50-Year Application of Nondestructive Assay in Science and Technology of Nuclear Material Security at Los Alamos National Laboratory

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Outline

- Introduction
- Gamma-Ray Emission
  - Gamma-ray spectrometry-based nondestructive assay (NDA) measurements:
  - Low resolution (NaI, LaBr) $\frac{\Delta E}{E} \approx 3 - 6\%$
    - Example: Uranium Assay (Enrichment Meter)
  - High resolution (HPGe) $\frac{\Delta E}{E} \approx 0.13 - 0.4\%$
    - Example: Plutonium Assay (FRAM)
- Heat emission - Calorimetry
- Neutron Emission
  - Shufflers
  - Well Neutrons Coincidence Counter (passive and active)
- Conclusion

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Introduction

- Los Alamos National Laboratory's (LANL's) work on measurement techniques for nuclear safeguards was started by Bob Keepin on his return from the IAEA in 1966.
- During 2016–17, LANL is celebrating its 50th anniversary of safeguards work
- This talk presents partial coverage of the range of methods that have been developed and used for measurement and verification of nuclear material worldwide.

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Gamma-Ray Spectrometry-Based NDA Methodologies – "Enrichment Meter"

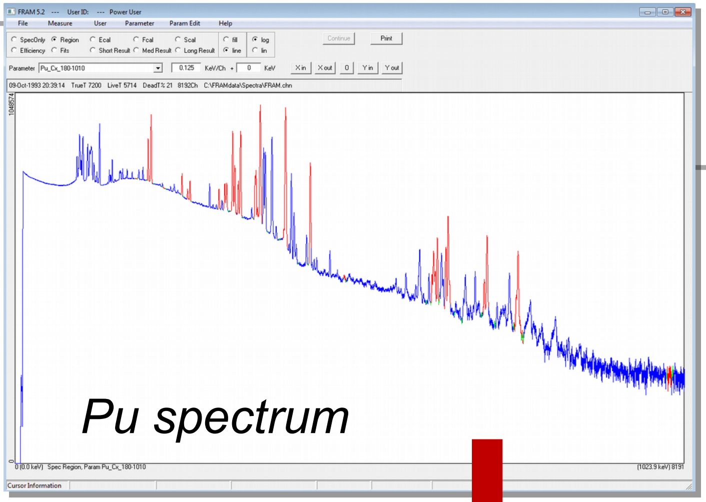
- Gamma-ray isotopic analysis measurements of material samples are probably the best examples of the practical application of gamma-ray spectrometry.
- Based on the concept of "infinite thickness" of a uranium sample that leads to the *concept* of "visible volume," which is determined by the measurement geometry; collimator, detector, and the mean free path of the 186 keV gamma ray in the sample.
- First applied to the measurement of UF_6 in transportation cylinders (1969)

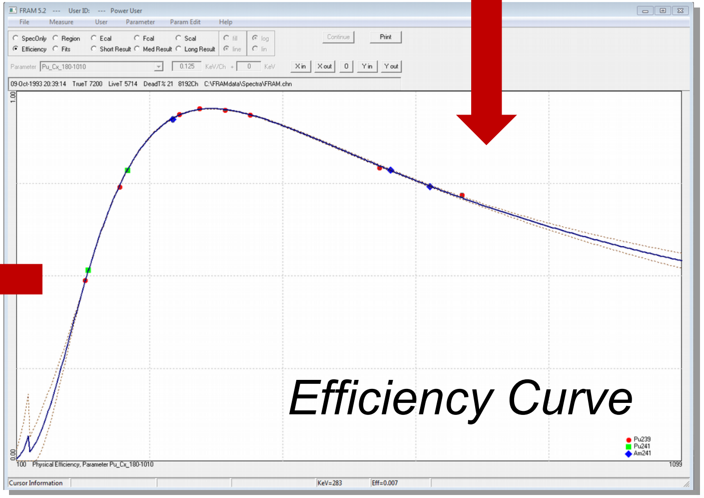
Fig. 10. Field measurement of ^{235}U enrichment of UF_6 in a 2-1/2-ton shipping container. A portable NaI detector is used to measure 186-keV ^{235}U gamma rays. A portable ultrasonic gauge is used to determine the cylinder wall thickness for the attenuation correction.

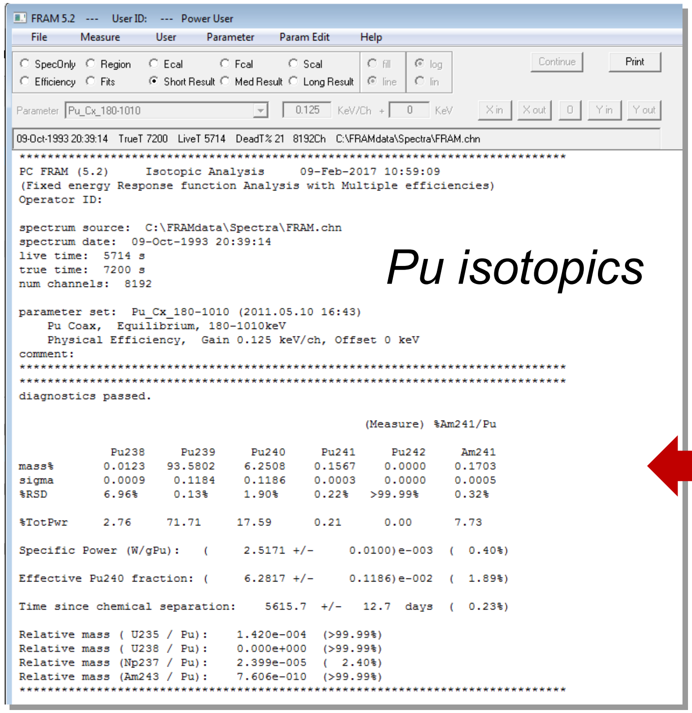
LOS ALAMOS SCIENCE/Summer 1980 97

High-Resolution Gamma Analysis

- **FRAM** uses the spectrum itself to determine relative efficiency curve and calculates Pu and U isotopics





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*******************************************************************************
PC FRAM (5.2)     Isotopic Analysis     09-Feb-2017 10:59:09
(Fixed energy Response function Analysis with Multiple efficiencies)
Operator ID:

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

parameter set:  Pu_Cx_180-1010 (2011.05.10 16:43)
    Pu Coax,  Equilibrium, 180-1010keV
    Physical Efficiency,  Gain 0.125 keV/ch, Offset 0 keV
comment:
*******************************************************************************
*******************************************************************************
diagnostics passed.

                                            (Measure) %Am241/Pu

             Pu238     Pu239     Pu240     Pu241     Pu242     Am241
mass%       0.0123   93.5802    6.2508    0.1567    0.0000    0.1703
sigma       0.0009    0.1184    0.1186    0.0003    0.0000    0.0005
%RSD        6.96%     0.13%     1.90%     0.22%   >99.99%     0.32%

%TotPwr     2.76     71.71     17.59      0.21      0.00      7.73

Specific Power (W/gPu):   (     2.5171 +/-     0.0100)e-003  (  0.40%)

Effective Pu240 fraction: (     6.2817 +/-     0.1186)e-002  (  1.89%)

Time since chemical separation:    5615.7  +/-   12.7  days  (  0.23%)

Relative mass ( U235 / Pu):    1.420e-004  (>99.99%)
Relative mass ( U238 / Pu):    0.000e+000  (>99.99%)
Relative mass (Np237 / Pu):    2.399e-005  (  2.40%)
Relative mass (Am243 / Pu):    7.606e-010  (>99.99%)
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Calorimetric Assay

- Calorimetry is a technique used to measure heat.
- Used for nuclear safeguards, MC&A purposes, shipper-receiver confirmation, process and quality control, and waste characterization
- When applied to MC&A, calorimetric assay determines the rate of heat generation (power) from radioactive nuclei, including nuclear material calorimetry.
- First used in 1903 to measure heat produced by radium

The world's first ice calorimeter was used during the winter of 1782–83 by Antoine Lavoisier and Pierre-Simon Laplace to determine the heat involved in various chemical changes.

Calorimeter R&D Program

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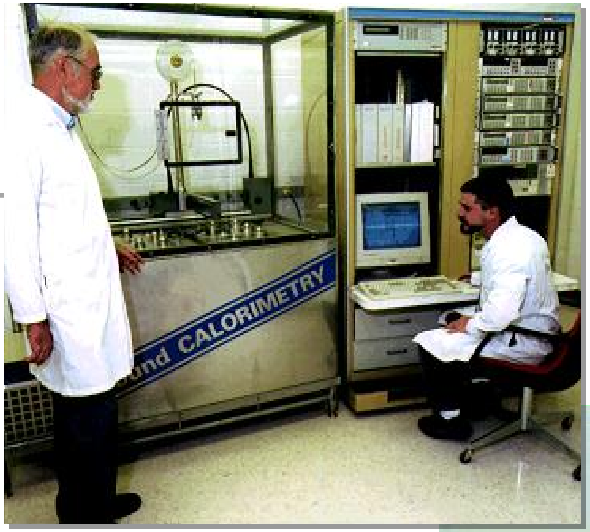
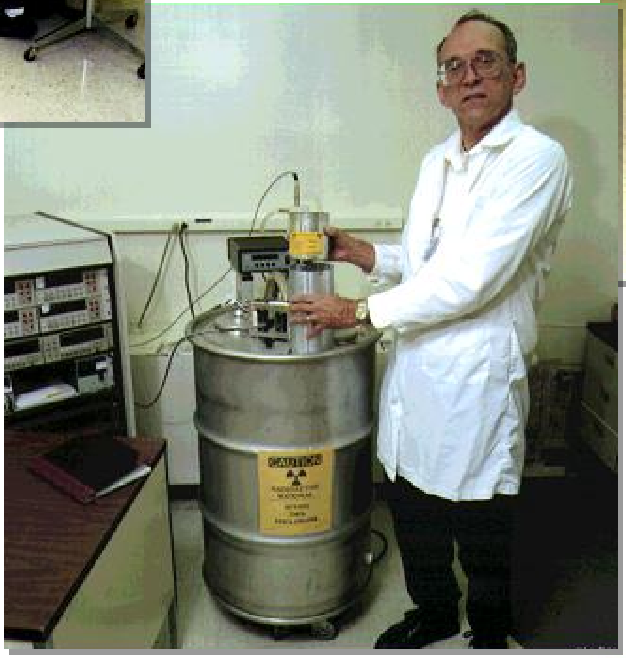
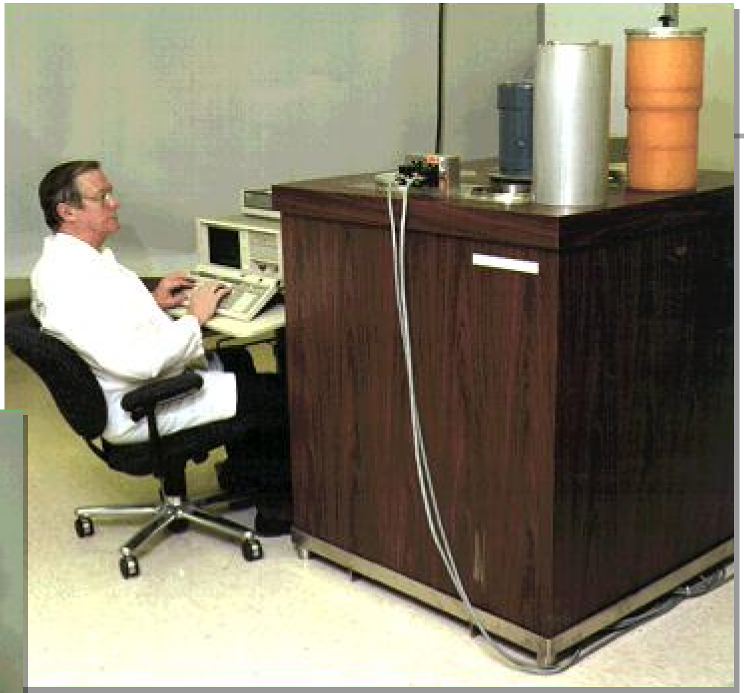


Heat Standards Calorimeter





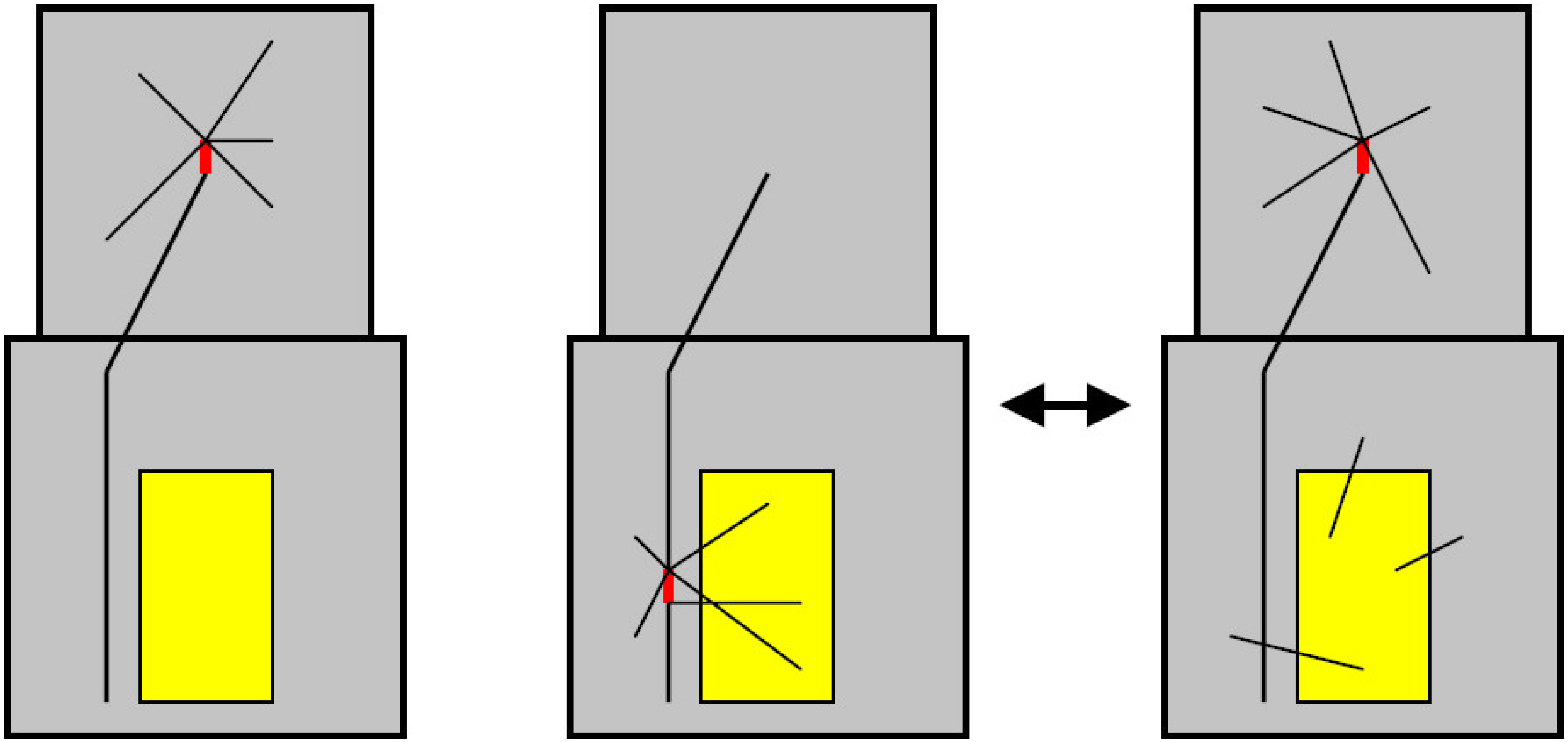
Water Bath Calorimeter

Shuffler

- The ^{252}Cf source alternates between the storage block and the assay chamber that contains the object to be assayed.
- The source is stored for the background count and again after the irradiation to count delayed neutrons.
- The irradiation and counting steps are repeated many times until the desired precision is reached. This “shuffling” of the source between two positions gives the instrument its name.
- Detector tubes surround the object to record the delayed neutrons. (Ref. 6)

This billet shuffler had a precision of 0.25% (1σ) and an accuracy of 0.5% (1σ) even though the assay time (including background) was only 10 minutes. Such performance was possible because of the large ^{235}U loading (1.7 kg) and the billet’s hollow core.

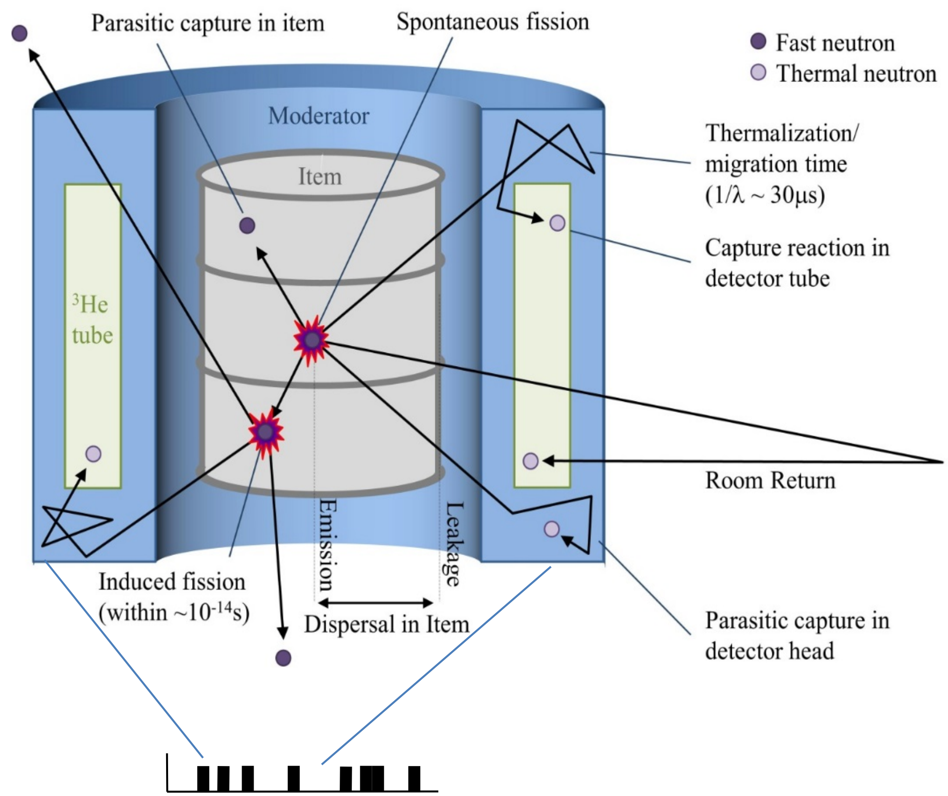
Operating Principle



(a) Background (b) Irradiation (c) Delayed Neutron Counting

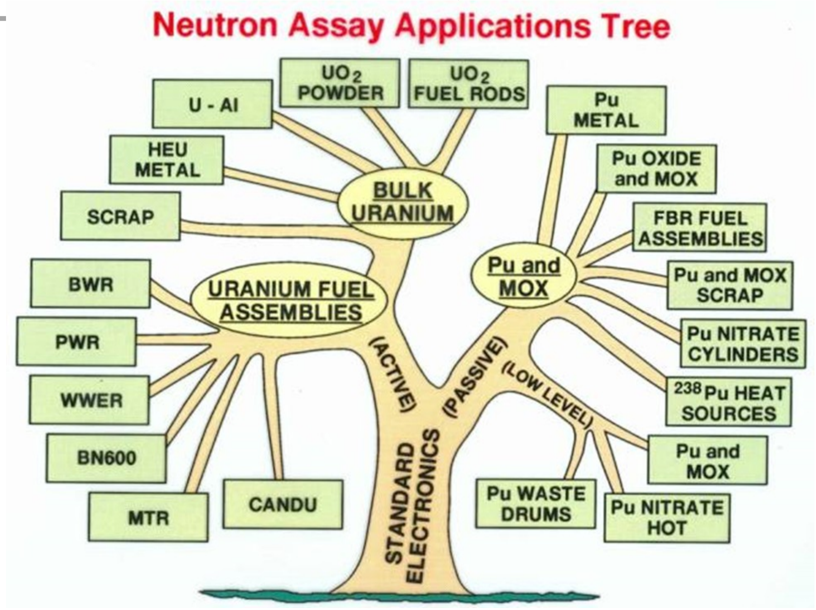
Passive Neutron Counting

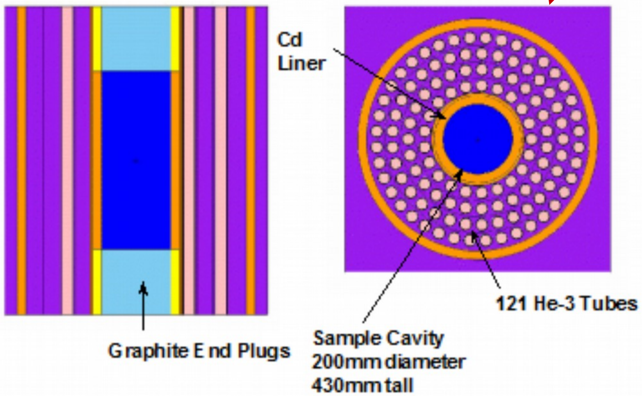
- The Pu item is surrounded by ^{3}He detectors embedded in polyethylene.
- The bursts of neutrons produced by fission can be distinguished from the single neutrons produced by (α,n) reactions by measuring the degree of correlation in the pulse train.
- Total neutron counting measures all neutrons.
- Passive neutron coincidence counting measures Pu mass and multiplication (M).
- Passive multiplicity counting determines Pu, M, and (α,n)—good for impure items.



Neutron Pulse Stream

Neutron Detector Family





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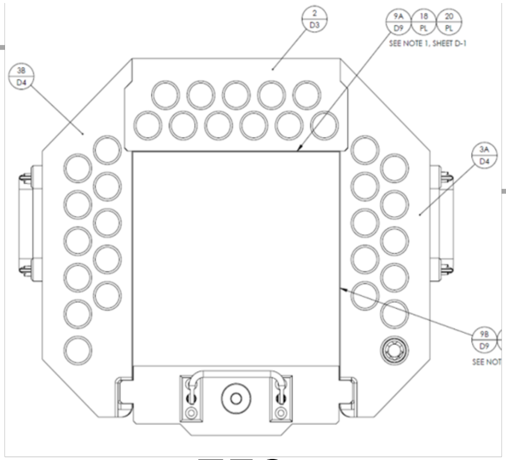
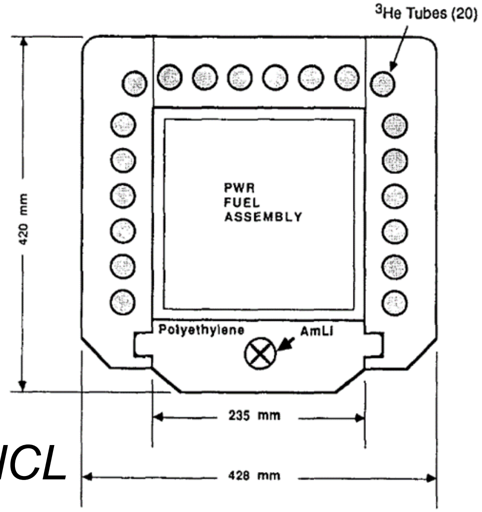
Active Coincidence Counters

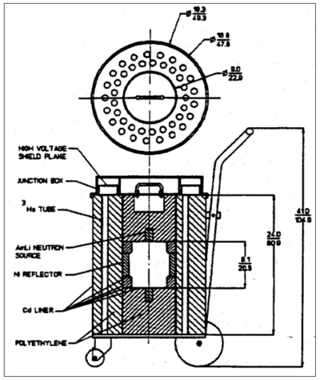
- Active coincidence counting: using an Am/Li neutron source to induce fission in the fissile material—used to measure ^{235}U mass
- **Uranium Neutron Coincidence Collar (UNCL)** (1982) for fresh PWR and BWR fuel assemblies
- **Active Well Coincidence Counter (AWCC)** (1985) for U metal and oxide
- **Euratom Fast Collar (EFC)** (2015) for PWR fuel assemblies with burnable poisons



EFC



UNCL



AWCC

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Waste Measurement

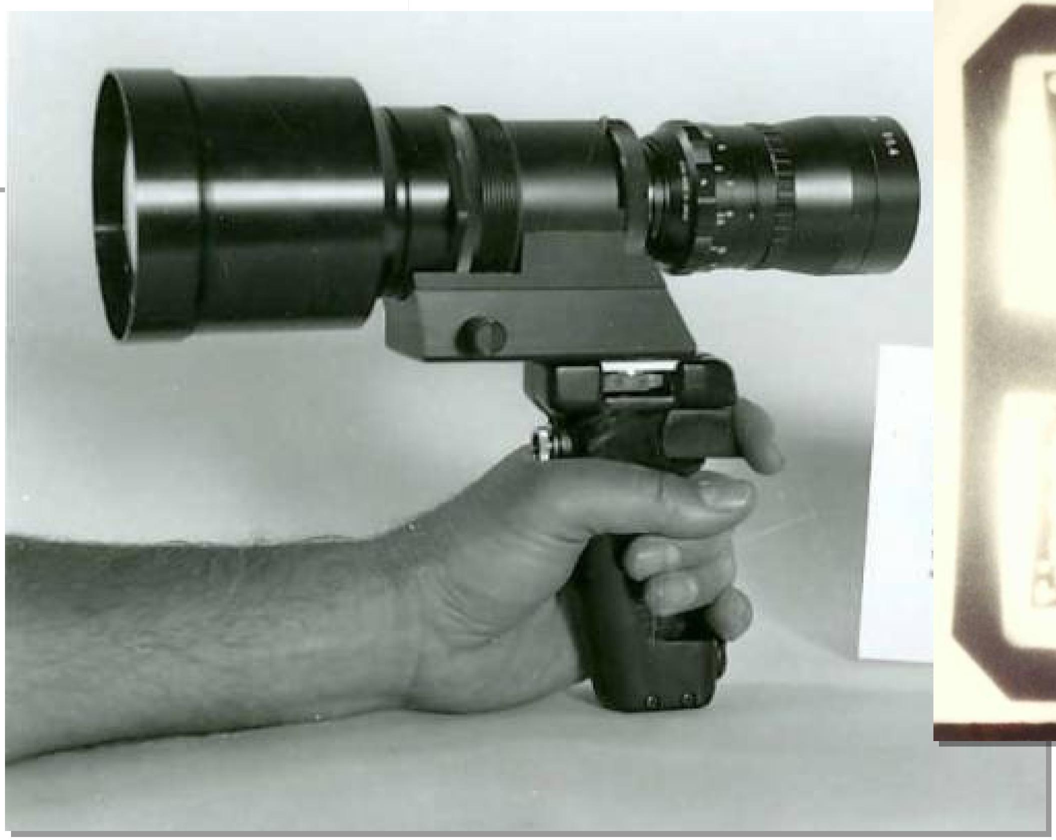
Super high-efficiency neutron counter (year 2000)

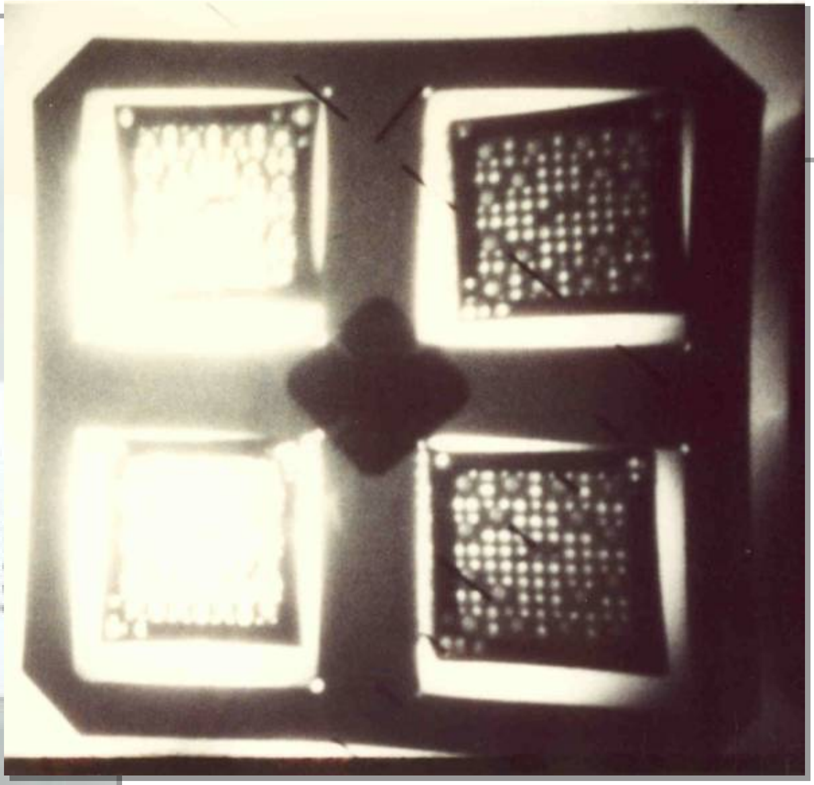
Largest Waste Counter for Rokkasho Reprocessing Plant

WCAS-B

Concrete Block Load Testing
Albuquerque, New Mexico
October 2001

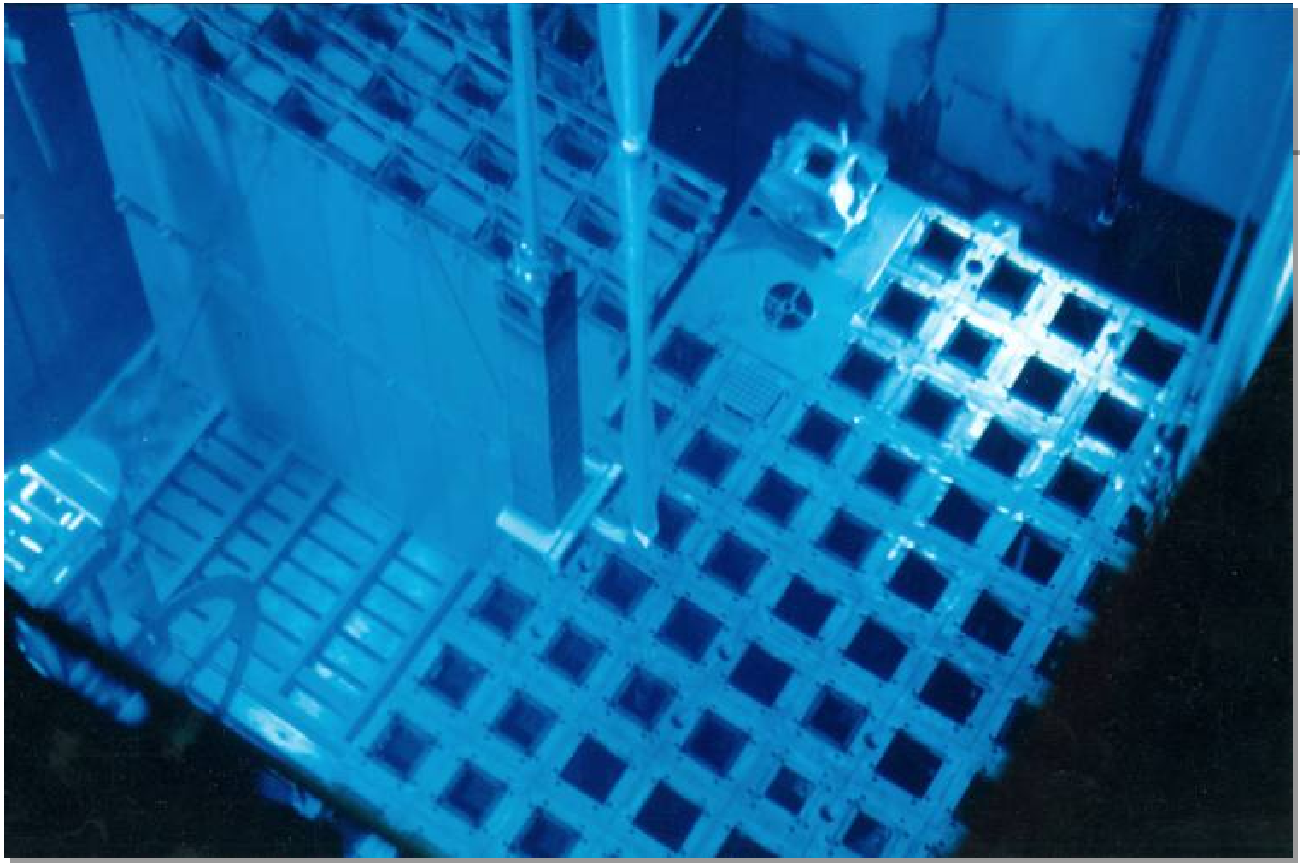
Spent Fuel Measurements





Cerenkov Viewing Device (CVD) for
Spent Fuel Verification (circa 1979)

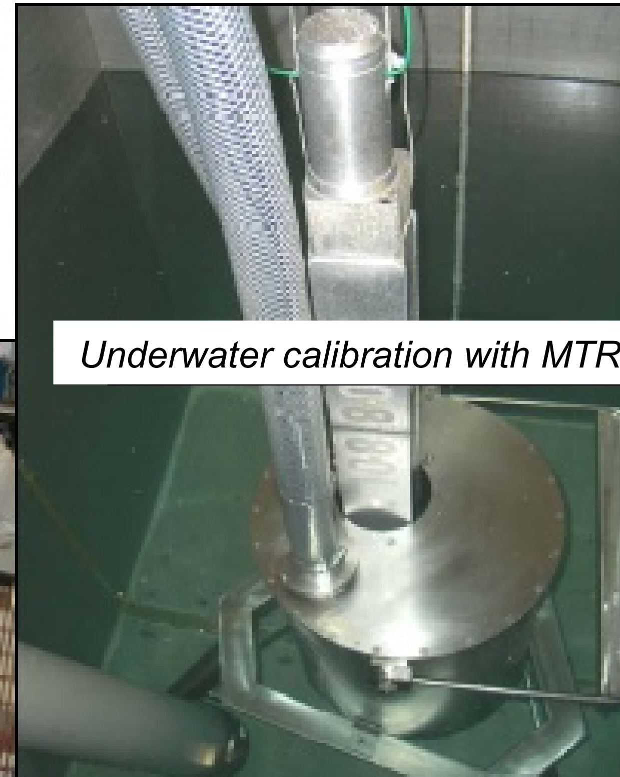
Fork Detector for Spent Fuel Verification – 1978



Gross neutron and gamma detectors to determine burnup and cooling time

Los Alamos Advanced Experimental Reactor Fuel Counter (AEFC) – 2007

In-pool testing


Underwater calibration with MTR fuel

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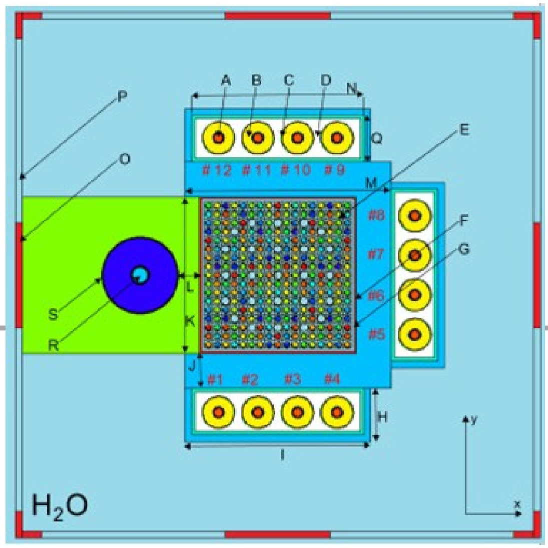
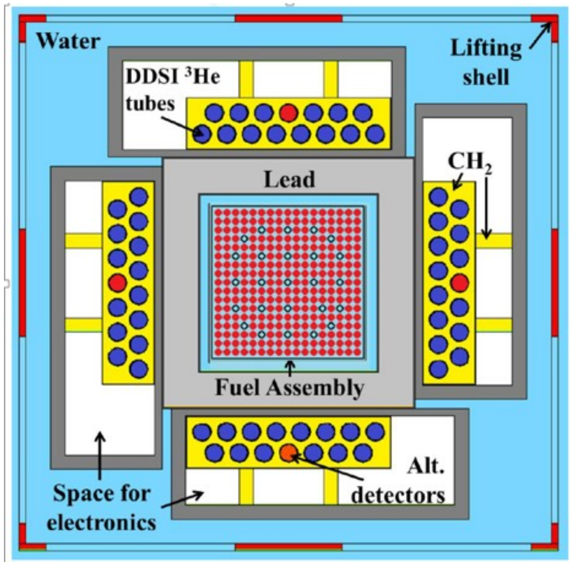
Current NDA Systems for Spent Fuel – 2017

- Aiming to measure initial enrichment, burnup, and cooling time and to detect diversion
- Differential Dieaway – A 14 MeV neutron generator is used to induce fission in the fuel
- Differential Dieaway self-interrogation: passive measurement recording coincidences from neutrons that thermalize and re-enter the fuel





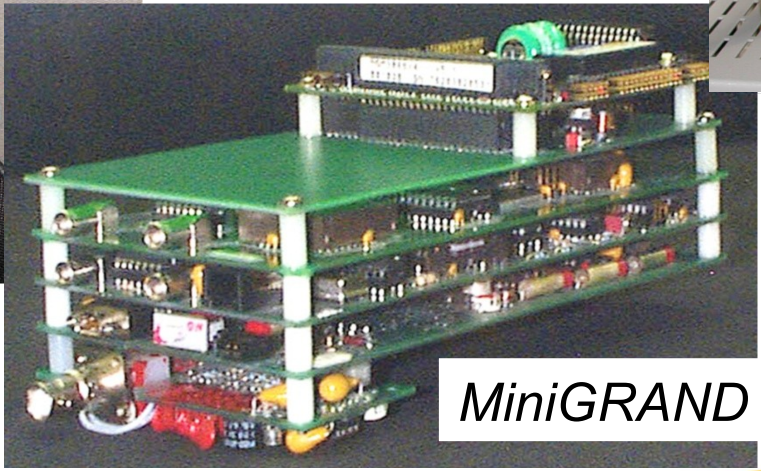
Electronic Development

- multichannel analyzer
- neutron multiplicity
- gross neutron
- gross gamma (current)


PMCA

AMSR

JSR-15


MiniGRAND Family

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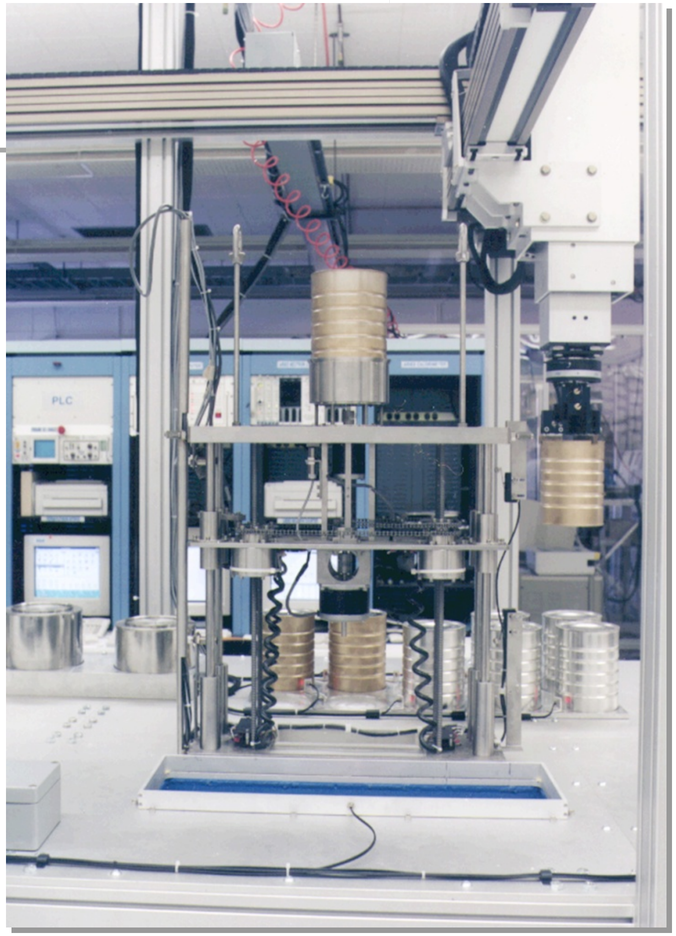
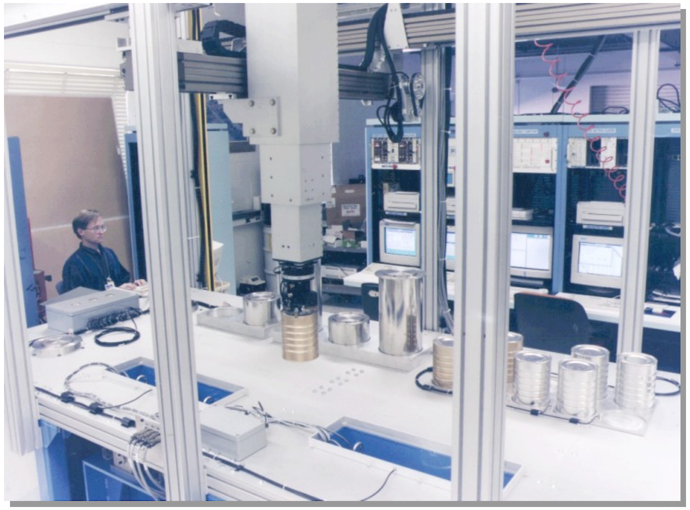
Advanced Recovery and Integrated Extraction System (ARIES)



Neutron and gamma NDA are used extensively for nuclear material accountancy during pit dismantlement.



1987–2014 Russian Program

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1988–2007 BN-350, Aqtau, Kazakhstan

- Neutron measurement of discharged fuel assemblies underwater
- Neutron and gamma monitoring of spent fuel stored casks

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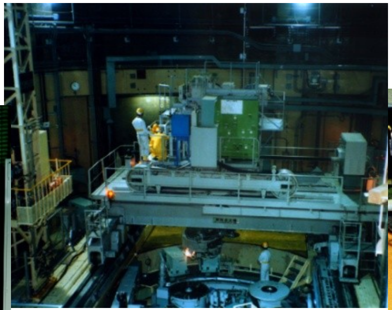
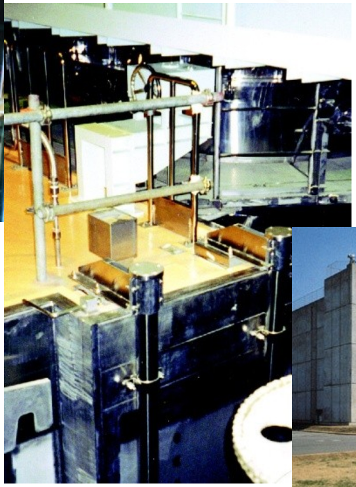
Evolution of NDA and Remote Monitoring

PFPF

JOYO



Rokkasho



PFPF

1973	1987	1992	1997	2000
Portable NDA	Unattended NDA	Integrated NDA & Rad. Monitoring	Integrated Video & Rad. Monitoring	Remote Transmission
	Quantity	**Quantity Movements**	**Operation Quantity**	**Remote Operation**

Safeguards Technology Training

50th Nondestructive Assay Inspector Training Course
Los Alamos, New Mexico
August 12 – 22, 2008

- Offers a comprehensive suite of courses on safeguards measurement techniques and general MC&A principles, with focus on hands-on activities
- Since 1973, more than 324 safeguards training courses have been held, with more than 5600 students attending.
- Courses cover all three types of NDA measurements (neutron, gamma, calorimetry) using a unique and broad set of nuclear material standards.
- Courses are taught to both international customers (IAEA, China, Indonesia, Kazhkstan, etc.) and domestic customers (DOE/NNSA laboratories, NRC, private companies)

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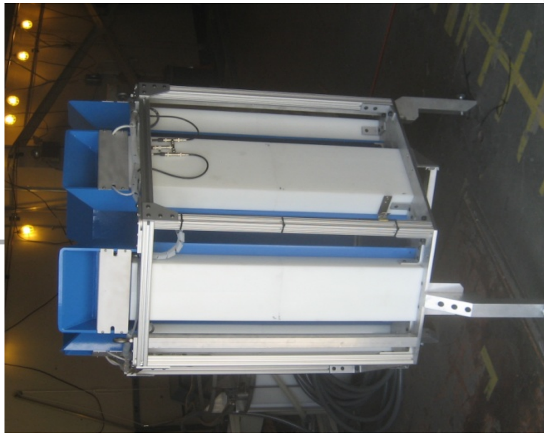
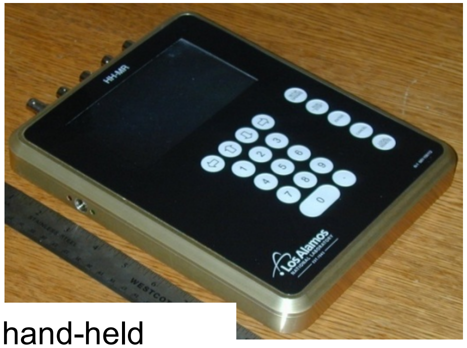
Current Thrust Areas for Safeguards Instrument Development

- Next-generation treaty verification instruments
- Spent fuel assay
- Safeguards for large-throughput plants
- High-accuracy nondestructive assay to replace destructive analysis in large-scale facilities
- Mass balance measurement capability for uranium enrichment plants
- Next-generation data acquisition systems
- Intractable safegua



Chernobyl Drum Assay System (CDAS) at TA-35 before deployment at Chernobyl to identify and quantify special nuclear material discovered during excavation for New Safe Confinement (NSC) building.



Portable hand-held multiplicity register (JSR-15) commercialized in 2008.

The Attribute Verification System–Neutron and Gamma (AVNG) was Russian built in close collaboration with LANL and installed at the VNIIEF weapons facility in Sarov, Russia. The detectors (foreground) are housed in the shielded room (background) during operation to prevent disclosure of classified information.

Summary

- Threat of terrorist individuals or organizations gaining unauthorized access to the special nuclear material (SNM) is real.
- The vision of a terrorist attack involving nuclear devices or nuclear material is abhorrent, but it must be seriously considered as a potential risk.
- An international effort to interdict unauthorized possession, transportation, sale, or use of SNM is needed.
- Such an effort will challenge the current capabilities of any nation for detection identification and control of illicit SNM and is perhaps beyond the current capabilities of all nations combined, making cooperation in these vital technologies all the more urgent.

Conclusions

- Most of the NDA methodologies used today for accountancy purposes were developed during the 1960s and 1970s.
- A fresh look is needed to address today's measurement requirements for the new generation of safeguard policies.

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  - Lynn Foster
  - Michael Kaufman

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THANK YOU!
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