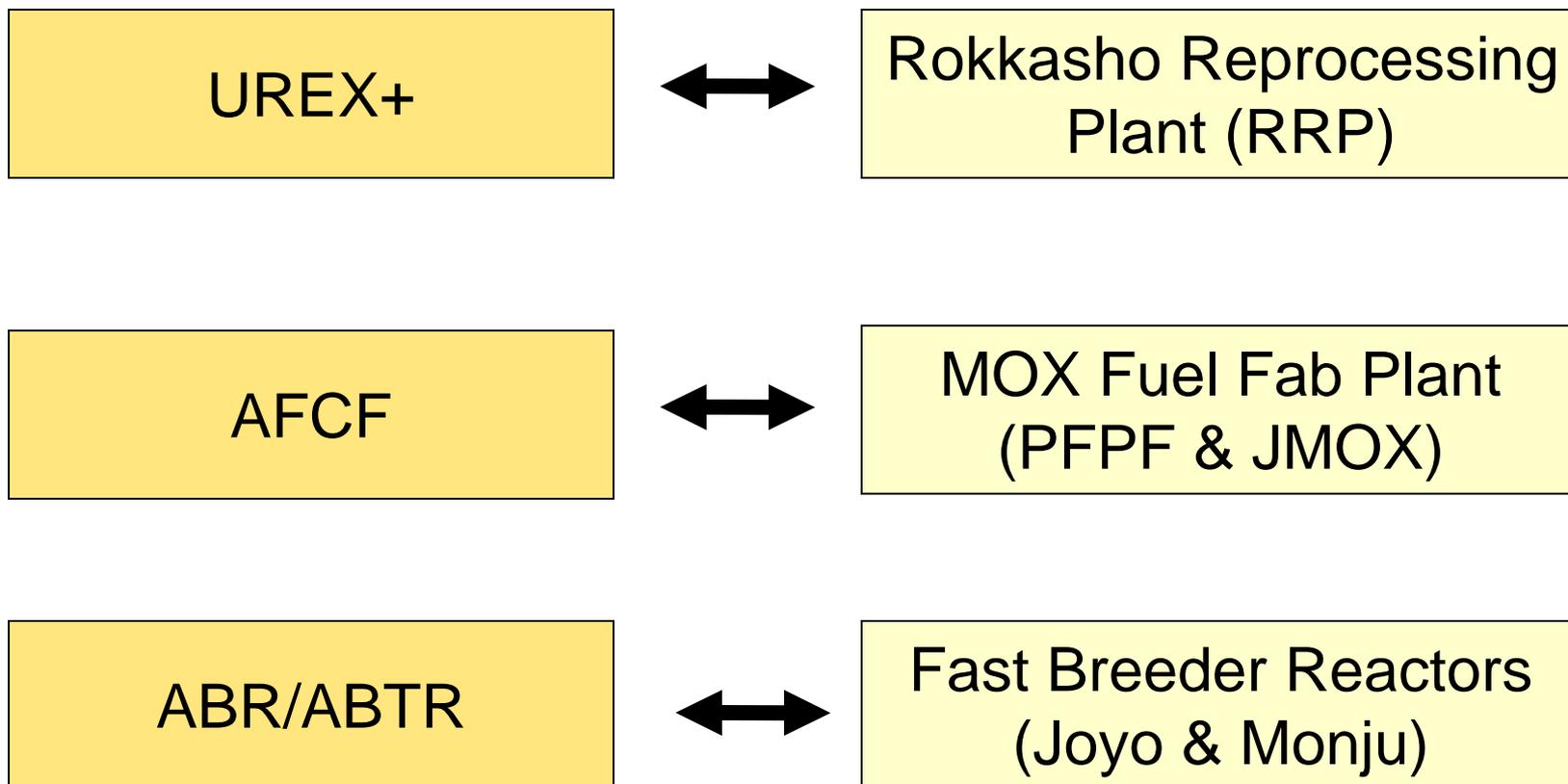


Safeguards for Advanced Fuel Cycle Facilities





Safeguards Applications in Advanced Facilities





Reprocessing





Rokkasho Reprocessing Plant (RRP)



South Side

Features common to UREX

- Spent fuel storage pool
- Head-end shear and dissolver
- Input accountability tank (IAT)
- Mixture with Pu in product
- High level liquid waste (vitrification)
- Solid wastes

Safeguards Features

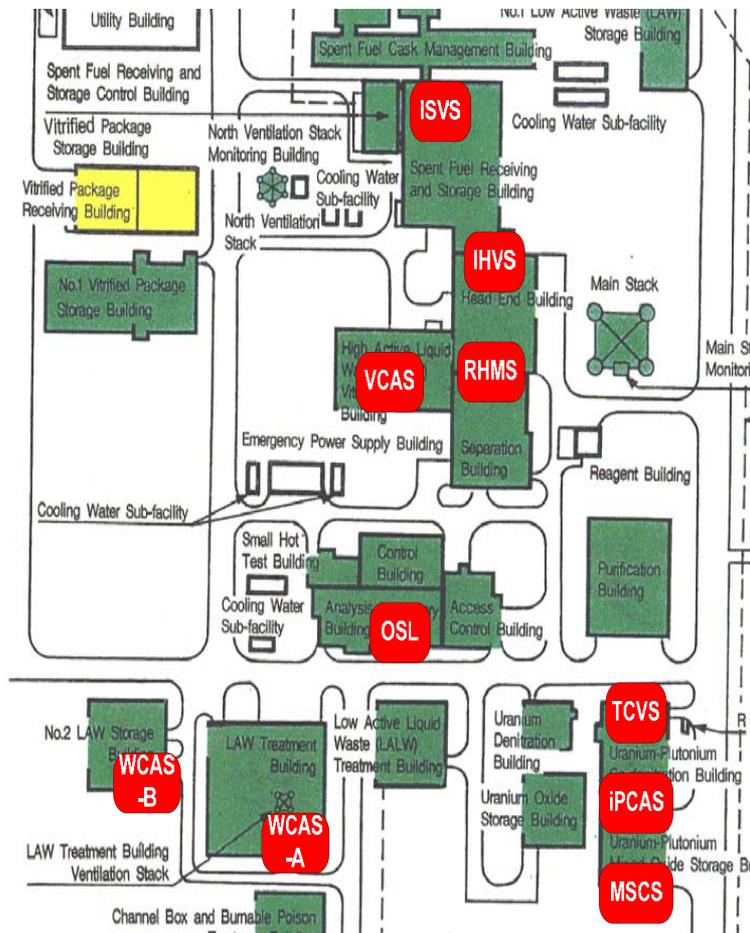
- Remote, unattended monitoring
- Near real-time accounting (NRTA)
- Process monitoring
- Data authentication
- Containment and surveillance
- On-site inspector presence
- Approx. 20 different SG systems





NDA Systems Installed Throughout the Process

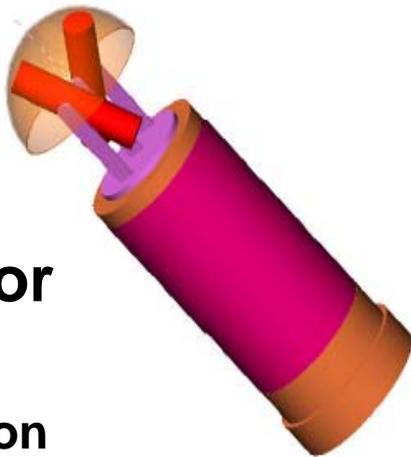
- ISVS – Integrated Spent fuel Verification System
- IHVS – Integrated Head end Verification System
- RHMS – Rokkasho Hulls Measurement System
- VCAS – Vitrified waste Canister Assay System
- HKED – Hybrid K-Edge Densitometer
- TCVS - Temporary Canister Verification System
- iPCAS - improved Plutonium Canister Assay System
- WCAS A/B - Waste Crate Assay System



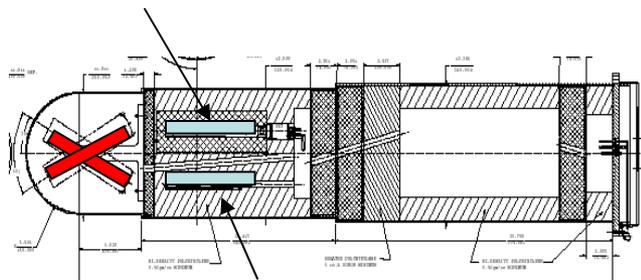


Head-End Fuel Assembly Verification Camera & Radiation Detector (CRD)

**CRD
Detector**

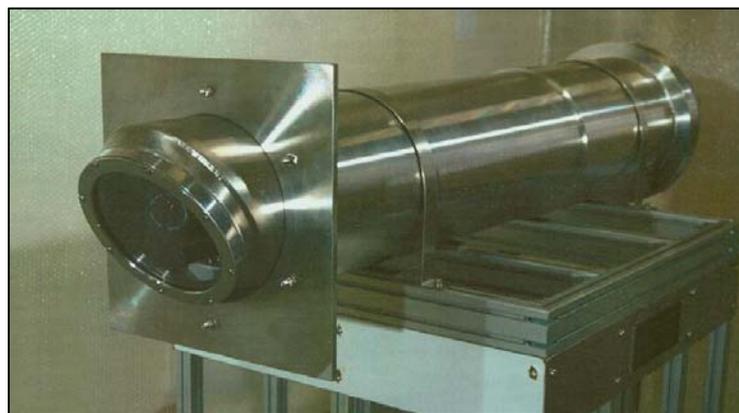


**Neutron
detector**



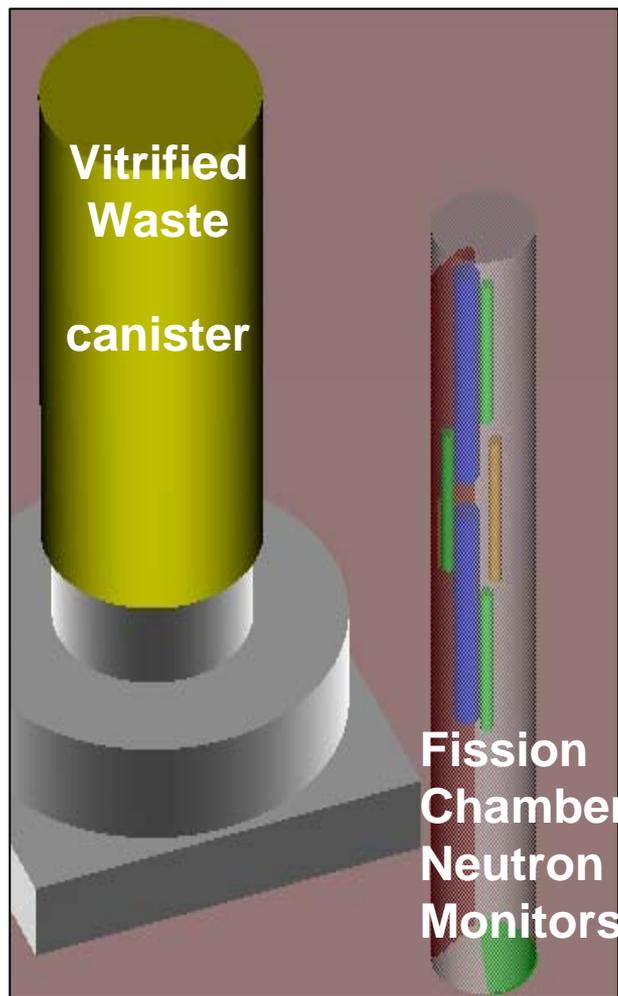
**Ion
chamber**

- Verification of LWR fuel assemblies inside shearing cell
- Penetrates the 2-meter thick concrete shielding wall
- Shielded detector head includes cameras, neutron and gamma sensors
- Provides ID, type fuel, and shearing history
- Continuous operation by IAEA and Operator



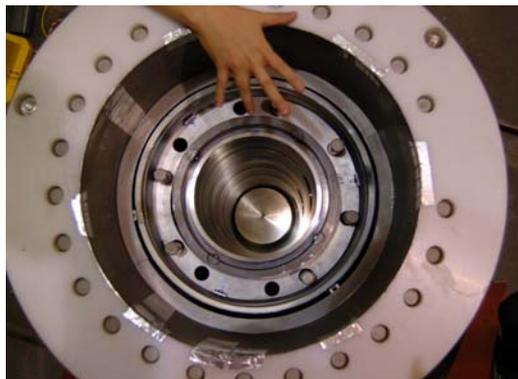


Verify Quantity of Pu in Waste Streams

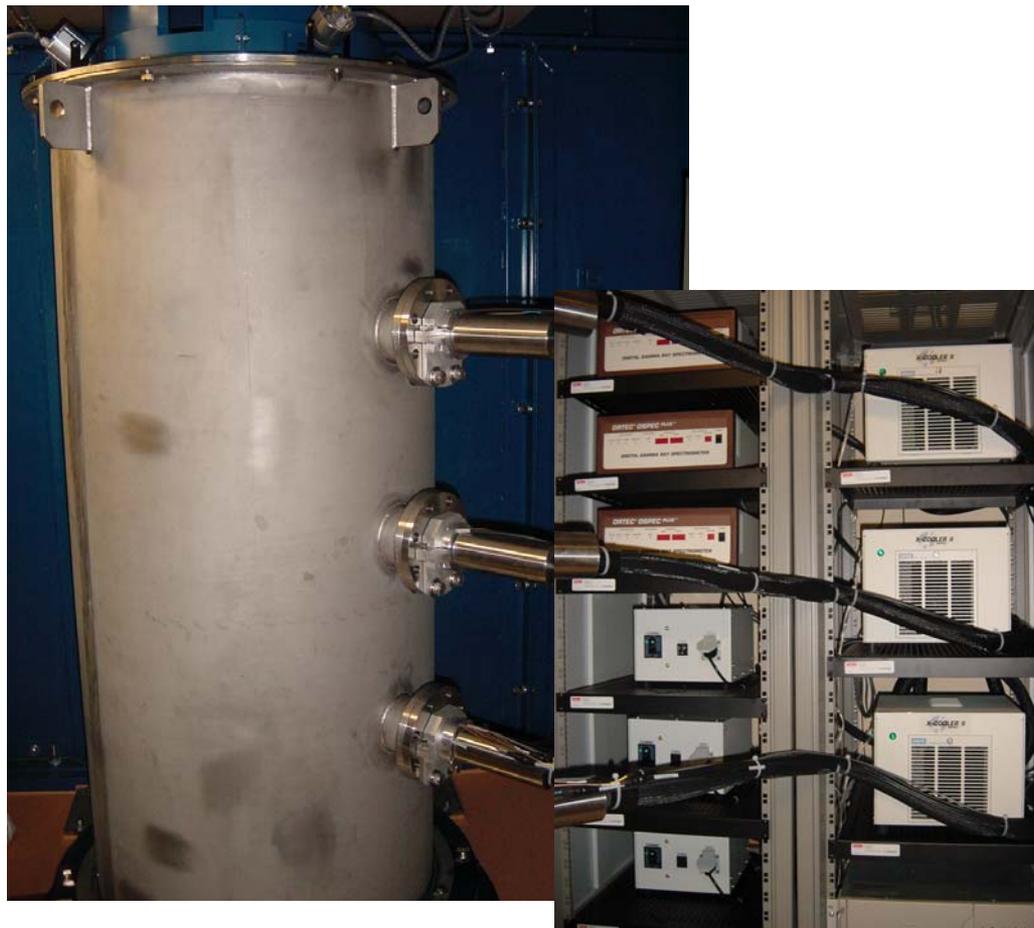




Example of impact of new safeguards measurement technologies: improved Plutonium Canister Assay System (iPCAS)



**Installed in RRP
March 2004:
decreased the
uncertainty of each
measurement (36kg
containers of MOX)
by ~200g Pu**



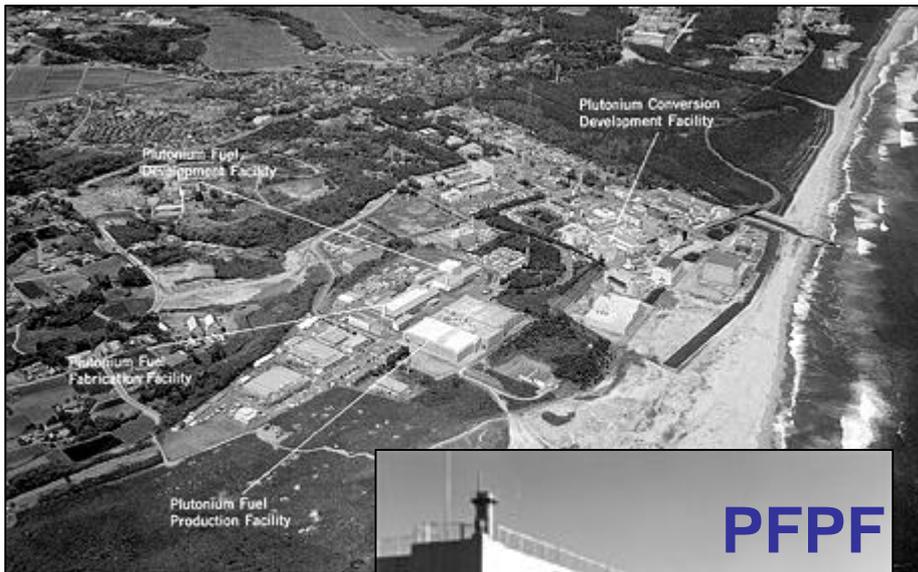


Fuel Fabrication





Plutonium Fuel Production Facility (PFPF)



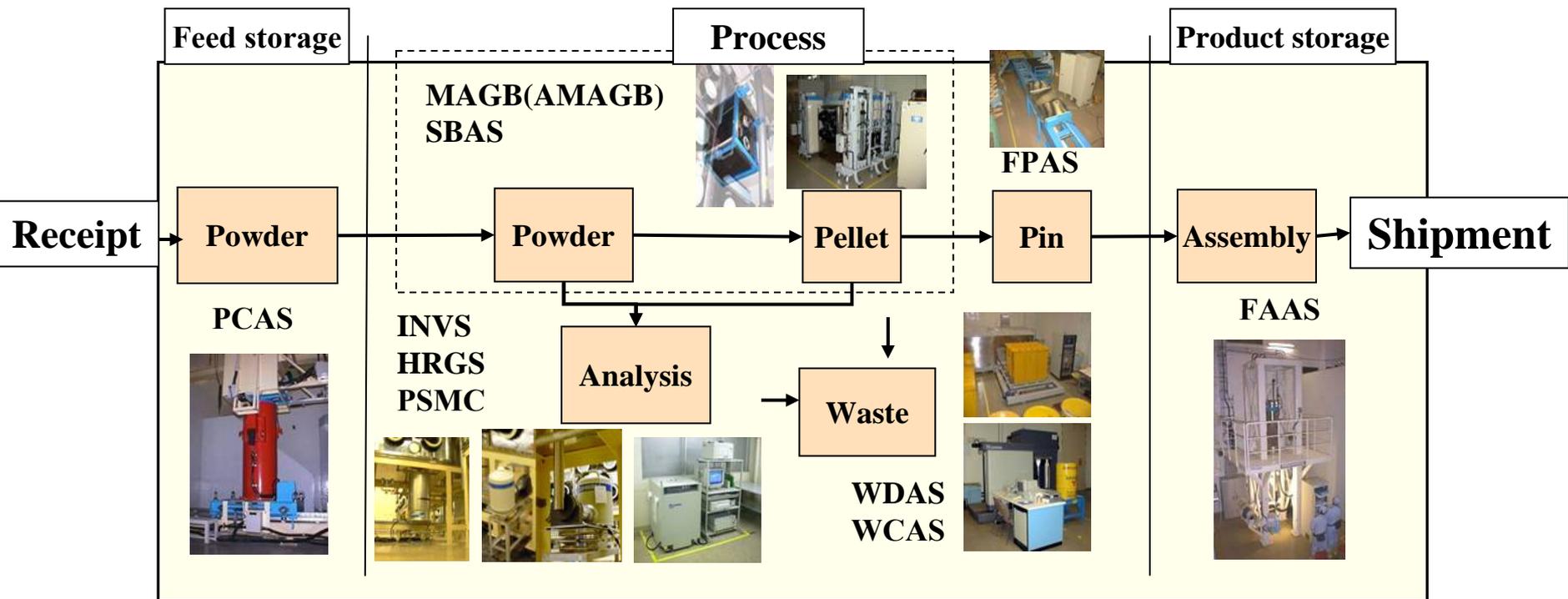
Safeguards Features:

- Measurement and monitoring of MOX feed, process, and product
- Glove-box holdup measurements
- NRTA and remote/unattended systems
- Containment/surveillance systems
- ~ 11 different SG systems at PFPF





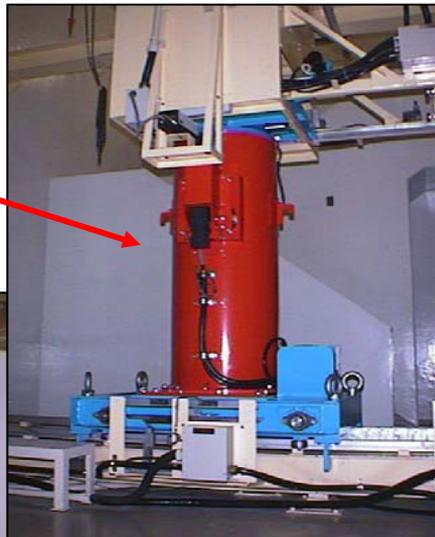
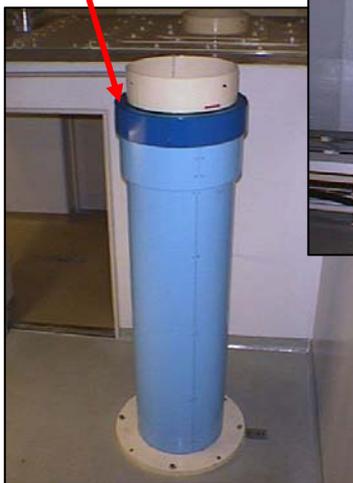
NDA Systems Installed Throughout the Process





Input MOX verification (PCAS), Process Holdup (GBAS)

Measure PuO₂ & MOX into facility



Process Line Holdup Detectors



- Features
- * Installed as part of facility robotics transfer system
 - * Continuous and unattended mode operation





Fuel Pin Measurement (FPAS), Fuel Assembly (FAAS)

Measure MOX fuel pins during fabrication



Measure MOX product assemblies leaving facility



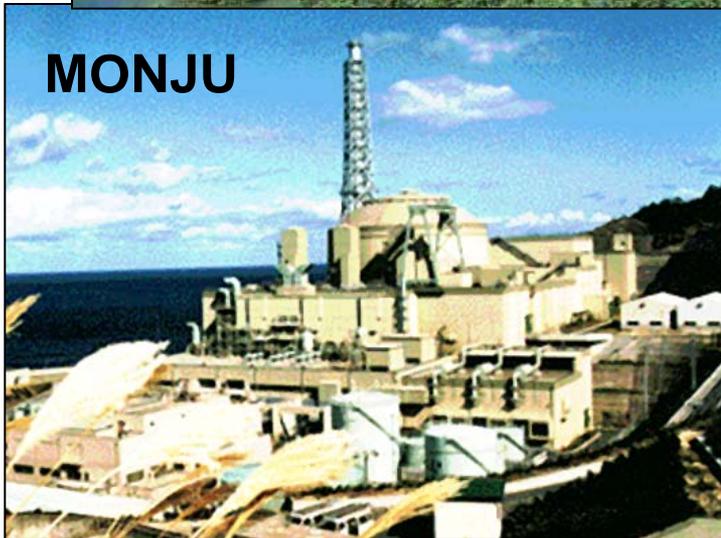
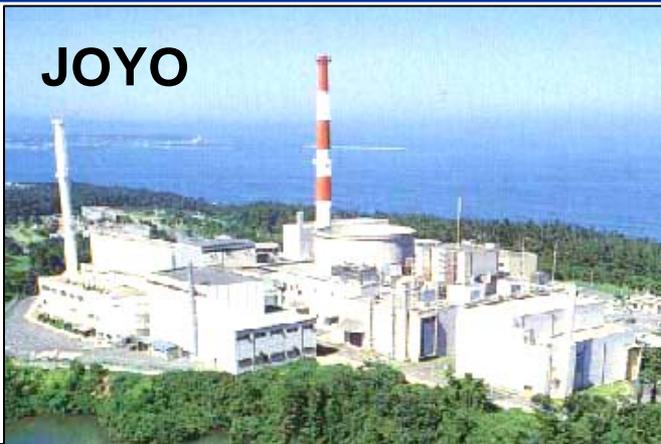


Fast Reactor





Fast breeder reactors in Japan under full-scope IAEA safeguards



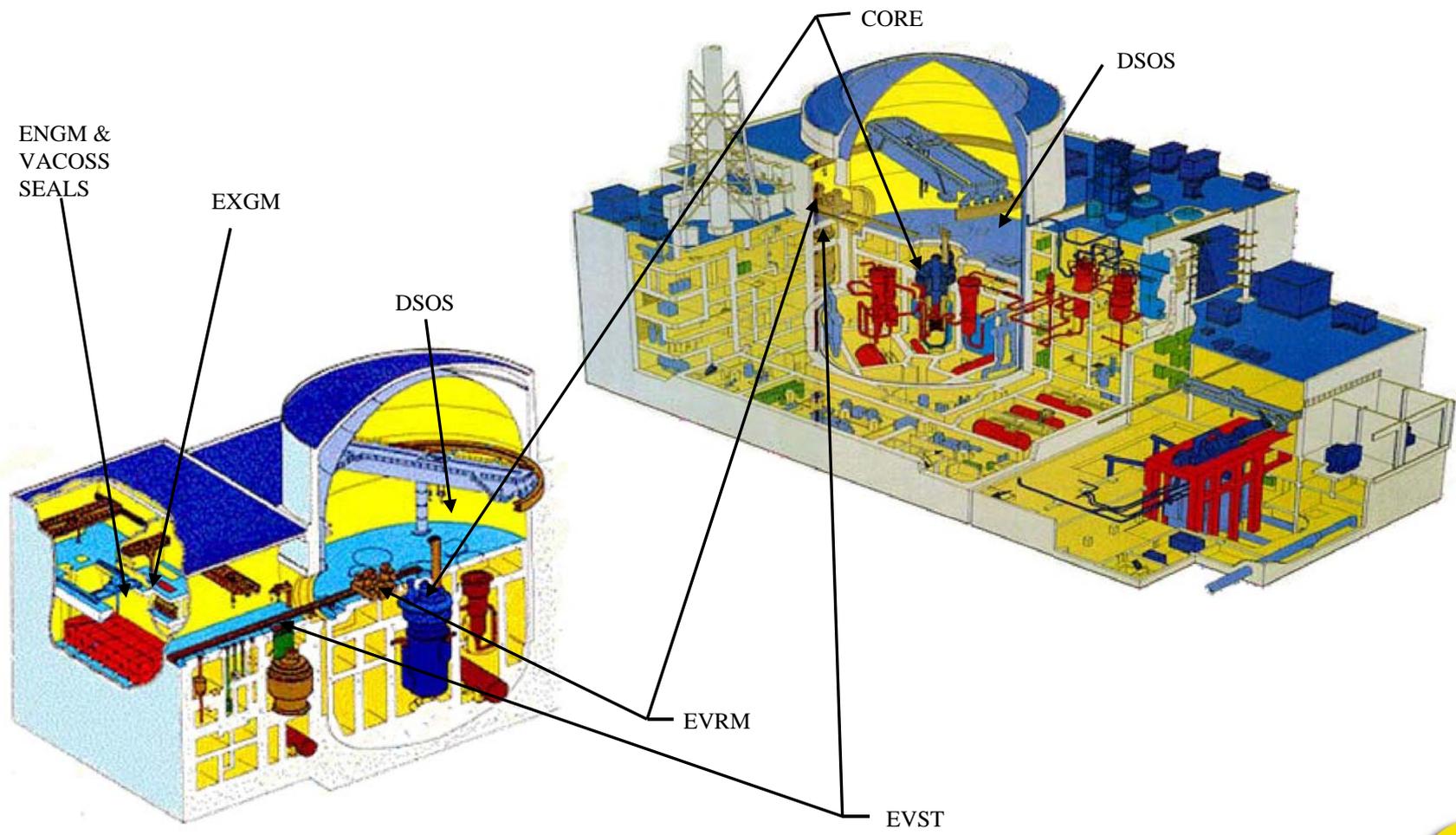
Safeguards Features:

- Fresh fuel assembly input gate monitors
- Reactor head seals and surveillance
- Reactor reload monitors and surveillance
- Fresh and spent fuel storage monitors
- Spent fuel discharge monitors





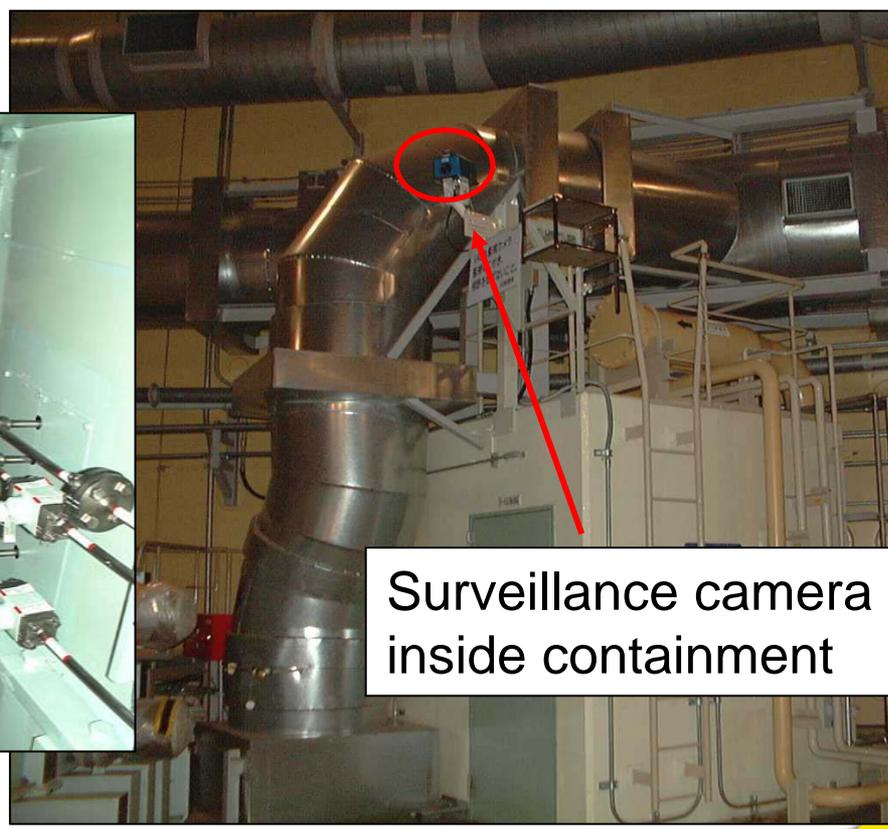
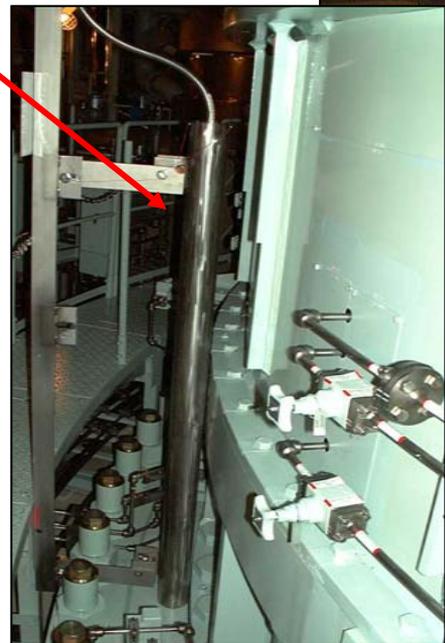
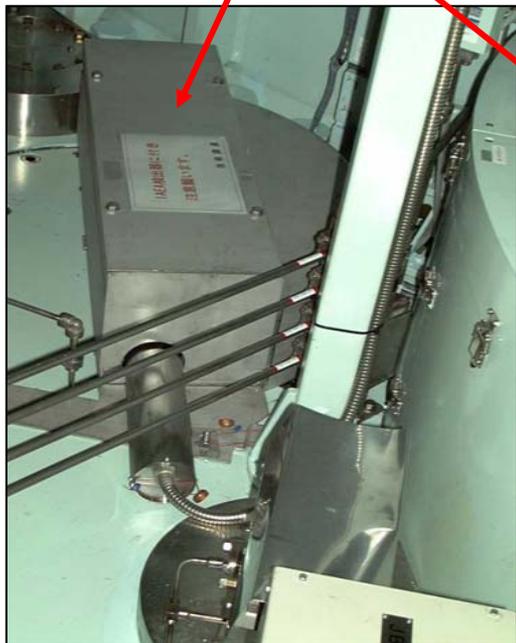
Safeguards at Monju





Core Monitoring

Two detectors monitoring top of CORE



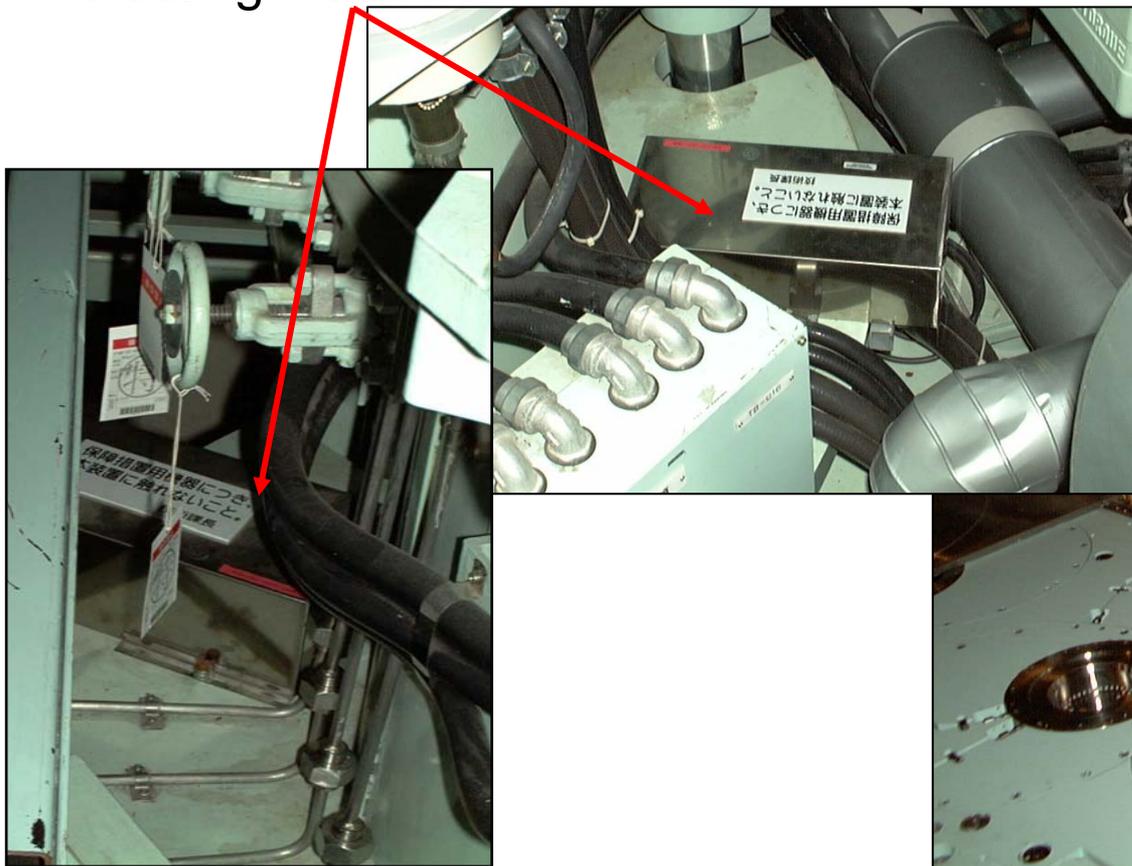
Surveillance camera inside containment





Refueling and Transfer Monitoring

Two detectors monitoring refueling machine



One detector monitors transfers between storage and refueling machine





Entrance Gate Monitor, Seals

Entrance gate monitor

Seals on fresh fuel storage

Seal on fresh fuel storage room access portal

