DEVELOPMENT AND CALIBRATION OF THE SHIELDED MEASUREMENT SYSTEM FOR FISSILE CONTENTS MEASUREMENTS ON IRRADIATED NUCLEAR FUEL IN DRY STORAGE

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ABSTRACT

In recent years there has been a trend towards storage of Irradiated Nuclear Fuel (INF) in dry conditions rather than in underwater environments. At the same time, the Department of Energy (DOE) has begun encouraging custodians of INF to perform measurements on INF for which no recent fissile contents measurement data exists. INF, in the form of spent fuel from Experimental Breeder Reactor 2 (EBR-II), has been stored in close-fitting, dry underground storage locations at the Radioactive Scrap and Waste Facility (RSWF) at Argonne National Laboratory-West (ANL-W) for many years. In Fiscal Year 2000, funding was obtained from the DOE Office of Safeguards and Security Technology Development Program to develop and prepare for deployment a Shielded Measurement System (SMS) to perform fissile content measurements on INF stored in the RSWF. The SMS is equipped to lift an INF item out of its storage location, perform scanning neutron coincidence and high-resolution gamma-ray measurements, and restore the item to its storage location. The neutron and gamma-ray measurement results are compared to predictions based on isotope depletion and Monte Carlo neutral-particle transport models to provide confirmation of the accuracy of the models and hence of the fissile material contents of the item as calculated by the same models. This paper describes the SMS and discusses the results of the first calibration and validation measurements performed with the SMS.

INTRODUCTION

The SMS was developed to perform fissile content measurements on INF items stored in individual dry underground storage liners at the ANL-W RSWF. The SMS provides a capability to perform fissile content verification in INF items for which measurement data is not available. The DOE has given guidance to INF custodians that measurement data is needed to provide fissile material assurance for INF items (Reference 1).

The SMS consists of a support base designed for use on uneven soil surfaces, an instrumented shield ring containing and supporting neutron coincidence and gamma-ray spectroscopy instrumentation, a scan shield to provide shielding for the INF item as it is scanned upwards through the instrumented shield ring, and a sample positioning system which lifts the INF item out of its storage location, supports and positions the INF item during measurement, and returns the item to storage after completion of measurements (Figure 1).

Measurement data taken at a number of stations along the length of an INF item include a neutron coincidence count rate and high-resolution gamma-ray spectra. The neutron coincidence count rate is indicative of spontaneous-fission isotope contents, which in the case of the Experimental Breeder Reactor II (EBR-II) fuel at ANL-W is almost entirely Pu-240. The gamma-ray spectra contain gamma-ray lines of Cs-137 among other isotopes. The Cs-137 distribution along the axis of an INF item is indicative of the burnup profile and hence the distribution of Pu-239 and Pu-240 in the item, provided no cesium migration occurs during irradiation. Other gamma-rays which have been observed are Co-60 and Eu-154. Taken together, the neutron and gamma-ray profiles provide a fingerprint for a given INF assembly, allowing periodic reconfirmation that the assembly has not been tampered with.

SMS measurements are used along with calculationally-based determinations of fissile contents in an INF item. The measurement data are compared with detector response characteristics calculated using isotope depletion and Monte Carlo computer software. A comparison of measured detector response with the calculated detector response allows a check on the validity of the model results and hence a check on the calculated fissile isotope contents of the INF item.

The SMS measurement systems were calibrated using point sources of Cf-252, Cs-137, and Co-60. Basic detector characteristics such as efficiency, sensitivity profile, and neutron dieaway time were determined. These characteristics will be used to verify and calibrate the performance of the Monte Carlo detector response models.

Validation measurements of representative, well-characterized INF items (EBR-II blanket assemblies) have been performed. The results of these measurements will be compared to detector response predictions calculated with the isotope-depletion models and the Monte Carlo detector response models to confirm the validity of the modeling process. Other fuel types, starting with EBR-II driver assemblies, will be measured in the future.

SHIELDED MEASUREMENT SYSTEM DESCRIPTION

The SMS is designed to perform measurements on INF items contained in waste cans in common use at ANL-W called HFEF-5 cans. These containers are approximately 6 feet long and 13 inches in diameter and contain from one to six spent fuel assemblies from EBR-II. Spent EBR-II fuel may be divided into two main categories, driver and radial blanket assemblies. Both are metallic alloy fuels consisting predominantly of uranium with a few percent zirconium as well as plutonium and higher actinides resulting from neutron exposure in the EBR-II reactor. Both types are clad in stainless steel. The fuelled portion of driver assemblies are approximately 13 inches long. Radial blanket assembly fueled portions are either 33 or 55 inches long depending on type. Neither type of fuel contains significant quantities of curium from a neutron-coincidence measurement standpoint since the drivers are highly enriched and the blankets, while consisting of depleted uranium, achieve only a low burnup. Thus, the bulk of the neutron coincidence signal may be assumed to be produced by Pu-240 spontaneous fission.

Figure 1 displays the main features of the SMS. The SMS is composed of several sections, most of which are not rigidly attached to each other but are merely stacked one atop the other for operational flexibility and easy handling. The total weight of the SMS is approximately twenty tons. The heaviest component, the scan shield/transition piece assembly, weighs approximately 9 tons.

The base of the SMS is a ¾-inch thick circular plate which increases the footprint of the SMS to decrease the bearing pressure on the surface supporting the SMS. Stacked atop this is the funnel/shield-ring assembly. This is split diagonally so that it can be adjusted to make the remainder of the SMS level when used on uneven soil. It also provides a guide funnel to guide the measured item during lifts.

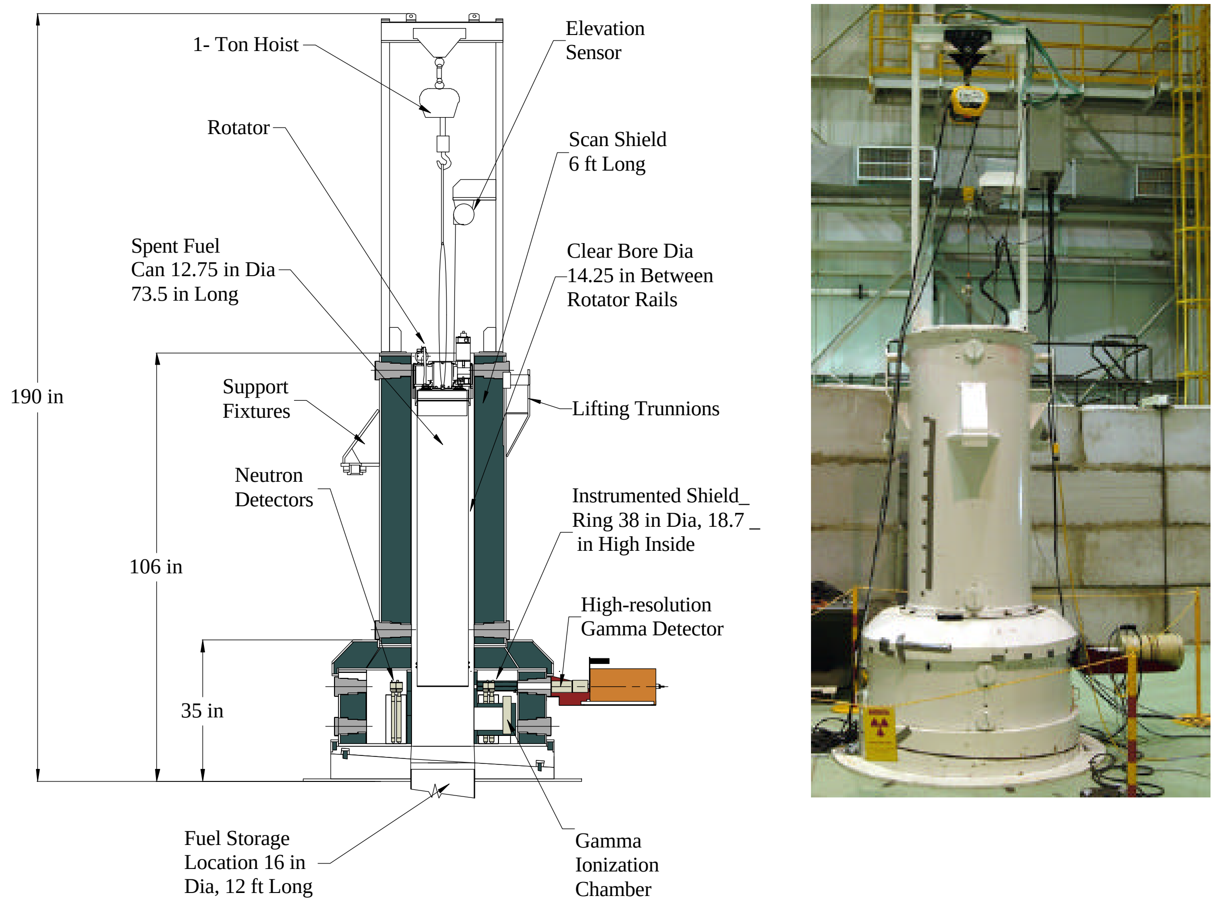

Figure 1, Longitudinal Section and Photograph of Shielded Measurement System

The instrumented shield ring sits atop the funnel/shield ring assembly. The instrumented shield ring contains a neutron coincidence measurement system, a gamma ionization chamber, and a mount and variable-height collimator for an externally-mounted high resolution gamma detector. The instrumented shield ring is shown in Figure 2.

The next component is the scan shield/transition piece assembly. This provides a shielded space into which the measured item moves as it is being scanned upwards. The motion control system is mounted on the scan shield/transition piece assembly and consists of a rotator, which rides on top of the measured item, and a hoist, which lifts the item. The rotator allows azimuthal positioning of the item and can also be used to continuously rotate the item to provide azimuthal averaging of gamma-ray data. A linear transducer is provided to allow accurate determination of the axial position of the item during measurement. The motion control system and data acquisition systems are computer-controlled. The SMS can be controlled remotely, reducing the potential radiation exposure to the operators except during loading and unloading of the measured item.

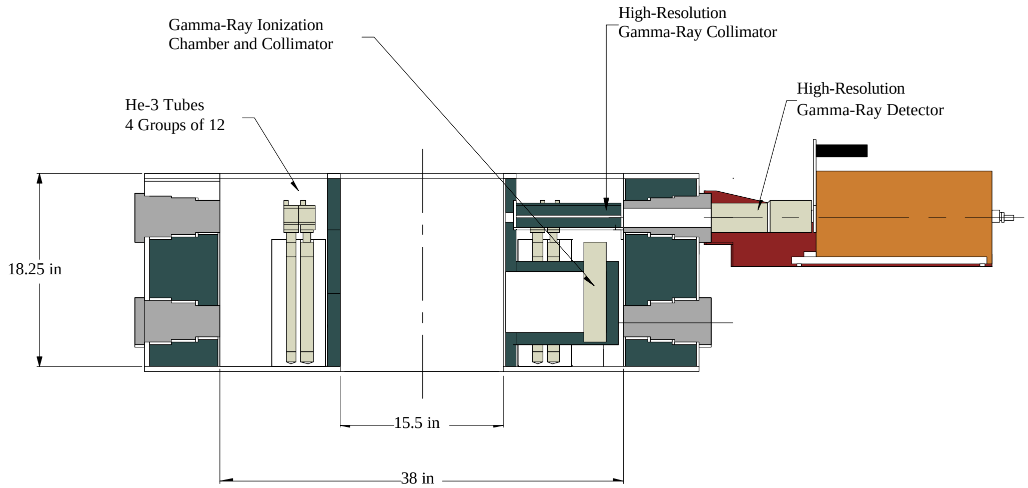

Figure 2, Measurement Systems Installed in the Instrumented Shield Ring

NEUTRON MEASUREMENT SYSTEM

The neutron measurement system of the SMS contains 48 ten-atmosphere He-3 tubes arranged in 4 sets of 12 at intervals of 90 degrees to form a circular array of two rows of tubes. Figure 2 presents a sectional view of the neutron detection system. Signal conditioning and amplifier/discriminator electronics are attached to the tops of the tubes. All tubes are interconnected to give a single signal output. The detector moderator is high-density polyethylene, which is wrapped in cadmium to preclude thermal neutron self-interrogation of the measured item. A standard (Canberra JSR-12) shift-register-based neutron coincidence counter is used with the neutron detection system.

A significant and highly-variable background neutron coincidence signal is caused by spallation neutrons induced by cosmic-ray interactions in the lead shielding of the SMS. Spallation events occasionally produce bursts of dozens of coincident neutrons, leading the shift register to detect hundreds of coincidences at once. Counts of small quantities of Pu-240 (approximately 1 gram) for times on the order of 5 minutes were found to possess a standard deviation of approximately 30 percent, compared with just a few percent that would be expected from counting statistics. Since the INF items to be measured often have Pu-240 loadings in this range, the spallation neutron signal must be subtracted. The rate of occurrence of spallation neutron bursts is low enough (approximately one every 30 seconds) that data collection times necessary for a simple background-subtraction process would be inordinately long. The solution adopted is to collect a large number of counts of very short duration (0.1 seconds) and to throw out counts with “high” values. To accomplish this, a histogram of the counts is constructed, a Gaussian function is fitted to the central, symmetrical part of the histogram, and only counts within +/- 3 standard deviations of the mean of the fitted function are summed.

Figure 3 illustrates this process. The abscissa of the plot in Figure 3 is the count value of the histogram bins, while the ordinate is the base-10 logarithm of the number of occurrences of the count value. The actual count represented by each bin of the histogram is the bin number times its number of occurrences. It will be noted that counts to the left of the origin subtract from the total. Negative net counts occur because of the short count times and the fact that the net count is the difference between two large numbers. In Figure 3, data from – 4 to + 11 on the x-axis would be used. Note that since the ordinate of Figure 3 is logarithmic, the difference between the measured histogram and the fitted Gaussian function is overemphasized. A test of this method with a source strong enough to overcome the spallation count interference shows that the short time bin counts add up to the same total count obtained by one long count within counting statistics. The neutron count profile in Figure 5 illustrates the effectiveness of this method in eliminating spallation neutron noise. The dashed line is the profile without spallation correction. It shows a great deal of variability compared to the spallation corrected profile (solid line).

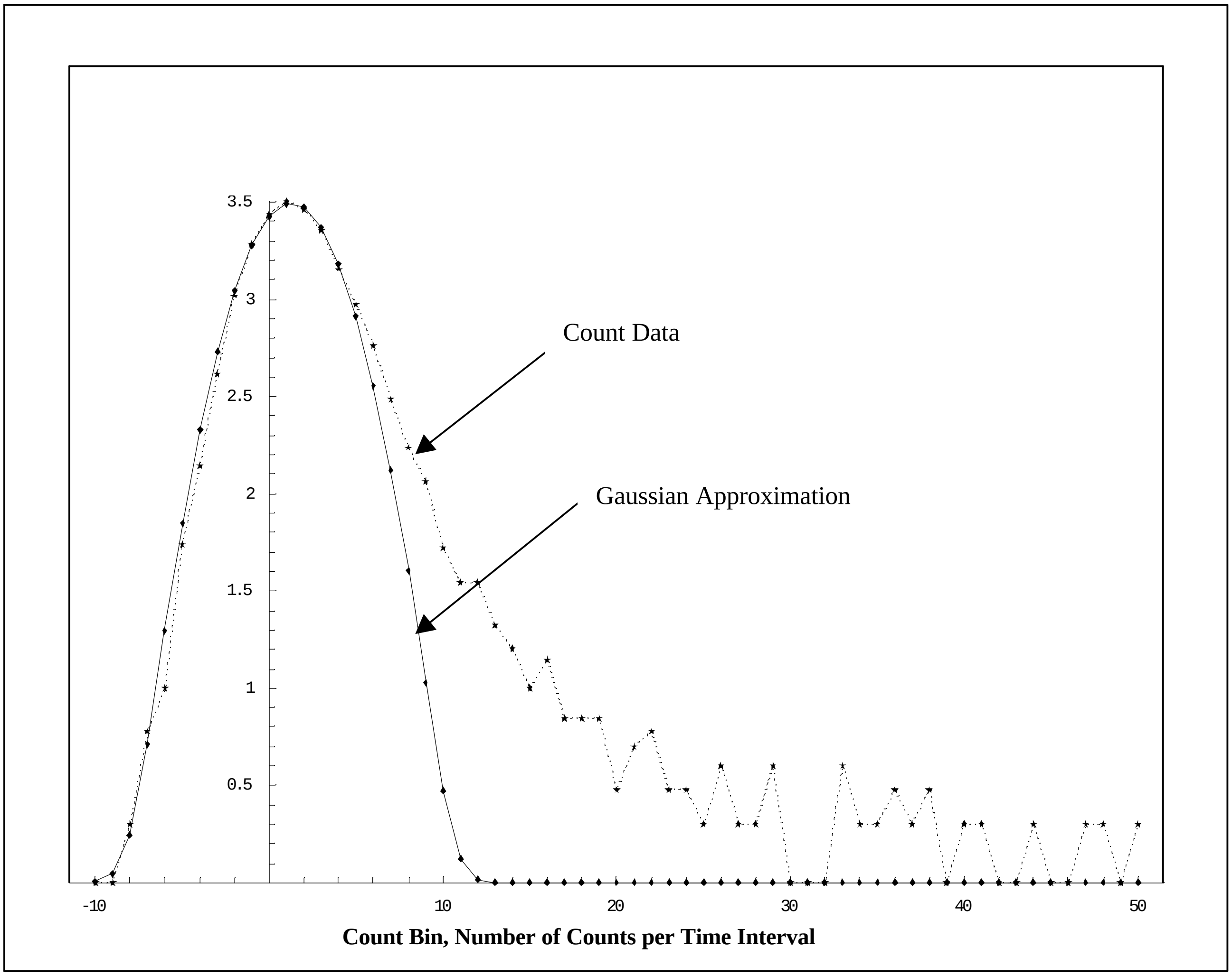

Figure 3, Histogram of Neutron Signal with Gaussian Fit to Non-Spallation Neutrons

GROSS GAMMA MEASUREMENT SYSTEM

A gross gamma-ray measurement system remains installed in the SMS as a holdover from an earlier phase of the instrument in which only gross neutron and gamma-ray emissions were to be measured. This instrument consists of an ionization chamber collimated to view approximately +/- 16 inches axially and 3 inches horizontally at the centerline of the SMS. Data from this instrument produced a dose rate profile very similar to the Co-60 gamma-ray profile shown in Figure 6. The maximum magnitude of the dose was approximately 15 R/hr, with a variation of approximately 50 percent from one side of the assembly to the other. The Co-60 gamma-ray intensity exhibited this same variation. This instrument is shown in Figure 2.

GAMMA-RAY SPECTROSCOPY MEASUREMENT SYSTEM

Gamma-ray spectra are acquired with a high-purity germanium detector mounted on the exterior of the instrumented shield ring. The detector views the measured item through a circular shield wall penetration and a horizontal, variable-slit collimator. The exterior mounting facilitates handling and operation of these liquid nitrogen-cooled detectors. The opening height of the collimator can be varied from approximately 0.25 inches to 0.001 inches to control the dead time of the gamma-ray data acquisition system. The gamma-ray data are used to determine the relative axial distribution of fission and activation products in the measured item. The absolute efficiency of the gamma-ray spectroscopy system does not need to be determined for this purpose and it has not been.

Figure 6 shows the Cs-137 and Co-60 profiles for the measured items. The cesium distribution is indicative of burnup and shows a plausible shape for a fuel assembly including fuelled and non-fuelled regions. The cesium profile may provide a qualitative indication of a partial fuel mass defect if significant departures from an expected, smooth profile are found in fuel types in which cesium migration during irradiation is not expected. The cobalt distribution shows the same general shape except for its left end, which indicates the presence of relatively heavy activated stainless steel hardware at the top of the assembly. Average distributions are shown in Figure 6. The gamma-ray intensities varied with azimuthal angle by about 50 percent for Co-60, and 20 percent for Cs-137, reflecting the steep neutron flux gradient in the blanket region of EBR-II.

NEUTRON CALIBRATION MEASUREMENTS

The singles efficiency of the neutron detection system has been measured to be 8.5 percent using a Cf-252 source positioned at the geometric center of the detector. The dieaway time has been measured to be 56 microseconds. The axial sensitivity profile of the neutron detector is presented in Figure 4. The profile is roughly Gaussian-shaped with a slight asymmetry 10 to 25 inches above the detector center. This asymmetry is likely caused by neutron scattering from the lead shielding pieces in the scan shield/transition piece assembly. The full-width at half-maximum for the profile is approximately 16 inches.

VALIDATION MEASUREMENTS USING EBR-II BLANKET FUEL ASSEMBLY

Neutron coincidence and gamma-ray count profiles (Figures 5 and 6, respectively) have been measured on an EBR-II radial breeding blanket assembly of relatively low burnup. Isotopic contents of irradiated EBR-II fuel are routinely calculated using the Origen isotope depletion code (Reference 2). For this assembly, the Pu-240 mass has been calculated to be 0.89 grams. This item will later be sampled for determination of Pu-240 and other irradiation products by destructive analysis. Count times for each data point were approximately 30 minutes. In operation, a count time of 10 minutes per data point would suffice, for a total count time of 2 hours. For higher-burnup items, the count time could be significantly less. For most materials, a 2-hour total count time would be sufficient.

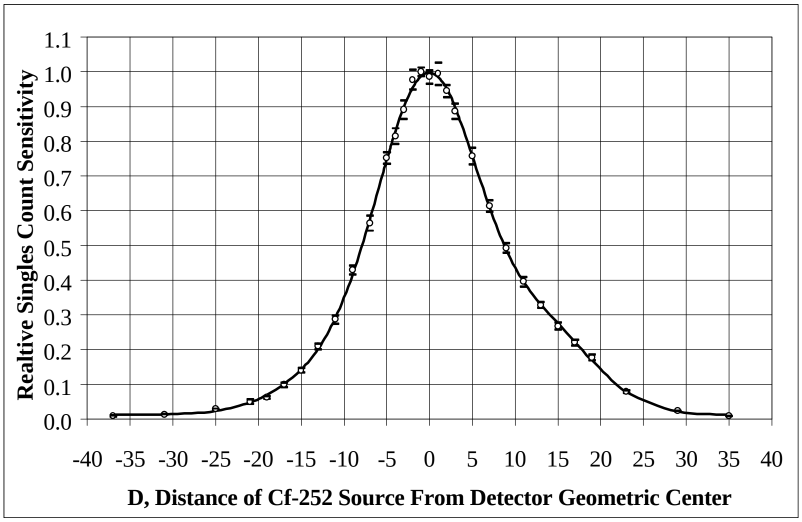

Figure 4, Neutron Measurement System Efficiency Profile

The neutron coincidence profile (Figure 5) shows the same general variation as the Cs-137 profile, as is to be expected, since both are indicative of burnup. Figure 5 also shows that removal of spallation neutron data often resulted in increased counts. A spallation neutron burst may appear within a short time of a triggering neutron event and be counted among the real plus accidental coincidences in the coincidence counter, or a burst may occur later, be counted among the accidental coincidences, and be subtracted from the reals plus accidentals. Removing a spallation burst in the first case decreases the coincidence count, and in the second case increases the count. One may thus expect some corrected counts to increase and others to decrease.

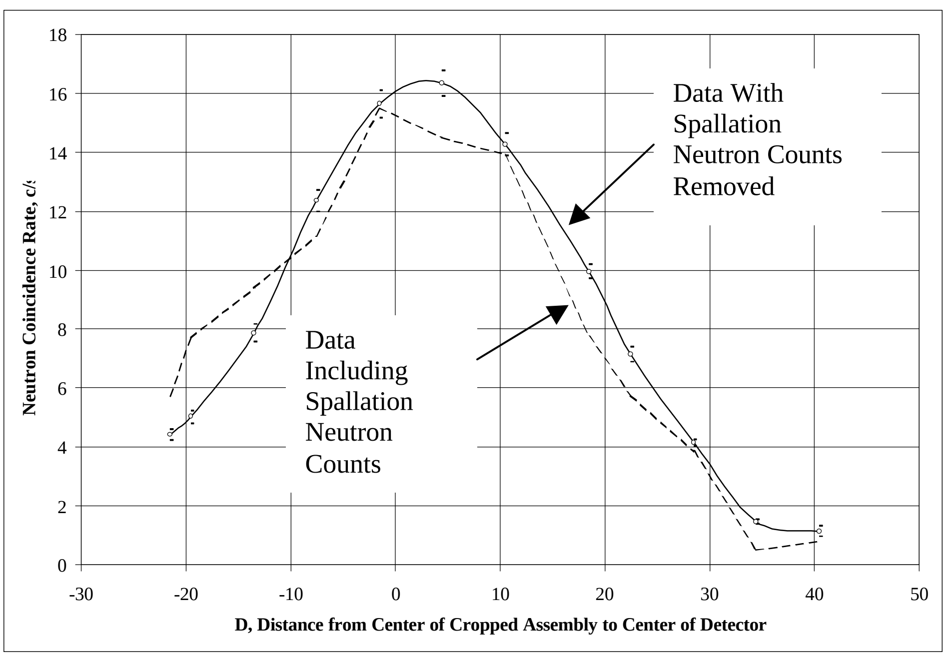

Figure 5, Neutron Coincidence Profile of Radial Blanket Assembly U1177.

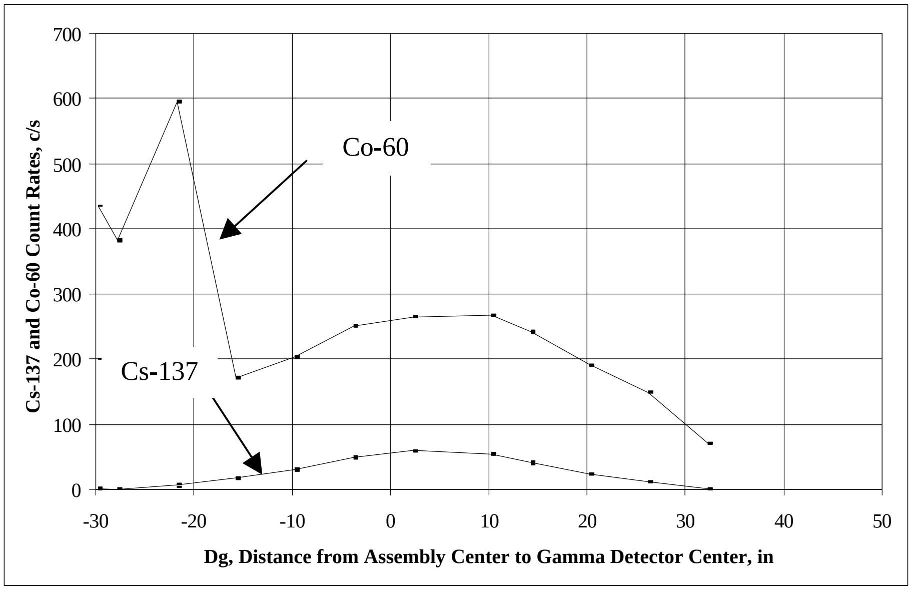

Figure 6, Gamma-ray Profiles of Radial Blanket Assembly U1177.

SUMMARY

This paper has described the design and use of the SMS. It has shown characteristics of the SMS neutron and gamma-ray measurement systems, described a method for removing spallation neutron noise from the count data, and shown results of the first validation measurements. The value of these measurements as signature measurements has also been shown. A more in-depth analysis of the validation measurement data awaits Monte Carlo detector response model calculations and destructive analysis of the blanket assembly. Fissile mass defect detection and other measurement capabilities of the SMS will be explored in the course of this further work.

DISCLAIMER

REFERENCES

[1] E.E. Habiger Letter to C. L. Huntoon, T. F. Gioconda, and M. A. Krebs, “Recommendations on Safeguarding Irradiated Nuclear Fuels”, August, 1999.

[2] R. D. McKnight, “ANL Calculational Methodologies for Determining Spent Nuclear Fuel Source Term, *Proceedings of the Embedded Topical Meeting on DOE Spent Nuclear Fuel and Fissile Material Management*, June 4-8, 2000, San Diego, CA, 253-258 (2000).

Shielded Measurement System Development and Calibration

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Measurements and Analysis Section

Introduction

- SMS developed to measure fissile contents of Irradiated Nuclear Fuel (INF)
- Response to DOE guidance
- Increased INF fissile material assurance desired
- INF fissile contents on measured basis
- INF increasingly moved to dry vs wet storage
- SMS designed for EBR-II fuel stored in underground liners at ANL-W
- SMS designed for measurements in dry storage
- SMS development funded by OSS TDP

Introduction

- SMS measurements
  - Neutron coincidence (and totals)
    - SMS designed for EBR-II fuel
    - Fast reactor spectrum- coincidence only from Pu-240
      - Blanket assemblies depleted but low-burnup
      - Driver assemblies high-burnup but high-enriched
      - Curies of Pu-240, millicuries/microcuries of Cm-242,-244
  - Gross gamma-ray dose (holdover from previous phase)
  - High-resolution gamma-ray spectra
    - Fuel at least 7 years cooling time
    - Cs-137 from fuel
    - Co-60 from clad
    - Eu-154 (weak)

Introduction

- Data interpretation
  - Detector properties from calibration measurements
    - Neutron- absolute efficiency, dieaway time
    - Gross gamma- dose as function of ion chamber current
    - High-resolution gamma- collimator view properties
  - Expected detector response from modeling
    - Isotope-depletion calculations predict source term in INF
    - Monte-carlo calculations predict detector response
  - Modeling process validated
    - Using calibration measurements (point sources)
    - Using validation measurements (well-characterized INF)

Introduction

- Data interpretation, continued
  - Expected detector responses calculated
    - INF source calculated by isotope-depletion (Origin) code
    - Detector resonses to INF source calculated by monte-carlo (MCNP) code
    - Neutron-coincidence profile along INF axis
    - Gamma-ray spectrum profile (Cs-137, Co-60, others)
  - Calculated and actual responses compared
    - Degree of agreement indicates confidence in source calculations
    - Fissile contents determined from calculations, confirmed by measurements
    - Diagnosis of disagreement- Cs-137 profile, direct Pu-240 measurement

SMS

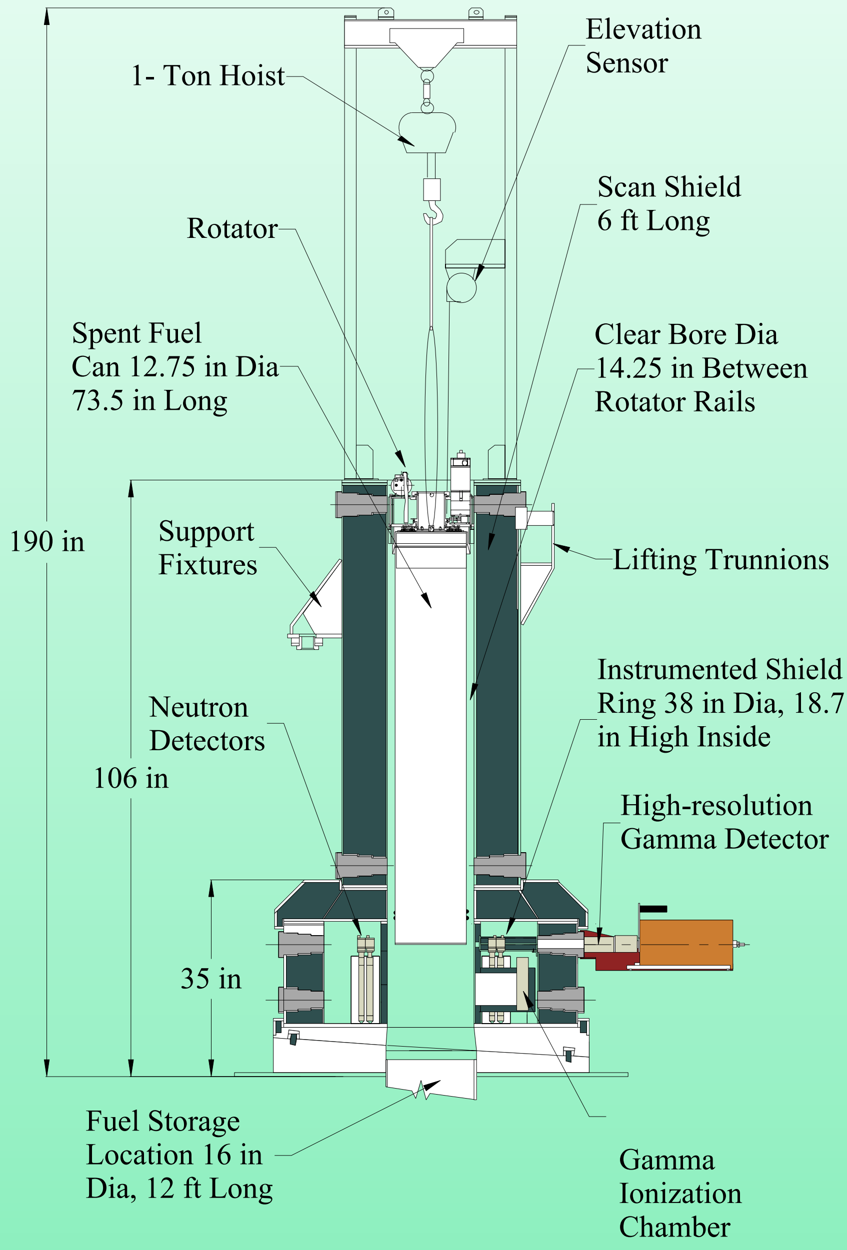
- Designed to measure EBR-II spent fuel
- Containers approximately 6 feet long, 13 in diameter
- Driver assemblies
  - Fuel 13.5 inches long
  - Initial enrichments 67 and 78 percent, metallic
  - Atom percent burnups to 10 routinely, 20 experimental
  - Stainless-steel clad
  - 2.5 kg total U, 0.5 g Pu-240
  - 61 elements 0.2 in diameter, usually stored in large-diametere circular array in basket

SMS Description

- **Radial blanket assembly**
  - Fuel approximately 60 inches long
  - Depleted uranium, metallic
  - Very low burnups
  - Stainless-steel clad
  - 47 kg total U
  - 0.9 g Pu-240, measured example
  - Up to 14 g Pu-240 in some examples
  - 19 elements 0.5 in diameter
  - Stored in original hexagonal assembly can
  - Usually 6 assemblies per storage can, hex layout

SMS Description

- Longitudinal section



SMS Description

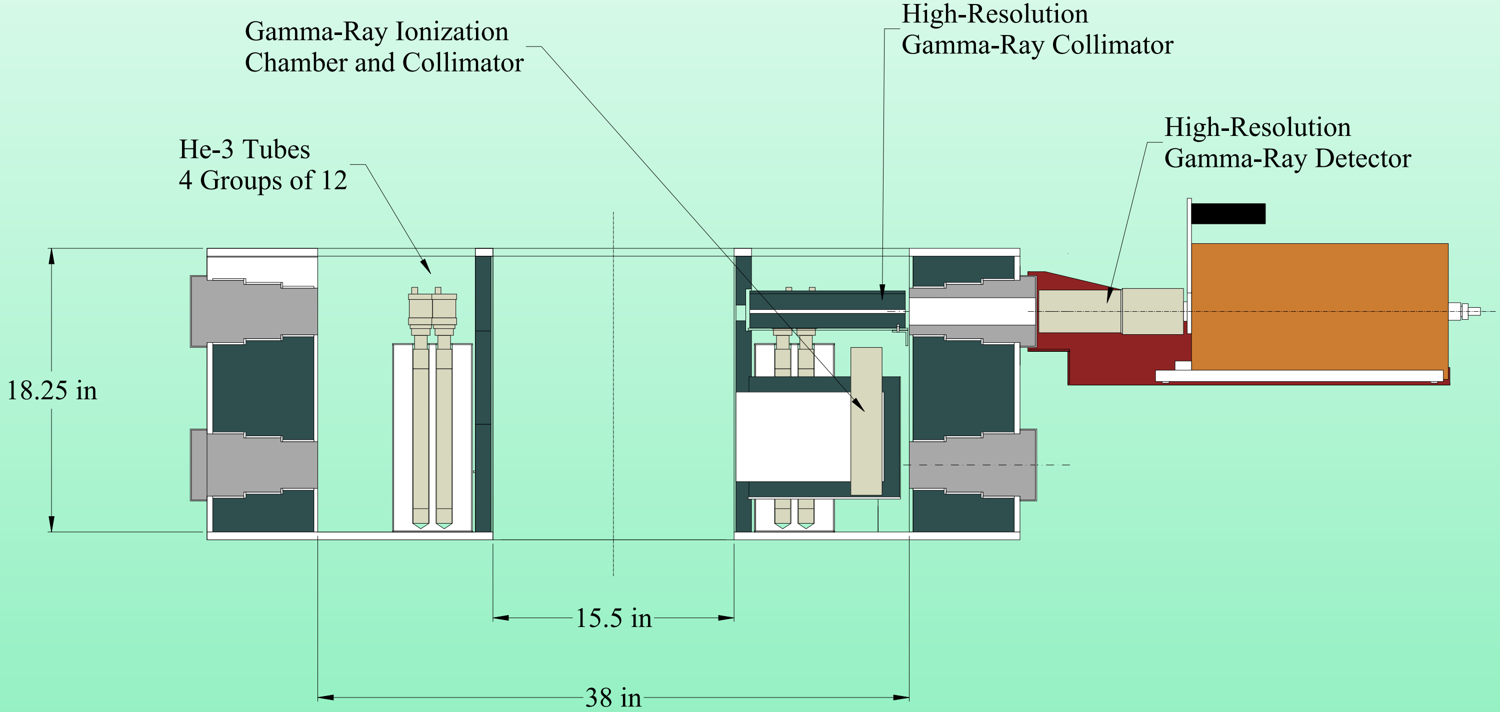
- **Photograph**

SMS Description

- **Components**
  - Base plate
    - Seismic stability, static load reduction
  - Funnel/shield-ring assembly
    - Leveling by rotating diagonally-split pieces
    - Guide funnel for item lifting and lowering
    - Shielding
  - Instrumented shield ring
    - Internal neutron detectors
    - Internal gamma ion chamber
    - External high-resolution gamma detector support
    - Penetrations for other detectors, cables

SMS Description

- Longitudinal section of detectors in ISR



SMS Description

- **Components, continued**
  - **Scan shield**
    - **Provides personnel shielding as item is scanned out the top of the ISR**
    - **Contains the item rotator**
    - **Supports the hoist frame assembly**
    - **Fitted with 4 penetrations for unusual measurement situations**
- **Total weight of SMS**
  - Approximately 19 tons
  - Limited by size and weight capacity of existing lifting systems at ANL-W

SMS Description

- Components, continued
  - Rotator
    - Supported by measured item storage can
    - Vacuum system + weight couples torque to can
    - Slides on rails in scan shield bore
    - Provides connection point for axial position transducer

SMS Description

- Photograph of top of rotator

SMS Description

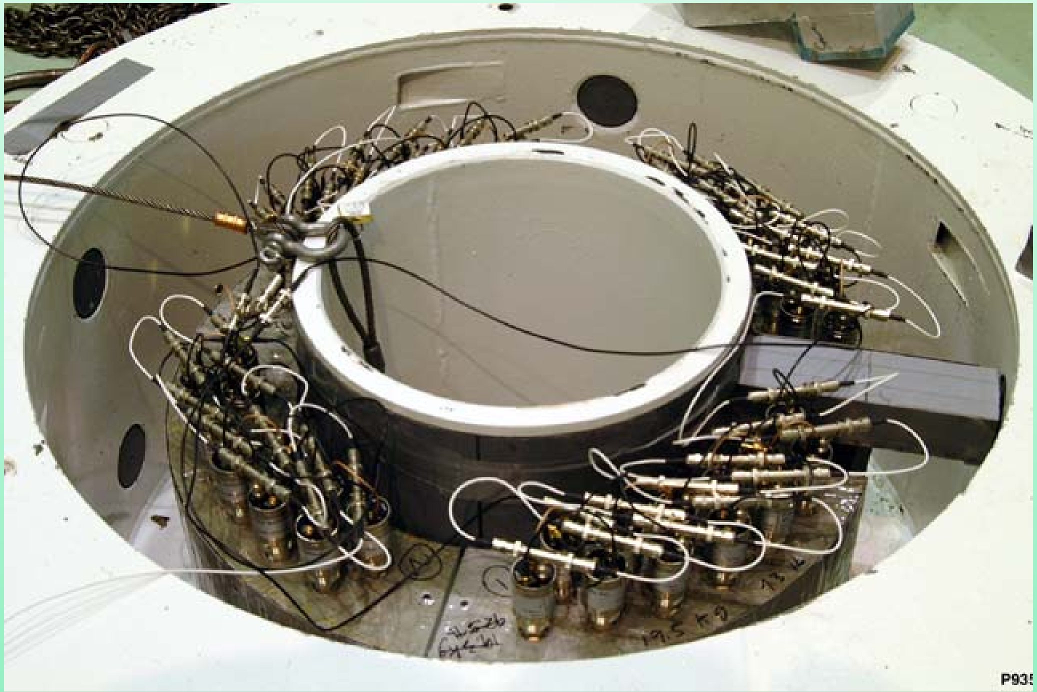
- Photograph of bottom of rotator

SMS Description

- **Neutron detector**
  - 48 He-3 tubes, 10-atmosphere, 8.5 percent efficiency
  - 4 sets of 12 tubes in two circular rows
  - Gaps between He-3 tube sets allow view of item through penetrations
  - 9-inch active length, 1-in diameter tubes
  - FWHM of approximately 16 inches for singles
  - Cadmium-wrapped polyethylene moderator prevents thermal self-interrogation of measured item
  - Tube-top electronics: signal-conditioning and amplifier/discriminator units
  - All amplifier/discriminator outputs collected together

SMS Description

- Photograph showing neutron detector



SMS Description

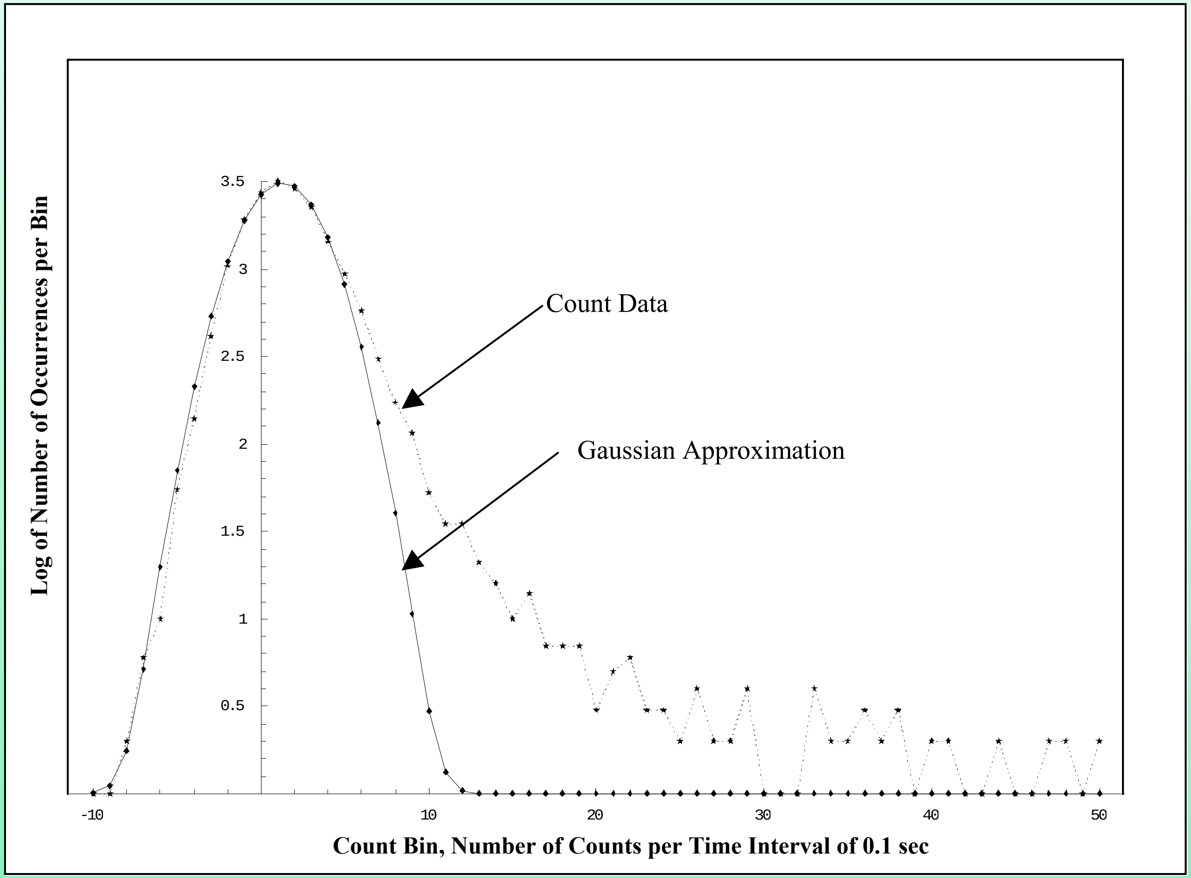
- Neutron detector, continued
  - Surrounded by approximately 18 tons of lead
  - Significant cosmic-ray spallation neutron count rate
  - High multiplicity counts at fairly low rate,
    - One every 10 to 30 sec
    - Apparent multiplicity of fifty to several hundred
    - Shift register greatly exaggerages high multiplicities
  - Continuous 5-minute counts exhibit high variance
    - Standard deviation of tens of percent
    - 1 or 2 percent expected from counting statistics

SMS Description

- Neutron detector, continued
  - Cosmic ray spallation neutron signal suppression
    - Count data in short time intervals (0.1 sec)
    - Histogram the resulting data
    - Throw out the "outlier" count data
    - Fission coincidence neutron data assumed to fit a normal distribution
    - Fit a gaussian to the main body of the histogram data
    - Count only data within +/- 3 standard deviations of the mean

SMS Description

- Log plot of count data and gaussian fit



SMS Description

- Neutron detector, continued
  - Cosmic ray spallation neutron signal suppression, continued
    - In the figure, count data from bins -4 to 11 would be used
    - Negative counts
      - Short count times
      - Net count is difference between two large numbers
    - Compared this method with long count
      - For high-rate Cf-252 source
      - Negligible spallation rate
      - Both methods statistically identical
      - Short count time case still showed some negative counts
    - Good noise reduction seen in neutron scans of INF items

SMS Description

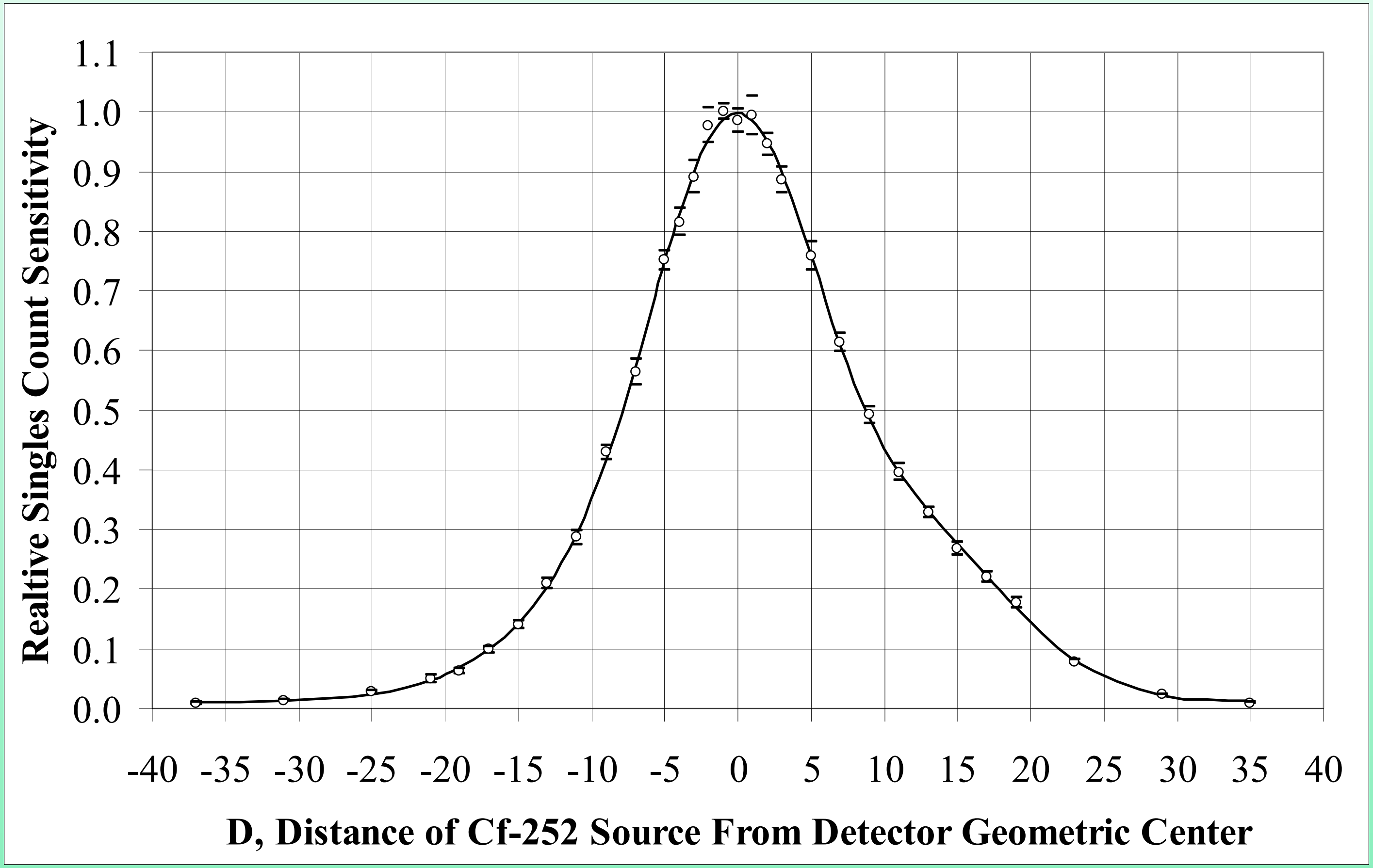
- Gross gamma-ray measurement system
  - Ionization chamber collimated to view approximately +/- 16 inches of item
  - Holdover from phase I of SMS, gross counts only
- High-resolution gamma-ray spectroscopy system
  - Detector mounted externally on ISR
    - Flexibility in detectors used
    - Accessible for liquid nitrogen filling
  - Internal rectangular collimator
    - Views 6-inch diameter at the centerline of the SMS
    - Vertical opening set using shims, 0.001 to 0.25 inches
  - 20 percent efficient coaxial detector used for this work

Calibration Measurements

- Neutron coincidence
  - 200 microCurie Cf-252 point source
  - Absolute efficiency 8.5 percent
  - Dieaway time 56 microseconds
  - Relative sensitivity profile
    - Roughly gaussian
    - Asymmetry 10 to 25 inches above center- neutron scattering from massive transition piece shielding
    - FWHM approximately 16 inches

Calibration Measurements

- Neutron measurement system efficiency profile



Calibration Measurements

- Gamma ionization chamber
  - Calibrated with Cs-137 point source
  - Calibrated together with picoammeter
  - Confirmed published dose-current relationship
  - Picoammeter used here not within calibration date
  - Dose measurements used only for general information

Calibration Measurements

- High-resolution gamma spectroscopy
  - Energy-channel calibration
    - Cs-137 point source, 9 microCurie
    - Co-60 point source, 9 microCurie
  - Absolute efficiency
    - Not needed, relative profile only
    - General idea of slit width vs activity per unit length of measured item
  - Geometric calibration
    - “Aiming point” on axis determined

Validation Measurements

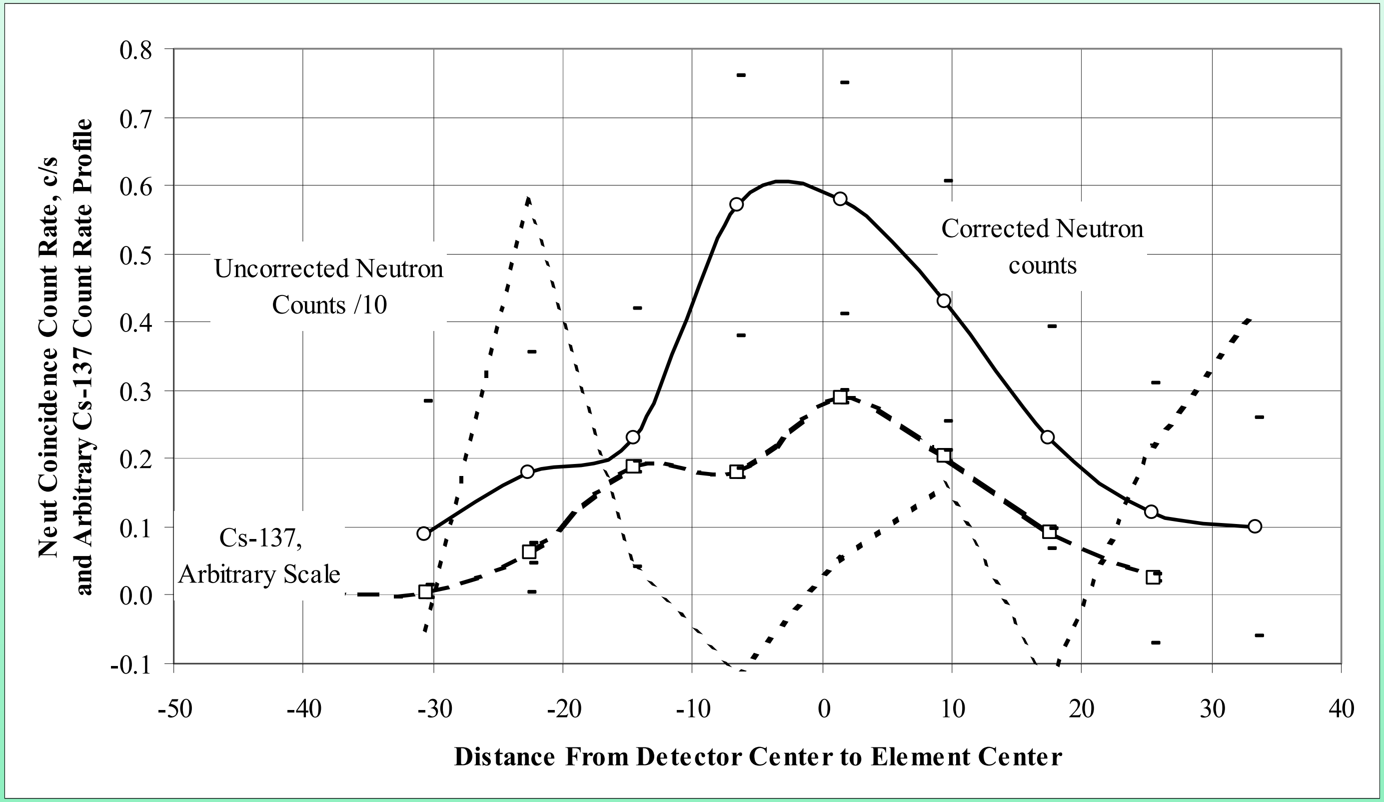
- Measure INF and cold fuel items
  - Real INF or cold mockups using U, Pu in desired ratios
  - INF composition, burnup distribution to be established by destructive analysis
- Accomplished
  - EBR-II blanket element
  - EBR-II blanket assembly (19 elements)
- Planned
  - EBR-II driver elements (61, in storage basket)
  - Cold mockups of blanket and driver
  - Cold assemblies of U, Pu items

Validation Measurements

- EBR-II blanket element
  - Medium-burnup example
  - One of 19 elements in original assembly
  - 0.5 g Pu-240 calculated using Origin code
  - Neutron coincidence scan data
    - Effect of removing spallation neutron counts shown by smoothness of plot
    - Corrected neutron coincidence profile very similar to Cs-137 profile and hence burnup

Validation Measurements

- Blanket element neutron coincidence profile



Validation Measurements

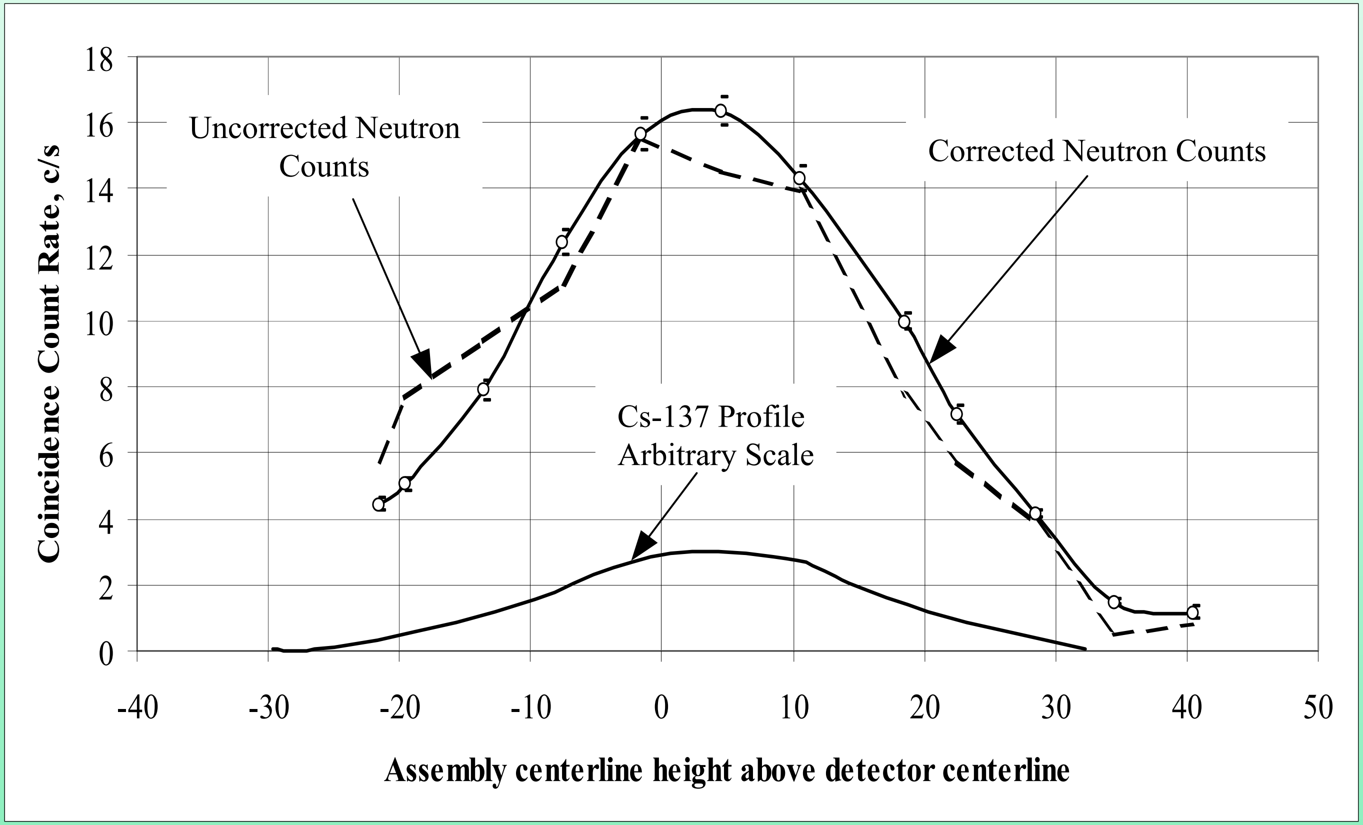
- EBR-II blanket assembly
  - Low-burnup example
  - 0.89 g Pu-240 calculated using Origin code
  - Neutron coincidence scan data
    - Effect of removing spallation neutron counts shown by smoothness of plot
    - Corrected neutron coincidence profile very similar to Cs-137 profile and hence burnup
    - Count time per segment 30 minutes
    - Count time could be reduced to 10 minutes with acceptable statistics, less for high-burnup assemblies
    - Apparent profile asymmetry due to shield plug neutron scattering and scan geometry.

Validation Measurements

- EBR-II blanket assembly
  - Gamma-ray scan data
    - Most gamma-ray emissions due to Co-60 in activated fuel clad
    - Gamma-ray emissions varied with azimuthal angle
      - Approximately 50 percent variation for Co-60 and total gamma-ray dose
      - Approximately 20 percent variation for Cs-137
    - High Co-60 count rate caused by increased hardware mass at top end of element
    - Total gamma-ray dose plot similar to that for Co-60
    - Neutron coincidence profile very similar to that for Cs-137
      - No increase in neutron detection rate at left end of plot
      - Neutron detectors successfully rejected gamma-rays

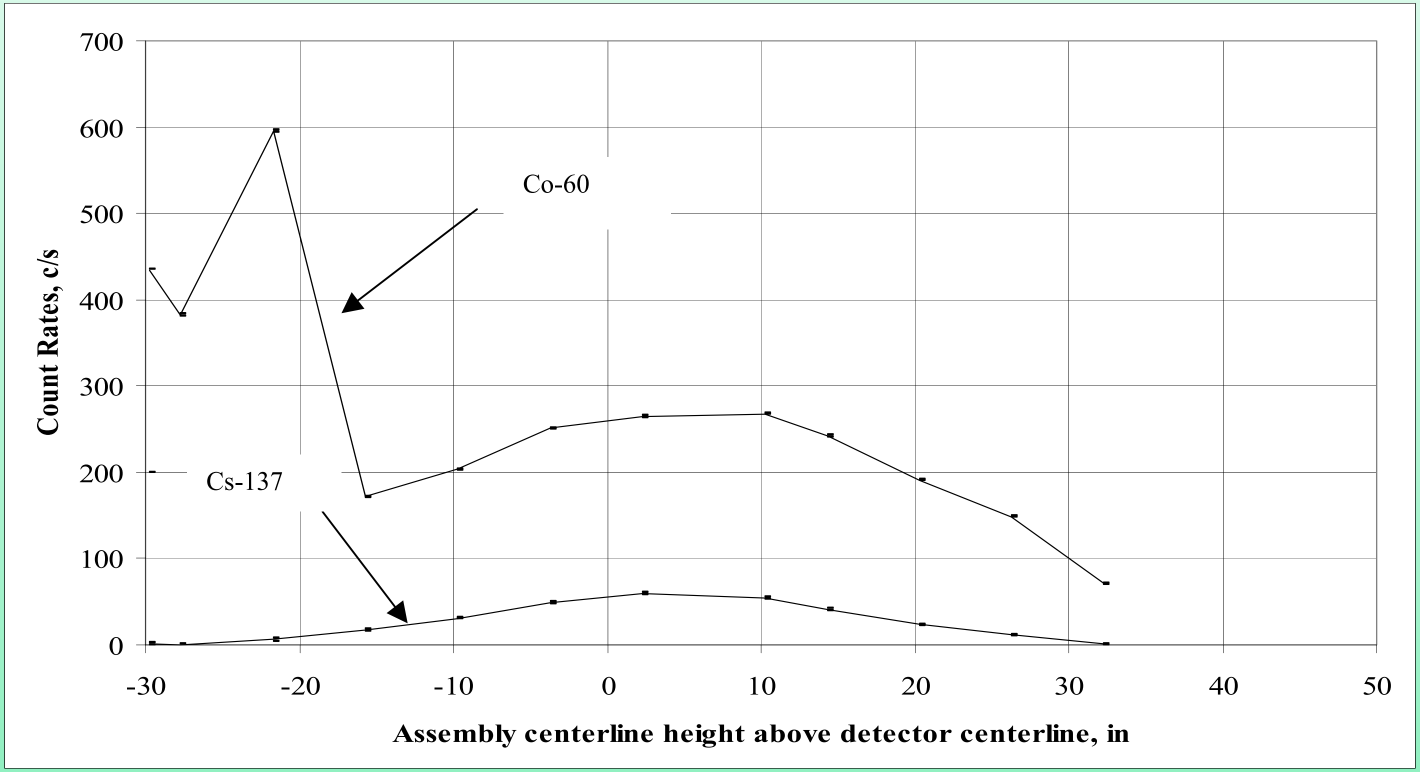
Validation Measurements

- Blanket assembly neutron coincidence profile



Validation Measurements

- **Blanket assembly gamma-ray profiles**



Summary

- Design of SMS described
- Measurement system characteristics described
- Spallation neutron count suppression method
- Example neutron and gamma-ray scans shown
- Future work
  - Measure cold materials
  - Destructive measurements on INF items
  - Perform isotope depletion and monte carlo simulations of measurements
  - Compare measurement and simulation results
  - Validate modeling method