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NMIS With Gamma Spectrometry For Attributes of Pu and HEU, Explosives and Chemical Agents

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April 24, 2002

EXECUTIVE SUMMARY

The concept for the Multiple Attribute System is a Nuclear Materials Identification System (NMIS) time-dependent coincidence processor that incorporates high resolution gamma ray spectrometry, proton recoil scintillation detectors, and in the active interrogation mode utilizes a small, lightweight, portable DT neutron (14.1 MeV) generator (capable of 1×10^7 n/s operations and less than 30 lbs. including power supply). This system, which is useful for highly enriched uranium (HEU) and plutonium measurements, could utilize a simple trusted processor that incorporates information barrier and authentication techniques using open software. The entire system, including the NMIS data acquisition board, can be built with commercially available components. In addition to use for fissile material, adding gamma ray spectrometry to the active system incorporates the ability to detect the presence of high explosive (HE), chemical agents, and, in some cases, drugs.

The system hardware and software can be configured to obtain the following attributes for Pu and HEU: Pu presence, Pu mass, Pu 240/239 ratio, Pu geometry, Pu metal vs. non-metal (absence of metal), time (age) since processing for Pu, U presence, U mass, U enrichment, U geometry, and U metal vs. non metal (absence of metal). The system hardware and software may also be configured to estimate a selected subset of these attributes. For bilateral uses, it could present the results as above (e.g., mass and HEU enrichment) or below (e. g., ^{240}Pu content) threshold values. The attributes measured, the method of determination, whether active or passive, and the measurement equipment involved are given in the following table. Some of these fissile attributes can be obtained by multiple data analysis methods. In addition, signatures from this system for fissile material can be used for template matching such as has been implemented for confirmation of inventories and receipts for weapons components at the Y-12 National Security Complex in Oak Ridge since 1996. Recently Y-12 personnel were trained and have been operating five NMIS systems at the Y-12 complex. The active gamma ray spectrometry method (activation analysis) is a fully developed commercially available methodology and is incorporated into this system with no significant increase in NMIS data accumulation time.

Two of the ten NMIS systems that exist are in Russia: one is located at the Russian Federal Nuclear Center All-Russian Scientific Research Institute of Experimental Physics (VNIIEF) and the other is located at the Zababakhin Russian Federal Nuclear Center All Russian Institute of Technical Physics (VNIITF). VNIIEF has already performed attribute analysis of NMIS measurements for Pu shells to obtain Pu mass and thickness. A passive system could be assembled and functionally tested in six months or less in preparation for passive measurements with fissile material.[1]

This passive system for measurements of Pu presence, Pu mass, Pu metal vs. non-metal and Pu 240/239 ratio, the latter by gamma ray spectrometry or time-dependent coincidence methods consists of two $500 \times 500 \times 80$-mm-thick proton recoil scintillators and a HPGe

[1] Estimates are times to perform technical work without delay introduced by other considerations which are not predictable.

detector for Pu 240/239 ratio and the hardware to process the time-dependent coincidence data (NMIS) and multichannel analyzer MCA for gamma-ray spectrometry.

An active system adds an additional small proton recoil scintillation detector for transmission and a small, lightweight, portable DT neutron generator to the passive system and thus extends the capability to HEU, HE, and chemical agents. The transition to active could take an additional two months. Building a system by augmenting existing system would reduce the time to six months. The existing non-fissile activation analysis methods using the DT neutron generator can be easily incorporated for HE and chemical agents.

This system is useful for a variety of nuclear material control and accountability applications since this one system detects and determines many attributes of fissile material and HE and chemical agents. This system applicability to a single cargo container would require a larger source (10^8 n/s) and detectors (>1 meter square).

Matrix of Attribute Measurements by NMIS with Gamma Spectrometry

Material	Attribute	Method (Option, Implementation, Basis)			Active or Passive	Measurement Equipment
plutonium	presence	1	time-dependent coincidence	detect internal spontaneous fission	active or passive	neutron source, scintillation detectors, time-correlator
		2	gamma spectrometry	detect Pu spectral lines	passive	high-resolution gamma detector, multi-channel analyzer
	fissile mass	1	time-dependent coincidence	measure induced fission rate	active	neutron source, scintillation detectors, time-correlator
		2	time-dependent coincidence	measure spontaneous fission rate	passive	scintillation detectors, time-correlator
	age	1	gamma spectrometry	measure in- and out-growth of impurities	passive	high-resolution gamma detector, multi-channel analyzer
	metal / non-metal	1	neutron-initiated gamma spectrometry	detect 6129 KeV gamma from 14.1 MeV neutron interactions with O & F	active	neutron source, high-resolution gamma detector, multi-channel analyzer
		2	time-dependent coincidence	measure density from neutron transmission	active	neutron source, scintillation detector, time-correlator
		3	time-dependent coincidence	attenuation of gammas emitted and multiplication depend on density	passive	scintillation detectors, time-correlator
		4	gamma spectrometry	detection of the 2438 keV gamma ray from the (α,n) reaction on oxygen	passive	high-resolution gamma detector, multi-channel analyzer
	geometry	1	time-dependent coincidence	measure axial density gradient from neutron transmission	active	neutron source, scintillation detector, time-correlator
	thickness	1	time-dependent coincidence	time-dependent coincidence depends on thickness and mass	passive	scintillation detectors, time-correlator
	relative ^{240}Pu-content	1	time-dependent coincidence	compare spontaneous and induced fission rates	active	neutron source, scintillation detectors, time-correlator
		2	gamma spectrometry	compare ^{240}Pu and ^{239}Pu spectral lines	passive	high-resolution gamma detector, multi-channel analyzer
		3	time-dependent coincidence	integral and width of distribution primarily related to ^{240}Pu and ^{239} Pu mass, respectively	passive	scintillation detectors, time correlator

Matrix of Attribute Measurements by NMIS with Gamma Spectrometry (Cont'd.)

Material	Attribute	Method (Option, Implementation, Basis)			Active or Passitve	Measurement Equipment
uranium	**presence**	1	time-dependent coincidence	detect induced fission and absence of internal spontaneous fission	active	neutron source, scintillation detectors, time-correlator
	fissile mass	1	time-dependent coincidence	measure induced fission rate	active	neutron source, scintillation detectors, time-correlator
	^{235}U-enrichment	1	time-dependent coincidence	compare induced fission rates and neutron transmission	active	neutron source, scintillation detectors, time-correlator
	metal / non-metal	1	neutron-initiated gamma spectrometry	detect 6129 KeV gamma from 14.1 MeV neutron interactions with O & F	active	neutron source, high-resolution gamma detector, multi-channel analyzer
		2	time-dependent coincidence	measure density from neutron transmission	active	neutron source, scintillation detectors, time-correlator
		3	time-dependent coincidence	attenuation of gamma emitted and multiplication depend on density	active	neutron source, scintillation detector, time-correlator
	geometry	1	time-dependent coincidence	measure axial density gradient	active	neutron source, scintillation detectors, coincidence gate
high explosive	**presence**	1	gamma spectrometry	N presence and ratios of N/C, H/C, and O/C	active	neutron source, HPGe[a] or BGO[b]
chemical weapon	**presence**	1	gamma spectrometry	ratios of N/C, H/C, and O/C	active	neutron source,HPGe[a] or BGO[b]
druge	**presence**	1	gamma spectrometry	ratios of N/C, H/C, and O/C	active	neutron source, HPGe[a] or BGO[b]

[a]Methods of A. J. Caffrey; see Reference 4.
[b]G. Vourvopolos and P. C. Womble, "Pulsed Fast/Thermal Neutron Analysis: A Technique for Explosive Detection," Applied Physics Institute, Western Kentucky University, Bowling Green, KY 42101 (2001).

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1. INTRODUCTION

The concept for the system described herein is an active/passive Nuclear Materials Identification System[2] (NMIS) that incorporates gamma ray spectrometry[3]. This incorporation of gamma ray spectrometry would add existing capability into this system. This Multiple Attribute System can determine a wide variety of attributes for Pu and highly enriched uranium (HEU) of which a selected subset could be chosen. This system can be built using commercial off the shelf (COTS) components. NMIS systems are at All-Russian Scientific Research Institute of Experimental Physics (VNIIEF) and Russian Federal Nuclear Center Institute of Technical Physics, (VNIITF) and measurements with Pu have been performed at VNIIEF and analyzed successfully for mass and thickness of Pu. NMIS systems are being used successfully for HEU at the Y-12 National Security Complex. The use of active gamma ray spectrometry for high explosive HE and chemical agent detection is a well known activation analysis technique, and it is incorporated here.

This report describes the system, explains the attribute determination methods for fissile materials, discusses technical issues to be resolved, discusses additional development needs, presents a schedule for building from COTS components, and assembly with existing components, and discusses implementation issues such as lack of need for facility modification and low radiation exposure.

1.1 BACKGROUND

1.1.1 Past Experience With Uranium And Plutonium Metal

Measurements of the time distribution of counts in a detector near a fissile assembly after a Cf fission in an ionization chamber also near the fissile assembly were first performed at the ORNL in 1968 with enriched uranium metal assemblies. The methodology at that time was referred to in the literature as the randomly pulsed neutron measurements because of the randomness of the ^{252}Cf fission events in the ionization chamber.[4] Prior to and subsequent to 1968, measurements of the time distribution of counts in one detector after a count in another detector or the same detector (the so called Rossi-α method) were performed at ORNL.[5] Through the 1960s and 1970s, thousands of these type measurements were performed with enriched uranium metal. Throughout the 1960s, pulsed neutron measurements for enriched uranium metal were also performed utilizing a DT neutron generator.[6] These three time-

[2] J. T. Mihalczo, J. A. Mullens, J. K. Mattingly, and T. E. Valentine, "Physical Description of Nuclear Materials Identification System (NMIS) Signatures, Nuclear Instruments & Methods in Physics Research A 450 (2000) 531-555.

[3] J. A. Mullens, J. E. Breeding, R. B. Perez, J. T. Mihalczo, T. E. Valentine, and J. A. McEvers, "A Multipurpose Processor for Arms Control and Nonproliferation and NMC&A," paper presented at the Institute of Nuclear Materials Management Conference, Phoenix, Arizona, July 25-30, 1999.

[4] J. T. Mihalczo, "The Use of ^{252}Cf as a Randomly Pulsed Neutron Source for Prompt Neutron Decay Measurements,: *Nucl. Sci. Eng*., **41**, 296-298 (1970).

[5] J. T. Mihalczo, "Prompt Neutron Decay and Reactivity Measurements in Subcritical Uranium Metal Cylinders," *Nucl. Sci. Eng.,* **32,** 292-301 (1968).

dependent coincidence methods involve detection of prompt neutrons and gamma rays from fission that are more numerous (by a factor of 500) than delayed neutrons.

In measurements with plutonium metal in early 1970s, the ^{240}Pu content varied from 1.5 to 20 wt% and the mass varied from 2.2 to 16 kg of Pu metal.[6] In the experiments with uranium metal, the enrichment varied from depleted to 93.2 wt% of ^{235}U and the masses varied over a wide range. Measurements were also performed in the early 1990s [7] with two 4 kg rings of Pu metal, one of which was mainly ^{242}Pu.

These measurement methods, for which ORNL has decades of measurement experience, are the basis of the present NMIS processor. The same functions are measured: time-dependent coincidence distribution between a detector and the source (randomly pulsed neutron method), the time-dependent coincidence between two detectors (two detector Rossi-α method), and time-dependent coincidence between pulses in a single detector and a previous pulse in the same detector (one detector Rossi-α measurement). The present NMIS processor measures these same quantities with modern day chip technology for up to five detector channels, with the option of one channel being the neutron source channel for active measurements. These two measurement methods define the NMIS methodology. The only difference between the past and the present configuration is the hardware (1 GHz synchronous sampling of five channels required for prompt time-dependent coincidence distribution measurements for fissile metal), software, and data analysis methods. The quantities from NMIS measurements have been measured at Oak Ridge with other hardware for decades (since the 1960s).

1.1.2 NMIS Experience With U And Pu Metal

There is considerable experience using the existing NMIS processors, first assembled in 1995, with Pu and highly enriched uranium metal. NMIS in the template-matching mode has been used for confirmation of inventory for weapons components at the Y-12 National Security Complex since 1996 for nuclear material control an accountability (NMC&A) to meet the requirements of the Department of Energy (DOE) orders.[8] Six years of template matching applications have occurred for large number of weapons components. NMIS has also been used at the Y-12 Complex for confirmation of 100% of the receipts from Pantex since 2000.

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[6] J. T. Mihalczo, "The Use of Californium-252 as a Randomly Pulsed Neutron Source for Prompt-Neutron Decay Measurement," *Nucl. Sci. Eng*., **53,** 393-414 (1974).

[7] J. T. Mihalczo, V. K. Pare, E. D. Blakeman, and T. E. Valentine, "Time and Frequency Analysis Measurements for a ^{242}Pu Metal Ring Using ^{252}Cf," Institute of Nuclear Materials Management 37th Annual Meeting, Naples, FL, July 1996.

[8] J. A. Mullens, J. K. Mattingly, L. G. Chiang, R. B. Oberer, and J. T. Mihalczo, "Automated Template Matching Method for NMIS at the Y-12 National Security Complex," Institute of Nuclear Materials Management, Indian Wells, CA, July 15-19, 2001.

In 1998 NMIS was used to determine the fissile mass and ^{235}U enrichment of a large number of HEU samples at the Y-12 Complex to ± 1.5% relative standard deviation.[9] This was the first determination of enrichment of uranium by methods other than mass or gamma ray spectrometry.

In 1999 NMIS was used for the first time for facility-to-facility transfers where reference templates were acquired at the shipper's site.[10] Upon receipt at the Y-12 Complex, NMIS signatures were matched to the reference templates for confirmation of these weapons components. In addition, NMIS was successfully blind tested by the Defense Special Weapons Agency (DSWA) in 1997 at the Los Alamos National Laboratory (LANL) on pits in containers.[11] It not only determined that the items matched or did not match the declarations but also successfully stated what the items were, based only on data acquired during the LANL measurements.

NMIS was field deployed at Pantex in 1998 for measurements of Pu and HEU weapons components and fully assembled nuclear weapons systems. This application required nuclear explosive safety approval of the use of NMIS with fully assembled nuclear weapons.

In addition, NMIS at VNIIEF was used in the year 2000 for unclassified spherical shell assemblies of Pu and HEU metal, where Pu attribute extraction methods for fissile mass and thickness were demonstrated. Measurements were also performed in AT400 containers with heavily borated inserts. Pu masses of these systems were estimated from passive measurements only and are discussed in Appendix A and Ref. 13.[12] These attributes were also determined by Russian scientists from active measurements and are discussed in Ref. 14 and Appendix C.[13] In addition, there were NMIS measurements of pits in containers by VNIIEF personnel in 1999. In February 2002, NMIS was demonstrated at VNIIEF in the template matching mode for three Pu classified components to a US delegation, and the demonstration at VNIIEF with classified parts was initially scheduled on April 23, 2002 to the US Ambassador to Russia.

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[9] J. K. Mattingly, T. E. Valentine, J. T. Mihalczo, L. G. Chiang, and R. B. Perez, "Enrichment and Uranium Mass From NMIS for HEU Metal," Institute of Nuclear Materials Management Conference, New Orleans, Louisiana, July 16-20, 2000.

[10] L. G. Chiang, J. K. Mattingly, J. A. Mullens, and J. T. Mihalczo, "NMIS Experience for Facility-to-Facility Transfer," Institute of Nuclear Materials Management, Indian Wells, CA, July 15-29, 2001.

[11] J. A. Mullens, J. T. Mihalczo, T. E. Valentine, and J. K. Mattingly, "Successful Blind Testing of NWIS for Pits at Los Alamos National Laboratory," 1998 Institute of Nuclear Materials Management Conference, Naples, Florida, July 26-30, 1998.

[12] J. K. Mattingly, J. T. Mihalczo, L. G. Chiang, and J. S. Neal, "Preliminary Analysis of Joint RFNC-VNIIEF/ORNL Measurements Performed in Year 2000," Y/LB-16,067, Y-12 National Security Complex, September 2001.

[13] V. V. Gurov, M. I. Kuvshinov, V. A. Popov, V. P. Dubinin, J. K. Mattingly, and J. T. Mihalczo, "VNIIEF-ORNL Joint Plutonium Measurements with NMIS and Results of Plutonium Attributes Preliminary Evaluations," July 2001.

1.2 FISSILE ATTRIBUTES ESTIMATED

The system hardware and software can be configured to obtain the following attributes: Pu presence, Pu mass, Pu 240/239 ratio, Pu geometry, Pu metal vs. non-metal (presence of oxygen or absence of metal), time since processing, U presence, U mass, U enrichment, U geometry, U metal vs. non-metal (oxide or fluoride or absence of metal). Some of these attributes can be obtained by multiple data analysis methods. The system hardware and software can be configured to estimate a selected subset of these attributes.

1.3 SYSTEM DESCRIPTION

The system is a NMIS time-dependent coincidence distribution processor that incorporates gamma ray spectrometry and utilizes a small, lightweight, portable DT neutron (14.1 MeV) generator (1×10^7 n/s and weighs less than 30 lbs. including power supply), and proton recoil scintillation detectors. For some applications such as large containers, a 10^8 n/s source would be required. This system could utilize a trusted processor with information barrier and authentication with no proprietary software. Since it is constructed of commercially available components, it can be Russian built including the NMIS data acquisition boards.

The system consists of three functional subsystems: (1) gamma ray spectrometry which will only be described for determining the presence of oxygen and fluorine in fissile materials (Fig. 1.1) and referenced for other attributes such as for HE and chemical agents; (2) two large detectors and a neutron source for time-dependent coincidence measurements (Fig. 1.2); and (3) a third detector and the same source for transmission/scanning measurements (Fig. 1.3).

In addition to the Pu age (time since reprocessing), Pu 240/239 ratio, and presence of Pu applications of gamma ray spectrometry, the system uses 14.1 MeV neutrons to detect the presence of PuO_2, U_3O_8, and UF_4 by source event time-gated gamma ray spectrometry measuring the 6129 KeV gamma ray from neutron interactions with oxygen and fluorine.

The second subsystem, consisting of the source and large detectors, differentiates enriched uranium from plutonium, estimates (a) relative Pu 240 content, (b) Pu mass, (c) ^{235}U enrichment, (d) fissile mass of uranium, and (e) absence of metal. The third subsystem for active transmission/scanning measurements determines geometry and absence of metal also. Subsystems share hardware, but software analysis methods are separated.

1.4 DATA ACQUIRED

The system incorporates gamma ray spectrometry data as well as data from the NMIS processor. The NMIS data includes time-dependent coincidence methods that obtain the time distribution of counts after a detection of an event in another detector, the same detector, or the source. In addition, NMIS now includes third order time-dependent coincidence

methods using coincidence in three detectors (including the source).[14] In addition, the number of times n pulses are acquired in a time interval are also measured and from these multiplicities can be obtained. These data acquired by NMIS have been described in detail in Ref. 3. The same basic signatures that are used for attribute estimation can be used for template matching.

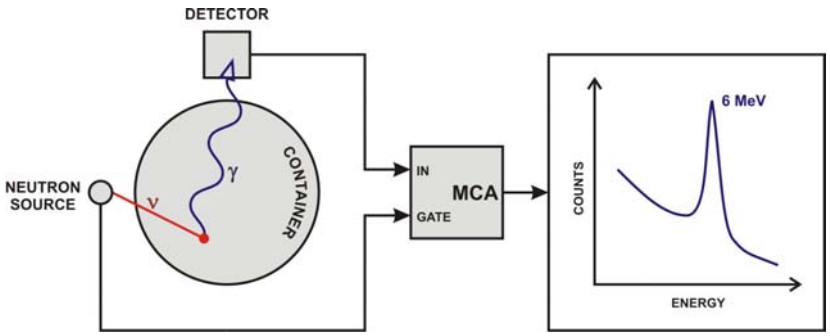


Fig. 1.1. The Functional Design of a Gamma Ray Spectrometry Measurement System For Detection of High Explosives, Chemical Agents, in Some Cases Drugs, and Fissile Oxides and Fluorides. [The system employs an active neutron source (DT neutron generator equipped with an associated particle detector), a HPGe gamma detector, and a multi-channel analyzer with appropriate software.]

[14] J. K. Mattingly, "Multivariate High Order Statistics of Measurements of the Temporal Evolution of Fission Chain-Reactions," ORNL/TM-2001, 45, March 2001.

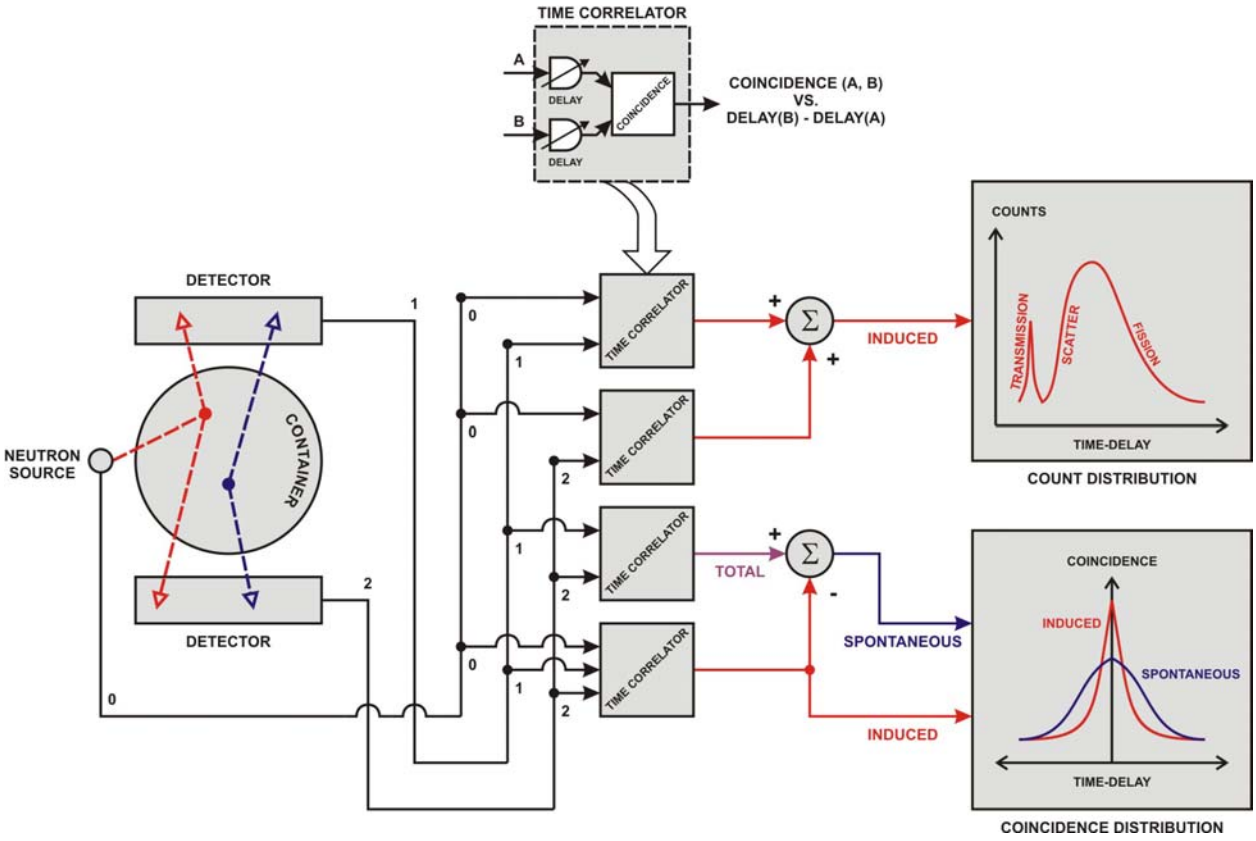


Fig. 1.2. The Functional Design of a Measurement System to Differentiate Plutonium From Uranium, to Estimate Relative ^{240}Pu-Content and Fissile Mass of Plutonium Samples, and to Estimate ^{235}U-Enrichment and Fissile Mass of Uranium Samples. [The system employs an active monoenergetic neutron source (DT generator equipped with an associated particle detector for detection of the time of emission of a neutron and its direction) and proton recoil scintillation detectors to acquire the time-distribution of detector counts induced by the active source and to acquire the time-distribution of detector coincidence partitioned into mutually exclusive induced and spontaneous components. Note that in the absence of plutonium, the spontaneous component of the time-dependent coincidence-distribution vanishes.]

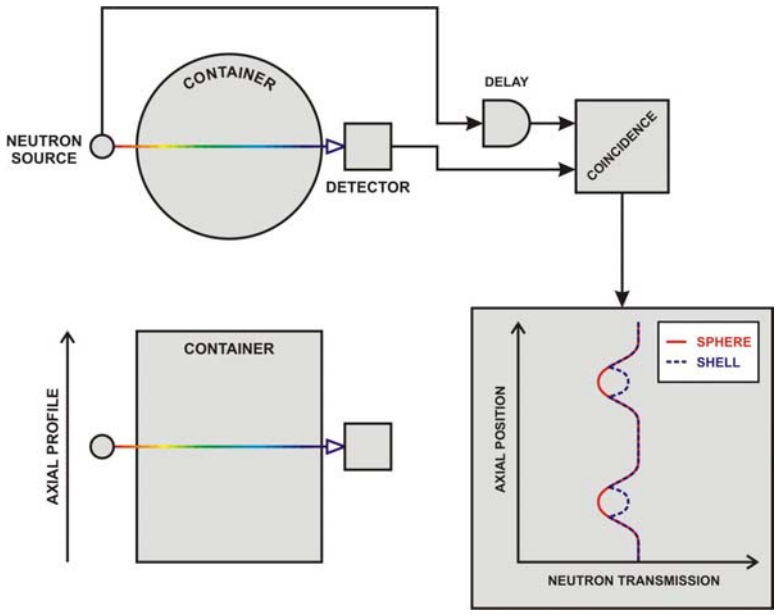


Fig. 1.3. The Functional Design of a Measurement System to Recognize the Geometry of Plutonium and Uranium Samples by Measuring the Axial Density Gradient Using Neutron Transmission. [Note the system could easily distinguish shells from spheres and also recognize rotational symmetry by rotating the inspected container and verifying the independence of transmission upon rotation angle (but this may not be necessary). The system employs an active neutron source (DT generator equipped with an associated particle detector) and a single proton recoil scintillation detector or a vertical array of small proton recoil scintillation detectors.]

2. CONFIGURATION OF ACTIVE/PASSIVE SYSTEM

This system is a combination of a NMIS system and gamma ray spectrometry utilizing the high-purity germanium (HPGe) methodology developed by other national laboratories. It employs a DT 14.1 MeV neutron generator as an active source and four detectors: two large proton recoil scintillators, a small proton recoil scintillator(s) for transmission measurements, and an HPGe system. A top view of the arrangement of the source and detectors is given in the following conceptual sketch (Fig. 2.1).

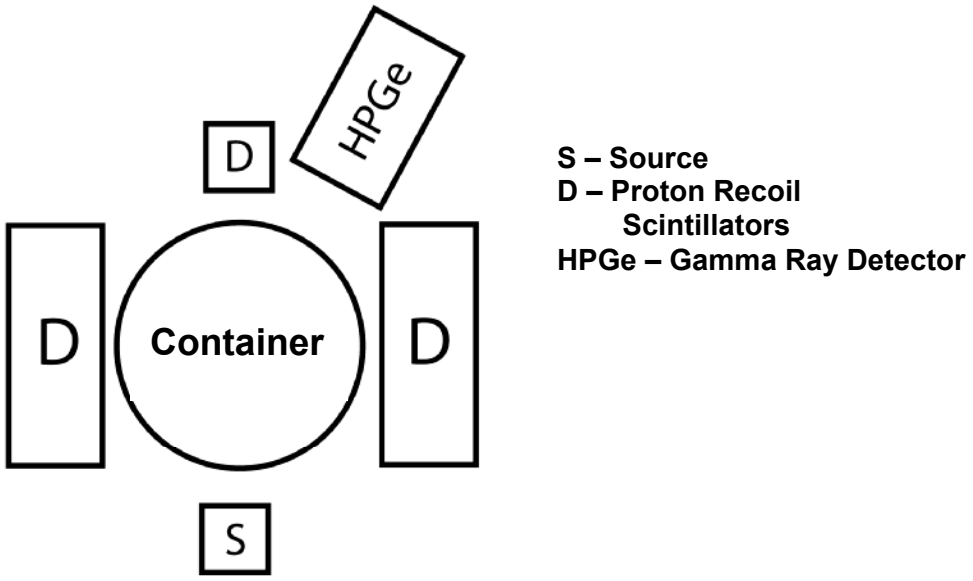


Fig. 2.1. Sketch of Configuration of System

2.1 DETECTORS

The detectors are three proton recoil scintillation detectors for NMIS type signatures and one HPGe detector for gamma ray spectrometry. All detectors are commercially available (COTS). Two of the proton recoil detectors are 500 × 500 × 80-mm-thick and are stationary on each side of the container. A third 100 × 100 ×80-mm-thick detector is for scanning to measure transmission and thus to obtain the geometry of fissile material. The proton recoil scintillators could utilize pulse shape discrimination. The HPGe detector is conventional and could be supplied with existing open software.

2.2 DT GENERATOR

The reliability and portability of present DT sources makes it practical to replace the existing ^{252}Cf source used with NMIS with a small, portable DT neutron (14.1 MeV) source.[15] This has a variety of advantages over the ^{252}Cf and is more useful for active measurements. For some containers, the presence of hydrogenous material in the container reduces the ability of ^{252}Cf source neutrons to penetrate the container and its inserts, and to induce fission in the fissile sample. Rather than raise the ^{252}Cf source intensity to compensate for increased attenuation, utilizing 14.1 MeV neutrons (which are more efficient in passing through hydrogenous material) is a way to limit the source size and thus reduce radiation exposure in active measurements. Between 2 MeV and 14.1 MeV, the total neutron cross section of hydrogen decreases by a factor of 4, allowing easier neutron penetration of materials containing hydrogen and thus lower source size. In addition, there are many other advantages to the use of a DT neutron generator of this type with NMIS which are associated with increased sensitivity: reduction of correlated background from floor and nearby objects,[16] reduced radiation exposure (see Section 10.6), and cost effectiveness. Since NMIS detects prompt neutron and gamma rays from fission, there is a factor of ~500 more particles available for detection than for measurement systems that utilize delayed neutrons.

A sketch of the DT neutron generator model under fabrication at Thermal MF Physics Corporation is given in Fig. 2.2. It is a cylinder 40 inches long by 3 inches in diameter. The neutron tube has a built-in alpha particle detector that is used to provide direction and emission time of some of the neutrons produced at the tube target. The alpha detector spatial resolution will only define a cone of neutrons (i.e. neutron 180° from the detected alpha particle). A mixed tritium/deuterium metal hydride target is oriented at 45 degrees to the face of the alpha particle detector.

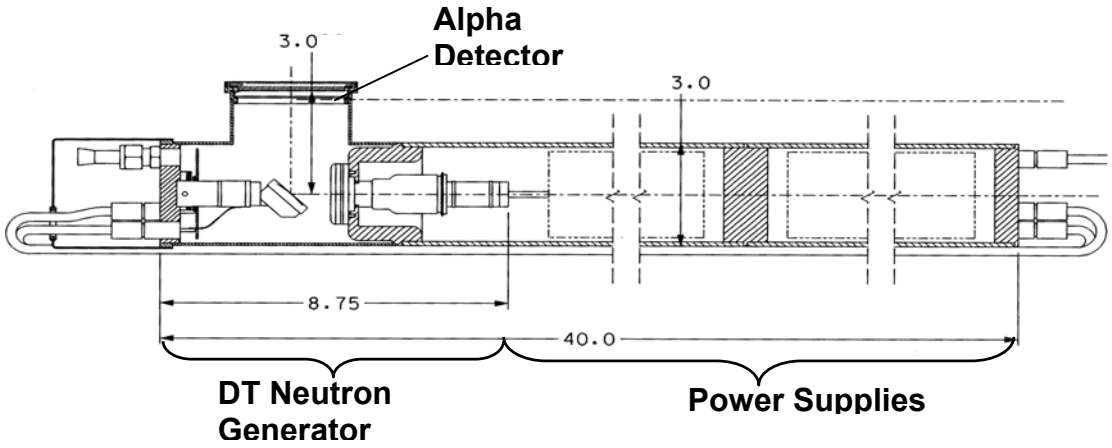


Fig. 2.2. Sketch of a Small, Portable, Lightweight DT Generator

[15] J. Reichardt, J. T. Mihalczo, R. B. Oberer, L. G. Chiang, and J. K. Mattingly, "Small, Portable, Lightweight DT Neutron Generator For Use With NMIS," Institute of Nuclear Materials Management, Indian Wells, CA, July 15-19, 2001.

[16] Sara Pozzi, "Preliminary MCNP-POLIMI Simulations for the Evaluation of the 'Floor Effect': Comparison of APSTNG and Cf Sources," ORNL/TM-2002/18, January 2002.

The design of the ion optics and the ion source is adapted from that used in the Thermo MF Physics Corp. miniature neutron tube, the A-3062, that is presently used primarily in very small neutron generators for oil field applications. The neutron tube, target assembly, and alpha particle detector are physically integrated as a sealed vacuum device. The accelerator section and ion source are welded onto the target chamber such that the entire structure may be evacuated to ultra-high vacuum.

The high voltage power supply is attached directly to the neutron tube and is inside the grounded pipe. The neutron tube and power supply are immersed in liquid Fluorinert FC-40 for dielectric insulation. Use of a liquid dielectric, rather than high-pressure gas such as SF_6, precludes any safety and transport constraints brought about by the use of high-pressure gas. The accelerator high voltage power supply can provide up to 75 kV accelerating potential with a maximum of 30 microamperes beam current. The neutron yield is 1×10^7 neutrons per second. Under these conditions, the expected operating life of the neutron tube is four thousand hours.

The neutron generator is entirely self-contained. All electronic circuitry necessary to control the neutron generator is integrated into the 3-in.-diameter, 40-in.-long package. The only input requirements are 50 watts power at 110 Volts AC and an adjustable 0-5 VDC signal to adjust the voltage output of the accelerator power supply. The total weight of the system including the alpha detector is less than 30 pounds. The neutron generator section of the system of Fig. 2.2 is only 8.75-in.-long and could be separated from the power supply.

2.3 ALPHA DETECTOR

Two concepts are under investigation for the alpha detection; one a gallium doped zinc oxide coating on a thick fiber optic window which prevents spatial distortion of the location of the alpha detection event, and a radially segmented semiconductor detector, which consists of eight rings and a central circle of the same area, that detect alpha particles with radial resolution. Present thinking is that defined cones of neutrons are adequate for most NMIS applications. For the case of the fiber optic window the cone can be defined by use of an iris between the PM tube and the outside of the fiber optic window. For the case of the radially segmented semiconductor detector, the signals will be summed electronically out to a selected radius to define the cone of neutrons. The alpha detector utilization limits the source intensity for this kind of use of a DT generator. If the alpha detector count rate limit is 10^6 cps and is divided into 10 rings which intercepts the alpha emissions from the target, then practical source of intensities would be $\sim 10^8$ n/s which would be a useful source size for large containers. Present experience is that a 5×10^6 n/s source is adequate for 3-ft.-diam containers.

2.4 SYSTEM

The signals from all of the detectors go via shielded cables to a processor. This processor contains the NMIS data acquisition boards and the multichannel analyzer for gamma ray spectrometry and could provide input to a red light-green light attribute display.

The detectors for a passive system for Pu consists of the two large plastic scintillators and the HPGe gamma ray detector. These same components are used in the active system and the passive system could be upgraded to an active system for HEU by addition of the neutron source and small transmission detector with appropriate software changes.

The data obtained by an active system for Pu also contains the data from a passive system since the total signal consists of two parts related to inherent source fission and external neutron source induced fission, which can be separated by correlation with the external source. The active system with HPGe has the capability for HE detection, chemical agents, and in some cases, drugs utilizing active (14.1 MeV Neutron generator) gamma ray spectrometry.

3. MATRIX OF POSSIBLE MEASUREMENTS

NMIS uses passive and active time-dependent coincidence methods for both template matching and attribute estimation. If supplemented by gamma ray spectrometry, additional attributes can be estimated, not only for fissile materials but also for HE, chemical agents, and, in some cases, drugs. The possible attributes estimated are given in Table 3.1. Table 3.1 also gives the method for determination and states whether it is active or passive. The system hardware and software can be limited to extract only certain attributes. Each of these attribute determination methods for plutonium and enriched uranium are discussed further in the next two sections of this report. Some attributes can be estimated in multiple ways. In addition, since there is extensive utilization of NMIS signatures for template matching at the Y-12 National Security Complex, this system could easily be used for template matching.

For a passive system for Pu mass, Pu 240/239 ratio, Pu presence, and Pu metal vs. non-metal, the system would use existing gamma ray spectrometry methods for Pu presence and Pu 240/239 and time-dependent coincidence distributions for Pu mass and Pu metal vs. non-metal. Time dependence coincidence could also be used for Pu initial presence.

For an active system for Pu (presence, mass, 240/239 ratio, and metal vs. non-metal) and HEU (presence, mass, enrichment, and metal vs. non-metal) the system would employ (1) gamma ray spectrometry, (2) NMIS active time-dependent coincidence counting with a DT source, and (3) transmission measurements. All these systems of Figs. 1.1, 1.2, and 1.3 would be used in the system. So, the system would consist of three proton recoil scintillators, an HPGe detector, and the DT generator with the processor. If NMIS time-dependent coincidence measurements also determines Pu presence, Pu 240/239 ratio, and presence of metal by transmission or time-dependent coincidence distribution measurements, then gamma ray spectrometry may not be needed for fissile materials. The red light-green light display of the attributes could indicate mass above threshold, and Pu 239/240 content below 0.1 and HEU enrichment of U > 20% ^{235}U. For HE and chemical agents, the ratios of N, H, and O to C are obtained from gamma ray spectrometry.

Table 3.1. Matrix of Attribute Measurements by NMIS with Gamma Spectrometry

Material	Attribute	Method (Option, Implementation, Basis)			Active or Passive	Measurement Equipment
plutonium	presence	1	time-dependent coincidence	detect internal spontaneous fission	active or passive	neutron source, scintillation detectors, time-correlator
		2	gamma spectrometry	detect Pu spectral lines	passive	high-resolution gamma detector, multi-channel analyzer
	fissile mass	1	time-dependent coincidence	measure induced fission rate	active	neutron source, scintillation detectors, time-correlator
		2	time-dependent coincidence	measure spontaneous fission rate	passive	scintillation detectors, time-correlator
	age	1	gamma spectrometry	measure in- and out-growth of impurities	passive	high-resolution gamma detector, multi-channel analyzer
	metal / non-metal	1	neutron-initiated gamma spectrometry	detect 6129 KeV gamma from 14.1 MeV neutron interactions with O & F	active	neutron source, high-resolution gamma detector, multi-channel analyzer
		2	time-dependent coincidence	measure density from neutron transmission	active	neutron source, scintillation detector, time-correlator
		3	time-dependent coincidence	attenuation of gammas emitted and multiplication depend on density	passive	scintillation detectors, time-correlator
		4	gamma spectrometry	detection of the 2438 keV gamma ray from the (α,n) reaction on oxygen	passive	high-resolution gamma detector, multi-channel analyzer
	geometry	1	time-dependent coincidence	measure axial density gradient from neutron transmission	active	neutron source, scintillation detector, time-correlator
	thickness	1	time-dependent coincidence	time-dependent coincidence depends on thickness and mass	passive	scintillation detectors, time-correlator
	relative ^{240}Pu-content	1	time-dependent coincidence	compare spontaneous and induced fission rates	active	neutron source, scintillation detectors, time-correlator
		2	gamma spectrometry	compare ^{240}Pu and ^{239}Pu spectral lines	passive	high-resolution gamma detector, multi-channel analyzer
		3	time-dependent coincidence	integral and width of distribution primarily related to ^{240}Pu and ^{239} Pu mass, respectively	passive	scintillation detectors, time correlator

Table 3.1. Matrix of Attribute Measurements by NMIS with Gamma Spectrometry (Cont'd.)

Material	Attribute	Method (Option, Implementation, Basis)			Active or Passitve	Measurement Equipment
uranium	**presence**	1	time-dependent coincidence	detect induced fission and absence of internal spontaneous fission	active	neutron source, scintillation detectors, time-correlator
	fissile mass	1	time-dependent coincidence	measure induced fission rate	active	neutron source, scintillation detectors, time-correlator
	^{235}U-enrichment	1	time-dependent coincidence	compare induced fission rates and neutron transmission	active	neutron source, scintillation detectors, time-correlator
	metal / non-metal	1	neutron-initiated gamma spectrometry	detect 6129 KeV gamma from 14.1 MeV neutron interactions with O & F	active	neutron source, high-resolution gamma detector, multi-channel analyzer
		2	time-dependent coincidence	measure density from neutron transmission	active	neutron source, scintillation detectors, time-correlator
		3	time-dependent coincidence	attenuation of gamma emitted and multiplication depend on density	active	neutron source, scintillation detector, time-correlator
	geometry	1	time-dependent coincidence	measure axial density gradient	active	neutron source, scintillation detectors, coincidence gate
high explosive	**presence**	1	gamma spectrometry	N presence and ratios of N/C, H/C, and O/C	active	neutron source, HPGe[a] or BGO[b]
chemical weapon	**presence**	1	gamma spectrometry	ratios of N/C, H/C, and O/C	active	neutron source,HPGe[a] or BGO[b]
druge	**presence**	1	gamma spectrometry	ratios of N/C, H/C, and O/C	active	neutron source, HPGe[a] or BGO[b]

[a]Methods of A. J. Caffrey; see Reference 4.
[b]G. Vourvopolos and P. C. Womble, "Pulsed Fast/Thermal Neutron Analysis: A Technique for Explosive Detection," Applied Physics Institute, Western Kentucky University, Bowling Green, KY 42101 (2001).

4. PU ATTRIBUTES

This section describes in some detail how the various Pu attributes are determined. Gamma ray spectrometry has been used by other laboratories to verify the presence of Pu, Pu 240/239 ratio, and time (age) since reprocessing. These methods are only mentioned here briefly and referenced. Alternative NMIS based methods are described in some cases, and the use of 14.1 MeV neutrons to determine PuO_2, U_3O_8 or UF_4 presence by measuring emitted gamma rays from neutron interactions is discussed in some detail. This chapter is our present thinking on attribute extraction, but improved methods from other features, ratios, products, or other combinations of features from the NMIS data will be developed during the development and testing of this system.

4.1 PU PRESENCE

Several of the subsequent attribute estimation procedures are specific to the material inspected, i.e., plutonium or enriched uranium. A measurement method to recognize that a container bears a plutonium object or objects is desirable.

4.1.1 Gamma Spectrometry Method

Passive gamma ray spectrometry for Pu presence is well known and documented for this application[17] and is not described here.

4.1.2 Time-Dependent Coincidence

This section proposes a second method to detect the presence of plutonium within a container. It is proposed as an alternative to the preceding passive high-resolution gamma spectrometry method.

4.1.2.1 *Physical basis*

Recall that ^{240}Pu spontaneously fissions such that a plutonium-bearing container will exhibit spontaneous emission of time-correlated neutrons and gammas. The intensity of this internal source depends upon the mass of the ^{240}Pu constituent and the self-multiplication of the plutonium contents. However, the total plutonium mass and relative ^{240}Pu-content of a plutonium sample create an easily detectable spontaneous fission source. Therefore, positive identification of an internal source of time-correlated neutrons and gammas in a passive or active measurement indicates that a container bears a plutonium object or objects. The absence of an appreciable internal source indicates the absence of plutonium for this application, where the accepted alternatives are plutonium *or* enriched uranium. The method proposed is designed to detect an appreciable internal source of time-correlated neutrons and gammas from either a passive or active measurement.

[17] D. Reilly, N. Ensslin, H. Smith, Jr., and S. Kreiner, *Passive Nondestructive Assay of Nucler Materials,* U.S. Nuclear Regulatory Commission, Washington, D.C., March 1991.

When a container bearing plutonium is interrogated by an active source of neutrons, it will exhibit both an *induced* response and a *spontaneous* response (Fig. 4.1). Of course, without interrogation by an active source, plutonium will only have a spontaneous component. That is, the induced response is prompted by the presence of the active source, and the spontaneous response is observed even in the absence of the active source. By using an active neutron source and a pair of scintillation detectors, the measurement system shown in Fig. 1.2 can acquire in a single measurement the time-distribution of detector coincidence and partition that distribution into mutually exclusive induced and spontaneous components. In the absence of an internal source, the spontaneous coincidence will vanish. In practice, in the absence of an internal source, spontaneous coincidence will be statistically insignificant compared to induced coincidence. Therefore, if the ratio of spontaneous coincidence to induced coincidence exceeds some threshold, then an internal source of time-correlated neutrons and gammas has been positively identified, and the presence of plutonium has been detected.

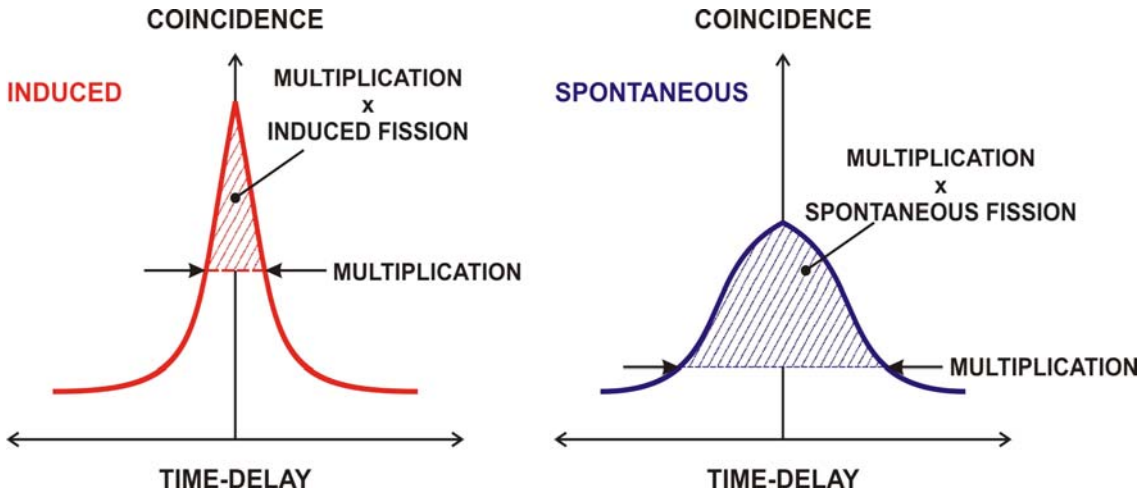


Fig. 4.1. Features of the Measurement Used to Infer the Presence, Relative ^{240}Pu-Content, and Fissile Mass Attributes of Plutonium Samples. (The widths of both the induced and spontaneous time-dependent coincidence-distributions are each proportional to multiplication. The integral of the induced coincidence-distribution is proportional to the product of multiplication and induced fission rate, and the integral of the spontaneous coincidence-distribution is proportional to the product of multiplication and spontaneous fission rate. Note that the spontaneous coincidence distribution vanishes in the absence of plutonium.)

4.1.2.2 *Technical issues*

The minimum threshold to assert positive identification of an internal source can be estimated by performing Monte Carlo calculations[18] over a matrix of relative ^{240}Pu-content (say in steps of 2% from 0 to 16% ^{240}Pu and relevant total plutonium masses. If possible, a selected number of these calculations should be verified via measurements on samples with varying ^{240}Pu-content, total plutonium mass, and geometry.

Note that because the proposed method relies upon the spontaneous response of the inspected container, the measurement to implement this method must be performed isolated from other plutonium-bearing containers. The proximity of other containers to the measurement apparatus should probably exceed 3 meters.

4.2 TIME (AGE) SINCE PROCESSING

The use of gamma ray spectrometry with HPGe for time (age) since chemical reprocessing is well known and documented for this application[19] and is not described here.

4.3 PU GEOMETRY

4.3.1 Physical Basis

The shape of the Pu sphere can be estimated by performing a vertical transmission profile of the fissile materials with the source and the small scintillation detector. Measurement time at each location would be less than one minute. This could be done by scanning or using a vertically segmented detector. For the vertically segmented detector, the whole profile could be obtained simultaneously. The location of edges of the sphere could be measured, from which the diameter of the sphere could be obtained. If pits are stored, both diameter and thickness of material could be obtained. The characteristic vertical profile of a sphere is like a U, while that for hollow shells is like a W, as shown in Fig. 4.3.1. From transmission the thickness can also be obtained knowing the material is Pu metal and performing measurements on an empty container.

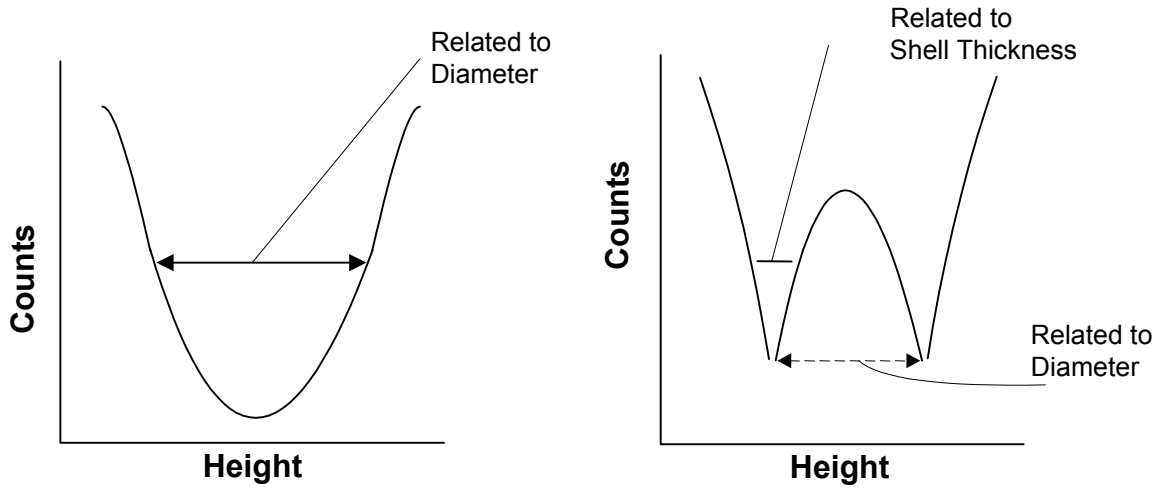
4.3.2 Technical Issues

NMIS has considerable experience with measurements of the shape of Pu on fully assembled systems and individual pits in containers. The diameter of a solid sphere can be estimated from such a scan (Fig. 4.3.1a) and the diameter and thickness of shells (Fig. 4.3.1b) can also

[18] All of the data acquired by the NMIS processor for active or passive measurements can be calculated by Monte Carlo coupled neutron gamma transport methods of T. E. Valentine described in the following reference: T. E. Valentine and J. T. Mihalczo, "Validation of the Monte Carlo Code MCNP-DSP," *Annals of Nuclear Energy* **24**, No. 2, 79-98 (1996).

[19] R. Gunnink, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Pu Isotopic Abundance," UCRL-LR-103220, Vol. 1 (April 1990).

be obtained. Such scans have also been performed for pits in containers at LANL for DSWA (1997), at Pantex (1998), and at VNIIEF (1999) by Russian specialists. Some limited testing of the final systems should be performed and supported by a calculational program.



(a) Vertical Scan for Solid Sphere **(b) Vertical Scan for Hollow Shell**

Fig. 4.3.1. Sketch of Characteristic Transmission Profile From Vertical Scans of Fissile Spheres and Shells

4.4 PU METAL VS. NON-METAL

Absence of Pu metal, described in this section, is detected by four methods: (1) secondary gamma rays from inelastic scattering of 14.1 MeV neutrons by the oxygen in PuO_2, (2) transmission measurements which show a reduced density from Pu metal, (3) changes in the time-dependent coincidence distribution between a pair of detectors, and (4) detection of gamma rays from (α,n) reactions with oxygen.[20] For an active system, the transmission measurement is the preferred method because of simplicity of interpretation. For a passive system, the time-dependent coincidence method could be used.

4.4.1 Active Gamma Spectrometry

4.4.1.1 *Physical basis*

The detection of oxide is possible by observing the 6129 KeV gamma ray from inelastic scattering of 14.1 MeV neutrons by oxygen. This type of measurement has been performed by A. J. Caffrey of the Idaho National Engineering and Environmental Laboratory (INEEL)

[20]D.J. Mercer, A.P. Belian, J.A. Bounds, J.E. Dyson, T.B. Gosnell, W.K. Hensley, P.L Kerr, C.E. Moss, L.F. Nakae, W.K. Pitts, and D.T. Vo, "Discrimination between PuO2 and Pu Metal by Gamma-Ray Spectroscopy", LA-UR-01-4870 (2001).

and others. The results of a 100 sec measurement with a DT neutron generator are shown in Fig. 4.4.1.[21] The sample in this measurement was simulated phosgene ($COCl_2$). In additional measurements in September 2001, A. J. Caffrey[21] has used also this method to detect the presence of oxygen in a ~430 g sample of iron oxide.

The gamma rays from inelastic scattering from nearby materials must be subtracted from the acquired data. The contribution of the oxygen from any of the surroundings can be minimized by gating on the MCA with the pulse from the alpha detector that defines only the cone of neutrons that hit the PuO_2 sample. This gating minimizes the background but does not eliminate it completely. A measurement without Pu metal can determine the background of 6129 KeV gamma rays from inelastic scattering by the oxygen of the container material and its surroundings.

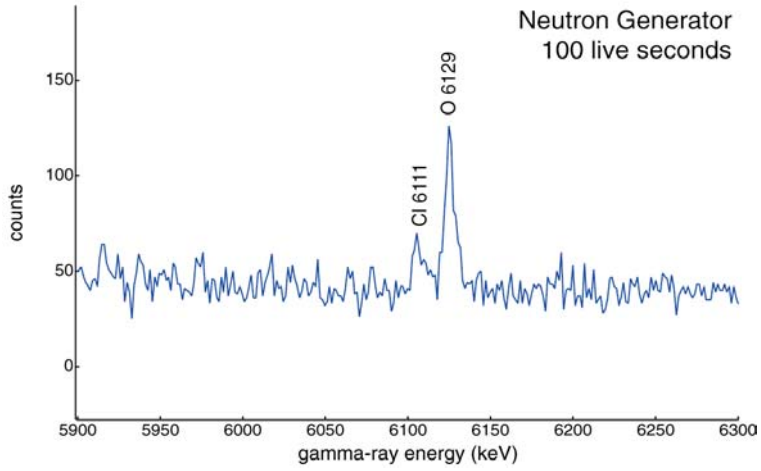


Fig. 4.4.1. Gamma Ray Spectrometry for 14.1 MeV Neutrons on Oxygen

4.4.1.2 *Technical issues*

Monte Carlo calculation of the expected measurement results should be performed prior to testing. Although the method has been tested for iron oxide out of a container, this method also needs testing in containers. This might be done initially with some metal oxide such as tungsten or iron oxide. These tests would establish the adequacy of the background subtraction method. The method would be tested in the final system with fissile metal and oxide.

4.4.2 Source Neutron Transmission

4.4.2.1 *Physical basis*

The density difference between Pu metal and non-metal (PuO_2) is such that active transmission measurements through the Pu sample will determine the presence of non-metal in conjunction with the vertical scan which determines the dimensions. This measurement is

[21] A. J. Caffrey, INEEL, personal communication (August 2000).

independent of both uncorrelated and time correlated background from nearby fissile material, and is perhaps the simplest method for metal presence. Most fissile compounds have significantly different densities from metal. There is much NMIS experience with transmission measurements.

4.4.2.2 *Technical issues*

This method needs a calculational program for confirmation for the source container configuration for a limited number of compounds and final tests with the fully developed system.

4.4.3 Time-Dependent Coincidence

4.4.3.1 *Physical basis*

A typical time-dependent coincidence function from a passive NMIS measurement at VNIIEF with 1.77 wt% ^{240}Pu is shown in Fig. 4.4.3 for two 4 kg masses of Pu: one with an inner radius of 1.0 cm and outer radius of 4.02 cm, and the other with an inner radius of 5.35 cm and an outer radius of 6.0 cm. One configuration is much thicker (nearly spherical) than the other (shell). The shell effectively has a low average density. For the shell, the attenuation out is much less so the gamma-gamma coincidence at $\tau = 0$ is larger. The width of the distribution is less because of lower multiplication. These differences are increased in active measurements.

Thus, the shape of the time-dependent coincidence for non-metallic components of Pu would be different from metal. Because gamma rays emerge more easily from the non-metallic material, the gamma-gamma coincidence rate would be higher than for metal. This effect is larger as density is reduced. Of course, this would be offset by lower multiplication, which reduces the source of coincidences and width of the time distribution.

4.4.3.2 *Technical Issues*

There is little NMIS experience with other fissile compounds. This method needs a calculational program for quantification of the effect, interim tests with a plutonium oxide and uranium oxide, and final tests with the fully developed system.

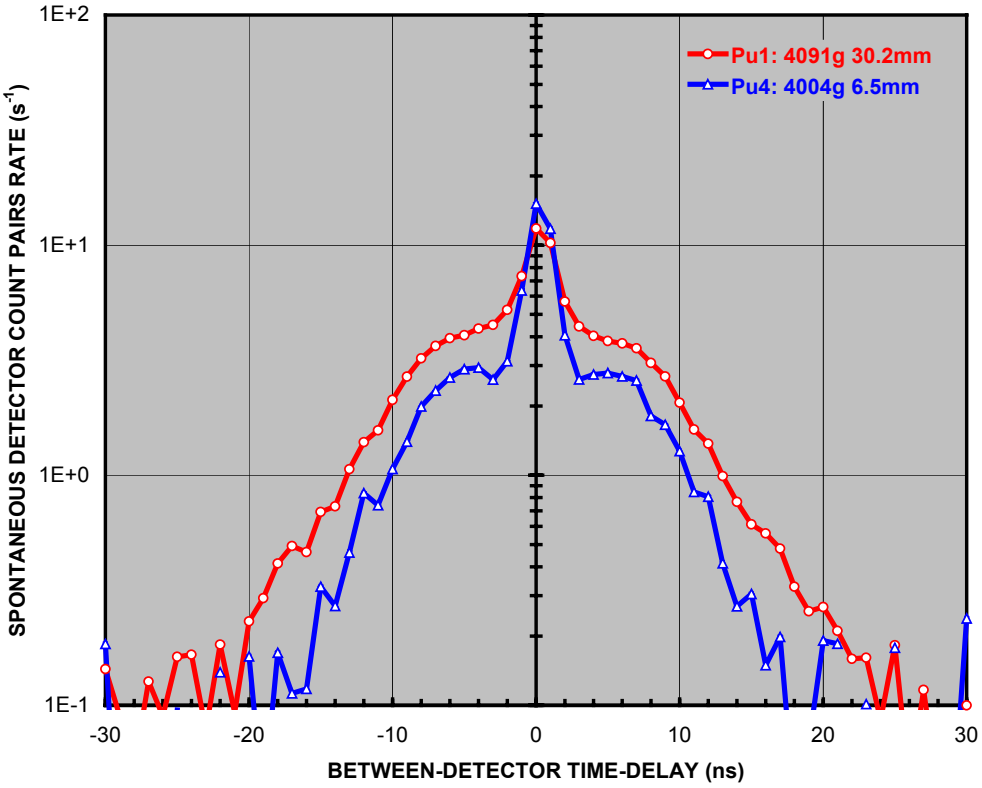


Fig. 4.4.7. Time-Dependent Coincidence Distribution Between Detectors in a Passive Measurement at VNIIEF With 1.77 wt% ^{240}Pu Shells of 3.02 and 0.65 cm Thickness and the Same Mass

4.5 PU 240/239 RATIO

This ratio has been usually obtained by gamma ray spectrometry or mass spectrographic analysis. For this application, others have recommended gamma ray spectrometry. However, there is an alternate time-dependent coincidence distribution method that may not be as accurate, but may be adequate for this application.

4.5.1 Gamma Spectrometry Method

Passive gamma ray spectrometry with HPGe for measuring the Pu 240/239 ratio is well known and documented for this application[22] and is not described here.

[22] H. Umezawa, T. Suzuki, and S. Ichikawa, "Gamma-Ray Spectrometric Determination of Isotopic Ratios of Plutonium, *Journal of Nuclear Science and Technology,* 13, 327-332 (1976).

4.5.2 Time-Dependent Coincidence

The subsequent development proposes a second method to quantitatively estimate the relative ^{240}Pu-content of a sample within a container. This method is proposed as an alternative to the preceding passive high-resolution gamma spectrometry method.

4.5.2.1 *Physical basis*

The physical basis for the method to quantitatively estimate the relative ^{240}Pu-content proceeds from that proposed to detect the presence of plutonium. The ^{240}Pu constituent is a spontaneous source of time-dependent coincidence neutrons and gammas. The ^{239}Pu constituent can be induced to fission by an active neutron source thereby creating an induced source of time-dependent coincidence neutrons and gammas. The relative intensity of these two sources depends upon the total plutonium mass and self-multiplication of the sample as well as its relative ^{240}Pu-content. The ratio of the spontaneous and induced sources, when adjusted for the effects of total mass and multiplication, is proportional to the ^{240}Pu / ^{239}Pu mass ratio.

By using an active neutron source and a pair of scintillation detectors, the measurement system shown in Fig. 1.3 can acquire in a single measurement the time-distribution of detector coincidence and partition that distribution into mutually exclusive induced and spontaneous components. Now refer to Fig. 4.1. The width (measured as, e.g., the full-width at tenth-maximum, or FWTM) of both the induced and spontaneous coincidence distributions is proportional to the sample's self-multiplication, though the dependence of the induced and spontaneous components' widths upon multiplication differs slightly due to the different neutron spectrum and spatial distribution of the initiating sources (the induced source is initiated by an external DT generator, while the spontaneous source is initiated by the internally distributed ^{240}Pu constituent). The integrals (measured as, e.g., the area beneath the respective FWTMs) of the induced and spontaneous components are respectively proportional to the induced and spontaneous fission rates, each augmented (e.g., multiplied) by multiplication. Consequently, the induced and spontaneous integrals are respectively proportional to the masses of the ^{239}Pu constituent and the ^{240}Pu constituent. When the integral of each component is adjusted for the effect of multiplication, the ratio of the spontaneous component to the induced component yields an estimate of the ^{240}Pu / ^{239}Pu mass ratio. This estimate can subsequently be compared to a pre-determined threshold corresponding to the criterion for identifying weapons-grade plutonium.

Alternately, in a passive measurement the total coincident pairs between detectors are primarily related to the ^{240}Pu mass; whereas, the width of the time distribution of coincidences (at 1/10 maximum value) is primarily related to the ^{239}Pu mass through self multiplication. Thus, this type of analysis will yield the ratio of isotopes (240/239) from a passive measurement.

4.5.2.2 *Technical issues*

The functional form relating the widths and integrals of the spontaneous and induced coincidence distributions to relative ^{240}Pu-content, total plutonium mass, and multiplication can be established by performing Monte Carlo calculations over a matrix of ^{240}Pu-content and total plutonium mass spanning the expected values. The results of these calculations will be used to parameterize prototypical trends of the preceding spontaneous-to-induced coincidence ratio versus total plutonium mass and multiplication. If possible, a selected number of measurements on samples with varying ^{240}Pu-content and total plutonium mass should be performed to verify the trends and parameters estimated from the Monte Carlo calculations.

Note that because the proposed method relies upon the spontaneous response of the inspected container, the measurement to implement this method must be performed isolated from other plutonium-bearing containers. The proximity of other containers to the measurement apparatus should probably exceed 3 meters. Furthermore, because multiplication is strongly dependent upon composition and geometry, the proposed method will be developed to address only plutonium-metal sample configurations of interest. Finally, it is recommended that prior to deployment this method be subjected to a reasonable number of blind tests to validate its implementation.

4.6 PU FISSILE MASS

The subsequent development proposes two alternative methods to quantitatively estimate the fissile (i.e., ^{239}Pu) mass of a plutonium sample within a container. The first proposed method can be implemented in a passive measurement. The second proposed method requires an active neutron source for implementation.

4.6.1 Passive Method

4.6.1.1 *Physical basis*

The physical basis for the method to passively estimate the fissile mass of plutonium samples is derived from that to estimate the relative ^{240}Pu-content. The ^{240}Pu constituent is a spontaneous source of time-correlated neutrons and gammas; the intensity of this source principally depends upon the mass of ^{240}Pu and the self-multiplication of the sample. When corrected for the effect of multiplication, the intensity of this spontaneous source is proportional to the total mass of ^{240}Pu present in the sample. With an independent passive measure of the relative ^{240}Pu-content, as might be obtained from passive gamma spectrometry, the ^{239}Pu mass, and hence the fissile mass, can be determined from the estimated ^{240}Pu mass.

For passive measurements, the time-distribution of detector coincidence provides a measure of the spontaneous ^{240}Pu source intensity as illustrated in Fig. 4.1. In particular, the width (measured as, e.g., the full-width at tenth-maximum, or FWTM) of this distribution is

proportional to multiplication. The distribution's integral (measured as, e.g., the area beneath the FWTM) is proportional to the spontaneous fission rate and multiplication (taken as, e.g., a product) and hence is proportional to ^{240}Pu mass. Consequently, the ^{240}Pu mass may be estimated from the time-distribution of detector coincidence after adjusting for multiplication[12,23,24] Given the relative ^{240}Pu-content from a supporting measurement of the 240/239 ratio, the ^{239}Pu mass can then be estimated using this passive method. The estimate of ^{239}Pu mass can then be compared to a pre-determined threshold corresponding to the criterion for a weapons-quantity of plutonium.

Some successful attribute analysis of the NMIS data from VNIIEF measurements in 2000 is given in Appendices A and B, and some results of Monte Carlo calculations for varying Pu mass at Mayak are given in Appendix C.

4.6.1.2 *Technical issues*

The functional form relating the spontaneous time-dependent coincidence distribution to relative ^{240}Pu-content, total plutonium mass, and multiplication can be established by performing Monte Carlo calculations over a matrix of ^{240}Pu-content and total plutonium mass expected. The results of these calculations will be used to parameterize prototypical trends of the coincidence width and integral versus total relative ^{240}Pu-content and multiplication. Note that multiplication is only weakly affected by the relative ^{240}Pu-content over the range of interest. Consequently, these trends will principally relate multiplication to the spontaneous coincidence distribution and will be relatively invariant with relative ^{240}Pu-content. If possible, a selected number of measurements on samples with varying ^{240}Pu-content and total plutonium mass should be performed to verify the trends and parameters estimated from the Monte Carlo calculations.

Note that because the proposed method relies upon the spontaneous response of the inspected container and a supporting estimate of the relative ^{240}Pu-content, the measurement to implement this method must be performed isolated from other plutonium-bearing containers. The proximity of other containers to the measurement apparatus should probably exceed 3 meters. Furthermore, because multiplication is strongly dependent upon composition and geometry, the proposed method will be developed to address plutonium-metal shape. Finally, it is recommended that prior to use, this method be subjected to a reasonable number of blind tests to validate its implementation.

4.6.2 Active Method

[23] T. E. Valentine, L. G. Chiang, and J. T. Mihalczo, "Monte Carlo Evaluation of Passive NMIS for Assay of Plutonium in Shielded Containers," Institute of Nuclear Materials Management, New Orleans, Louisiana, July 16-20, 2000.

[24] Previous studies have shown the passive method to be viable; see reference 19 and Appendix B of this report describing Monte Carlo simulations of Mayak plutonium spheres up to 2 kg each (see Appendix B). Both the passive and active methods have been previously demonstrated using plutonium spherical shells measured at the Russian Federal Nuclear Center, All-Russia Research Institute of Experimental Physics described in Ref. 14.

4.6.2.1 *Physical basis*

Unlike the preceding passive method to estimate plutonium fissile mass, the subsequent proposed method, which requires an active measurement, can be implemented without the support of passive gamma spectrometry instruments. Its physical basis is derived from that for estimating the relative ^{240}Pu-content. The ^{239}Pu constituent can be induced to fission by an active neutron source thereby creating an induced source of time-correlated neutrons and gammas. The intensity of this induced source is principally dependent upon the mass of ^{239}Pu and the self-multiplication of the sample. By measuring the intensity of the induced source and adjusting for self-multiplication, the ^{239}Pu mass, and hence the fissile mass, can be quantitatively estimated.

By using an active neutron source and a pair of scintillation detectors, the measurement system shown in Fig. 1.3 can acquire in a single measurement the time-distribution of detector coincidence and extract from that distribution an induced component arising due to fission-chains initiated by the active source. As depicted in Fig. 1.4 the width of this induced coincidence distribution (measured as, e.g., the full-width at tenth-maximum, or FWTM) is proportional to the sample's self-multiplication. The integral of the induced coincidence distribution (measured as, e.g., the area beneath the FWTM) is proportional to the induced fission rate and multiplication (taken as, e.g., a product) and therefore is proportional to the mass of the ^{239}Pu constituent. When the integral is adjusted for the effect of multiplication using an estimate obtained from the width, a quantitative estimate of the ^{239}Pu mass is obtained. The estimate of ^{239}Pu mass can then be compared to a pre-determined threshold corresponding to the criterion for a weapons-quantity of plutonium.

4.6.2.2 *Technical issues*

The functional form relating the induced time-dependent coincidence distribution to relative ^{240}Pu-content, total plutonium mass, and multiplication can be established by performing Monte Carlo calculations over a matrix of relative ^{240}Pu-content and total plutonium mass. The results of these calculations will be used to parameterize prototypical trends of the preceding induced coincidence width and integral versus relative ^{240}Pu-content and multiplication. Note that multiplication is only weakly affected by the relative ^{240}Pu-content over the range of interest. Consequently, these trends will principally relate multiplication to the induced coincidence distribution and will be relatively invariant with relative ^{240}Pu-content. If possible, a selected number of measurements on samples with varying ^{240}Pu-content and total plutonium mass should be performed to verify the trends and parameters estimated from the Monte Carlo calculations.

Finally, it is recommended that prior to use this method be subjected to a reasonable number of blind tests to validate its implementation.

5. URANIUM ATTRIBUTES (ACTIVE)

5.1 ENRICHED URANIUM PRESENCE

5.1.1 Time-Dependent Coincidence

5.1.1.1 *Physical basis*

The presence of induced fission and the absence of inherent source fission are indicative of the presence of enriched uranium.

5.1.1.2 *Technical issues*

NMIS has experience with the detection of induced fission in HEU. A calculational program of the material-container-source-detector configuration and final tests of the system for this application are needed.

5.2 GEOMETRY

5.2.1 Physical Basis

The shape of the uranium sphere can be estimated by performing a vertical transmission profile of the fissile materials with the source and the small detector. Measurement time at each location would be less than one minute. This could be done by scanning or by a vertically segmented detector. For the vertically segmented detector, the whole profile could be obtained simultaneously. The location of the edges of the sphere could be measured from which the diameter of the sphere could be obtained. There is a wide experience with NMIS in obtaining the shape of weapons components from active scanning measurements both on weapons systems and individual weapons components. This measurement is independent of both uncorrelated and time correlated background from nearby fissile material. The thickness of the fissile material could be obtained assuming metal and performing measurements with an empty container. Some unclassified work on obtaining thickness of materials has been performed by active NMIS measurements.[25]

5.2.2 Technical Issues

This method needs a calculational program for confirmation for the source container configuration and final tests with the fully developed system.

[25] M. S. Wyatt, T. Uckan, J. T. Mihalczo, and T. E. Valentine, "Characterization of an Enriched Uranyl Fluoride Deposit in a Valve and Pipe Intersection Using Time-of-Flight Transmission Measurements with ^{252}Cf," Institute of Nuclear Materials Management Annual Conference, Naples, Florida, July 26-30, 1998.

5.3 U METAL VS. NON-METAL[26]

Absence of U metal, described in this section, is detected by three methods: (1) detection of the 6129 KeV gamma ray from 14.1 MeV source neutrons interacting with oxygen (U_3O_8) and fluorine (UF_4), (2) transmission measurements which show a reduced density from U metal, and (3) changes in the time-dependent coincidence distribution between a pair of detectors. The transmission method may have the simplest analysis. These are essentially the same as for Pu metal vs. non-metal but are repeated here for Section 5 completeness.

5.3.1 Active Gamma Spectrometry

5.3.1.1 *Physical basis*

The detection of oxide is possible by observing the 6129 KeV gamma ray from inelastic scattering of 14.1 MeV neutrons on oxygen. This type of measurement has been performed by A. J. Caffrey of INEEL and the results of a 100 sec measurement are shown in Fig. 5.3.1.[21] The sample in this measurement was simulated phosgene ($COC1_2$).

The gamma rays from inelastic scattering from oxygen in the background must be subtracted from the measured. The contribution of the background oxygen can be minimized by gating on the HPGe MCA with the pulse from the alpha detector that defines only the cone of neutrons that hit the U sample. This gating minimizes the background but does not eliminate it completely. A measurement with a container without fissile material or with U metal can determine the background of 6129 KeV gamma rays from inelastic scattering by the oxygen of the container and its surroundings.

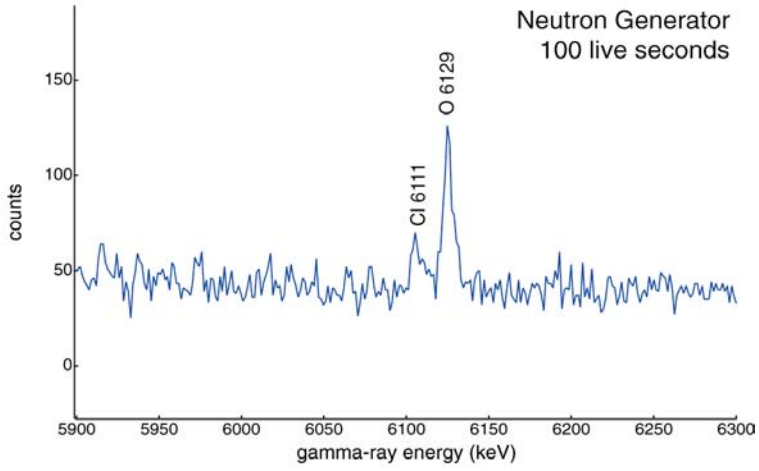


Fig. 5.3.1. Gamma Ray Spectra for 14.1 MeV Neutrons on Oxygen

[26] Much of this is the same as Section 4.4 but is stated here again for HEU to make this section complete.

The other compounds of uranium that are commonly stored are UF_4 and U_3O_8. A. J. Caffrey has also performed measurements of gamma rays from interaction of 14.1 MeV neutrons with fluorine.[21] In this case, the sample was Teflon. Here of course, the (n, alpha) reaction prevails generating the same 6129 KeV oxygen gamma ray. As a result, for interaction of 14.1 MeV neutrons with non-metallic uranium U_3O_8 or UF_4, the detection of this gamma ray would mean the absence of metal and the presence of either U_3O_8 or UF_4 without distinction. Alpha particle gating to minimize background and background subtraction is also necessary for UF_4.

5.3.1.2 *Technical issues*

Monte Carlo calculation of the expected measurement results should be performed prior to testing. Although the method has been tested for iron oxide out of a container, this method also needs testing in containers. This might be done initially with some metal oxide such as tungsten or iron oxide. These tests would establish the adequacy of the background subtraction method. The method would be tested in the final system with fissile metal and oxide.

5.3.2 Transmission

5.3.2.1 *Physical basis*

The density difference between U metal and non-metal compounds (U_3O_8 and UF_4) are such that neutron transmission measurements through the HEU sample will determine the presence of non-metal in conjunction with a vertical scan which determines the dimensions. This measurement is independent of both uncorrelated and time correlated background from nearby fissile material. Increased transmission by other uranium compounds can also be detected.

5.3.2.2 *Technical issues*

It needs a calculational program for confirmation for the source container configuration and final tests with the fully developed system.

5.3.3 Time-Dependent Coincidence

5.3.3.1 *Physical basis*

A typical time-dependent coincidence between detectors from a passive NMIS measurement is shown in Fig. A.2. Similar time-dependent coincidence between detectors in an active measurement result from induced fission. The shape of the time -dependent coincidence function for non-metallic components of U would be different from metal. Since gamma rays more easily come out of the material, the gamma-gamma coincidence rate would be higher than for metal. This effect is larger as density is reduced. Of course, this is offset by lower multiplication which reduces the source of coincidences, but the ratio of gamma-gamma to gamma-neutron coincidence may change. As a result of these various effects, the

shape of the time-dependent coincidence distribution between detectors will change. This effect was shown in Fig. 4.4.7 for Pu.

5.3.3.2 *Technical issues*

NMIS has little experience with other fissile compounds. This method needs a calculational program for quantification of the effect, interim tests with a limited number of materials and final tests with the fully developed system.

5.4 ^{235}U-ENRICHMENT

The subsequent development proposes a method to estimate the ^{235}U-enrichment of a uranium sample in a container. This has already been used successfully (± 1.5% relative uncertainty both on mass and enrichment) for HEU at the Oak Ridge Y-12 Plant. At this time, no viable alternative method has been identified for optically thick samples, e.g., solid uranium-metal spheres.

5.4.1 Physical Basis

The ^{235}U constituent of a uranium sample can readily be induced to fission by an active neutron source (the ^{238}U constituent can also be induced to fission, but the probability of induced ^{238}U fission is lower than that of induced ^{235}U fission for typical neutron energies). In contrast, the total (238 + 235) uranium tends mainly to attenuate the transmission of active source neutrons through inelastic scattering and absorption. Consequently, by measuring the rate of induced fission and the rate of direct source-neutron transmission, the ^{235}U-enrichment can be estimated from the ratio of these quantities.

The measurement system shown in Fig. 1.3 is capable of acquiring the time-distribution of detector counts arising from reactions initiated by active source neutrons and the time-distribution of detector coincidence as illustrated by Fig. 5.4.1. For uranium-bearing containers, the spontaneous portion of the time-dependent coincidence distribution essentially vanishes. Recognize that if a DT generator (or functionally equivalent source of monoenergetic neutrons) is used as the active source, the detection rate of transmitted, i.e., uncollided source-neutrons is readily measured from the count distribution at a time-delay corresponding to the direct flight time of 14.1 MeV neutrons. Consequently, the attenuation of source neutrons, which is proportional to the bulk uranium mass, can be measured. Further consider that the integral of the coincidence distribution (measured as, e.g., the area beneath the full-width at tenth-maximum, or FWTM) is proportional to induced fission rate and multiplication (taken as, e.g., a product), and hence is proportional to the fissile (i.e., ^{235}U) mass. The width of the coincidence distribution (measured as, e.g., the FWTM) is proportional to multiplication alone. Consequently, the ratio of the coincidence integral, adjusted for multiplication, to the attenuation of source-neutrons is proportional to the ^{235}U-

enrichment.[27] The resulting estimate of ^{235}U-enrichment may then be compared to a pre-determined threshold corresponding to the criterion for identifying high-enriched uranium (HEU).

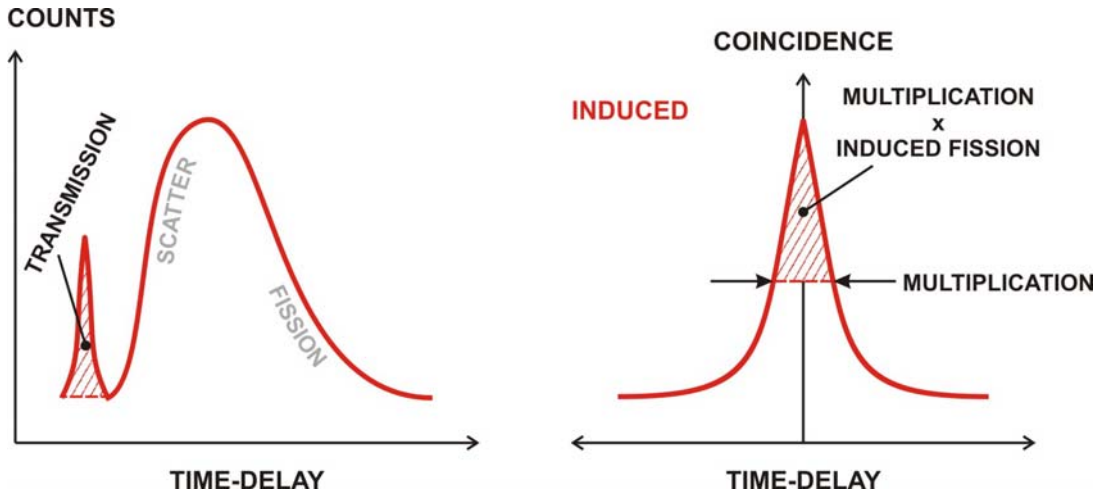


Fig. 5.4.1. Features of the Measurement Illustrated in Fig. 1.3 Used to Infer the ^{235}U-Enrichment and Fissile Mass Attributes of Uranium Samples. [The integral of the first peak in the count-distribution is proportional to the neutron transmission rate and thus total uranium. The width of the coincidence-distribution is proportional to multiplication and mass of ^{235}U, and its integral is proportional to the product of multiplication and induced fission.]

5.4.2 Technical Issues

The functional forms relating the source-neutron transmission rate and the width and integral of the time-dependent coincidence distribution to ^{235}U-enrichment, ^{235}U mass, and multiplication can be established by performing Monte Carlo calculations over a matrix of ^{235}U-enrichment and total uranium mass spanning those expected. The results of these calculations will be used to parameterize prototypical trends of the transmission rate and time-dependent coincidence distribution width and integral versus ^{235}U mass and multiplication. If possible, a selected number of measurements on samples with varying ^{235}U-enrichment and total uranium mass should be performed to verify the trends and parameters estimated from the Monte Carlo calculations.

Observe that because multiplication is strongly dependent upon composition and geometry, the proposed method will be developed to address these dependencies. Finally, it is recommended that prior to use this method be subjected to a reasonable number of blind tests to validate its implementation.

[27] A variation of this method has been previously implemented to verify both ^{235}U-enrichment and uranium fissile mass to within ± 1.5% relative error for a large number of HEU parts at the Y-12 Complex and maybe the first determination of enrichment by methods other than by gamma ray or mass spectrometry; Ref. 10.

5.5 URANIUM FISSILE MASS

The subsequent development proposes a method to quantitatively estimate the fissile mass (i.e., ^{235}U mass) of a uranium sample within a container.

5.5.1 Physical Basis

The ^{235}U constituent of a uranium sample can readily be induced to fission and multiply by an active neutron source (for typical neutron energies, induced and multiplied fission of the ^{238}U constituent is lower than that of the ^{235}U constituent). Consequently, by measuring the rate of induced fission, the ^{235}U mass, and hence the fissile mass, can be estimated.

The measurement system shown in Fig. 1.2 is capable of acquiring the time-distribution of detector coincidence. Note that the spontaneous component of the coincidence distribution vanishes for uranium-bearing containers. Now refer to Fig. 5.4.1. Consider that the integral of the coincidence distribution (measured as, e.g., the area beneath the full-width at tenth-maximum, or FWTM) is proportional to the induced fission rate and multiplication (taken as, e.g., a product), and is therefore proportional to the fissile (i.e., ^{235}U) mass; the width of the coincidence distribution (measured as, e.g., the FWTM) is proportional to multiplication alone. Consequently, the coincidence integral, when adjusted for multiplication using the coincidence width, is proportional to the ^{235}U mass.[27] The resulting estimate of fissile mass may then be compared to a pre-determined threshold corresponding to the criterion for identifying a weapons-quantity of ^{235}U. From transmission the thickness can be obtained knowing the material.

5.5.2 Technical Issues

The functional forms relating the width and integral of the coincidence distribution to ^{235}U-enrichment, ^{235}U mass, and multiplication can be established by performing Monte Carlo calculations over a matrix of ^{235}U-enrichment and total uranium mass spanning the expected values. The results of these calculations will be used to parameterize prototypical trends of the coincidence width and integral versus ^{235}U-enrichment and multiplication. If possible, a selected number of measurements on samples with varying ^{235}U-enrichment and total uranium mass should be performed to verify the trends and parameters estimated from the Monte Carlo calculations.

Observe that because multiplication is strongly dependent upon composition and geometry, the proposed method will be developed to address only uranium-metal spherical samples as is currently expected to be the nominal Mayak uranium sample configuration. Hence, the validity of the results obtained using this method rest upon the a-priori verification of uranium-metal composition and spherical geometry by the means described in preceding developments. Finally, it is recommended that prior to deployment this method be subjected to a reasonable number of blind tests to validate its implementation.

6. FISSILE MATERIAL TEMPLATES

Two different approaches have been put forward for the analysis of radiation signature data for monitoring of storage of special nuclear materials. These are referred to as the template matching and the attribute approach. The previous two sections of this report have addressed attribute determination methods. Attributes have the advantage of not needing to store classified signatures. A template is a measured characteristic radiation signature (such as a time-dependent coincidence distribution function) obtained from measurements of known items of the same type; it may be an average of such measurements. The template matching method is a procedure to identify an unknown item by comparison with an established template. A match is the result of a comparison in which the unknown object signature and the template are the same within an agreed variability.

Template matching with NMIS signatures has been in use at the Y-12 National Security Complex for confirmation of weapons components for stored items since 1996 and receipts since January 2000. These template matching confirmations are for internal NMC&A and confirmation of receipts. Template matching for NMIS has been described by J. A. Mullens[9] and template matching has been described in general by Lemley[28] and others.

The problem of storage of the classified templates can be circumvented by controlled, monitored storage of the classified item itself (first suggested to us by V. Dubinin of VNIIEF). The template can be reacquired daily as needed. If they arrive in campaigns, it still could be manageable. Between use of the reference containers for template acquisition, their storage could be monitored using tags and seals along with continuous video surveillance of the containers.

[28] James R. Lemley, Peter E. Vanier, and L. Forman, "Template Applications for Monitoring Warhead Dismantlement," Institute of Nuclear Materials Management, Indian Wells, CA, July 15-19, 2001.

7. HE AND CHEMICAL AGENTS

The activation analysis methods with HPGe for HE and chemical agent detectors are well known (Ref. 27) and have been implemented by other laboratories. The distinction of HE from chemical agents using ratios N/C, H/C, and O/C are illustrated in Fig. 7.1 (courtesy of J. Morgan of LLNL).

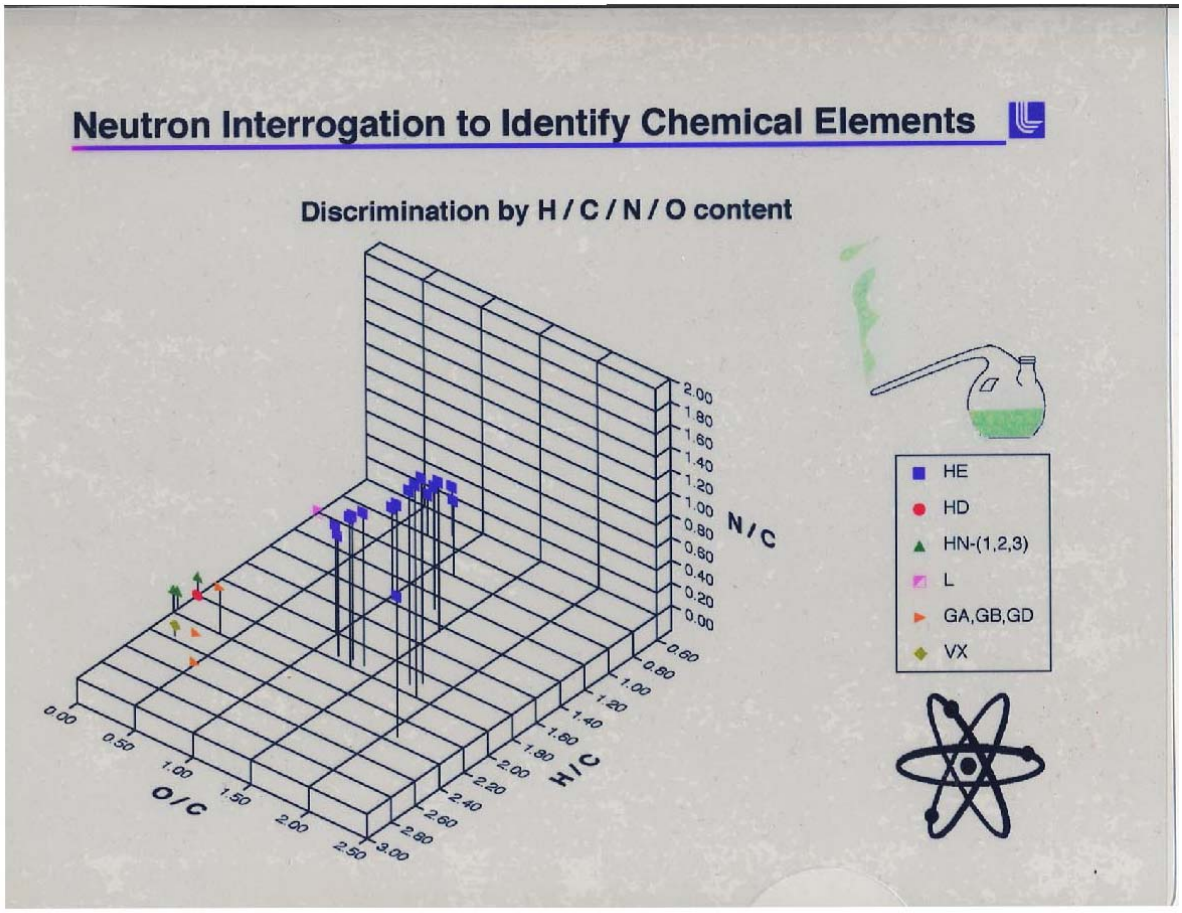


Fig. 7.1. Ratios From Gamma Ray Spectrometry for Presence of HE and Chemical Agents

8. ADDITIONAL CONSIDERATIONS

8.1 ATTRIBUTE METHODS DEVELOPMENT

This development requires measurements for two ^{240}Pu contents and two ^{235}U enriched measurements. In addition to the measurements for the attribute development, Monte Carlo neutron transport theory calculations are essential to the development of these attribute determination methods. Monte Carlo codes are capable of calculating all of the NMIS signatures for both passive and active measurements (see Appendix B for passive measurement calculations for the Mayak FMSF). MCNP-DSP can and has been used for these analyzes. A similar code, MCNP-POLIMI by Marseguerra, Padovani, and Pozzi[29] for this type of calculation which more properly follows the physics of the gamma ray production reactions.

In general, the attribute development (for example for HEU) proceeds as follows: (1) various NMIS signatures for a wide variety of ^{235}U enrichments, masses, and configurations that span the range of variables of interest are calculated to determine the dependence of the various attributes on the features or combinations of features (e.g. ratios) of the NMIS signatures. At this point, particular attributes are correlated to features or combinations of NMIS signatures to find out which are most sensitive and useful for particular attribute extractions. (2) NMIS signatures are measured to ascertain the bias in the calculations. This ties the calculations to reality by these measurements and provides the final relationships that relate features with attributes. Finally, (3) these attribute extraction methods are tested with measurements with unknowns [items not in step (2) but encompassed by step (1) and then refinements are made as necessary].

Approximately 100 days should be allowed for measurements with the four types of materials (i.e. two ^{240}Pu contents and two ^{235U} enrichment). For Pu half of the time would be for passive measurements and half would be for active measurements. The passive measurements could start after the procurement of the 500 × 500 × 80-mm-thick plastic scintillation detectors. All measurement programs include a supporting calculational program. The following sections address present thinking as to how this could be done but improvements in the attribute system extraction method will occur as the program proceeds.

8.1.1 Passive Measurements for Pu

In the passive system, time-dependent coincidence distribution would be used to obtain Pu mass and Pu metal vs. non-metal. Gamma ray spectrometry would be used for Pu presence and Pu 240/239 ratios. Time-dependent coincidence could also be used for Pu presence.

8.1.1.1 *Pu mass*

[29] M. Marseguerra, E. Padovani, and S. Pozzi, "The Use of MCNP-POLIMI Code For Time Correlation Safeguards Measurements," Smorn Symposium on Nuclear Reactors Surveillance (May 27-31, 2002).

The methods of Appendix A have been developed for analysis of passive NMIS measurements for Pu mass and show how the method works. This work is further described in Ref. 13 and the VNIIEF work in Ref. 14 (Appendix B). This work was for measurements with Pu not in containers and using small detectors 150 × 150 × 100-mm-thick plastic scintillators but does illustrate how the attribute estimation for mass would be performed. These previous measurements at VNIIEF have shown that the count rate from 150 × 150 × 100-mm thick detectors are not adequate and that 500 × 500 × 100-mm-thick detectors are needed for practical measurements in AT400 containers. The MCNP-DSP calculations of Appendix C for Pu in AT400 containers shows that NMIS can be used to obtain the mass of Pu in AT400 containers in short measurement times with 500 × 500 × 100-mm-thick detectors.

The path forward to obtain the Pu mass attribute from passive measurements involves three steps assuming that the Pu 240/239 ratio is known from gamma ray spectroscopy. (1) Calculate the NMIS signature for measurements with 500 × 500 × 80-mm thick proton recoil scintillators for Pu in an AT400 container over a variety of Pu 240/239 ratios (240 contents varying from 0 to 16 in 2% steps and masses varied between 500g and 6000g). (2) Perform selected measurements over this same range of variables to obtain the calculational bias and to establish the relationship between features of the data and attributes, and finally (3) test the methods by measurements.

8.1.1.2 *Passive measurements for Pu metal vs. non-metal detection*

The shape of the time-dependent coincidence distribution between Pu metal and less dense compounds depends on density as shown in Fig. 4.4.7. Two effects occur when the density is reduced and are shown in this figure. The gamma-gamma coincidence rate from fission is increased since the gamma rays more easily escape the material. The neutron multiplication decrease with reduced density and the width of the distribution decreases since fission chain decay is faster. To demonstrate this effect 2 to 4 kg Pu samples of both metal and PuO_2 should be used.

8.1.1.3 *Pu presence*

Gamma ray spectrometry methods for this exist and will be incorporated without testing into the system for final testing.

Time-dependent coincidence measurements can also determine Pu presence and this capability can be tested in measurements with Pu and other spontaneous fission sources. The time-dependent coincidence between detectors will be different for Pu because of multiplication.

8.1.1.4 *Pu 240/239 ratio*

Gamma ray spectrometry methods exist and will be incorporated into the system for final testing.

8.1.2 Active System

An active system is capable of measuring the additional attributes required for HEU as well as some additional attributes for Pu and may provide a way to obtain the Pu 240/239 ratio without gamma ray spectrometry. Recall that by correlating detector responses with the external source, all the time-dependent coincidences measured in a passive Pu measurement can also be obtained from an active measurement.

8.1.2.1 *Pu or U metal/non-metal*

Three types of measurements will be performed to address the determination of Pu metal/non-metal or U metal/non-metal. (1) Transmission time-of-flight measurements through the sample depend on density. Active neutron transmission measurements both out and in AT400 containers can be performed with ~2 kg metal spheres of Pu and PuO_2 containing ~2kg of Pu. Measurements will also be performed with ~8 kg spheres of U and a U_3O_8 sample with ~8 kg of U and with an empty container. (2) For the metal and oxide samples of the previous paragraphs, gamma rays emitted by the interaction of 14.1 MeV neutrons with the sample will be used to identify the presence of 6129 keV gamma rays from inelastic scattering on oxygen. (3) Changes in the time-dependent coincidence distribution between oxide and metal will also be measured for U and Pu. The three methods will be intercompared and the best method selected for incorporation into the system.

8.1.2.2 *Geometry*

Preliminary scanning measurements are not needed for this attribute based on previous experience with Pu and uranium metals. When scanning is included in the final active system, then final system tests can be performed.

8.1.2.3 *Pu 240/239 ratio*

Measurements to obtain the Pu 240/239 ratio from time-dependent coincidence distribution between two detectors should be performed. They will be performed outside of containers as well as inside AT400 containers. The first step in this work is the calculational program and comparison of calculations with existing data from measurements outside of containers (Refs. 13 and 14).

8.1.2.4 *Pu mass and thickness*

These can be determined from active measurements with Pu of two ^{240}Pu contents. The methods in Appendices A (Ref. 14) and C (Ref. 13) will be expanded to the use of large detectors adjacent to AT400 containers.

8.1.2.5 *U presence*

Detection of induced fission can be measured for various spherical shapes, masses, and enrichments of U in AT400 containers.

8.1.2.6 *Geometry*

No preliminary measurements needed because of past experience.

8.1.2.7 *U mass and enrichment*

The methods developed in Ref. 10 can be utilized here for mass and enrichment. Basically, transmission gives U content and induced fission determines the amount of ^{235}U.

8.1.3 Final System Tests

The system will be tested for the attribute determination methods it employs.

8.2 SOFTWARE FOR ATTRIBUTE EXTRACTION METHODS

The attribute extraction methods are incorporated into the system to display Pu presence, Pu mass above threshold, Pu 240/239 ratio <0.1, and Pu metal vs. non-metal for the passive system and for the case of HEU: presence, mass above threshold, enrichment >20% and metal vs. non-metal.

8.3 TEMPLATE MATCHING DEVELOPMENT

The advantage of template matching over attributes lies in the tight bounds that can be put on the radiation signature, as opposed to the loose bound the attribute threshold must use in order to protect classified attribute values like the isotopic ratio. The main problems of template matching are obtaining the initial template, storing the template, and the template's sensitivity to the "attributes of interest" of an inspection regime. These are additional problems that arise when template matching is applied in inspection regimes.

The approach suggested here is:

- Identify features of the measurement that will be in the template. In this case, the primary template values would be the numerical values of the attributes this system measures. Additional values could be selected for their sensitivity to other attributes

that give more assurance of the item's identity. In a strictly empirical approach, all of the measurement data could be used in the template.

- Make the initial template from items that have been monitored from their weapons origin to the facility. These become the reference items for the inspection regime.
- Make an attribute measurement on these reference items before using them as templates to verify that they are eligible.
- Set *a priori* bounds on the template so that it is assured that the reference item is within the attribute range expected in the regime. This is the template version of an attributes threshold decision. This step is useful if the template checks more attributes that the official "attributes of interest" measurement since it checks the reference item against these additional criteria.
- Instead of storing a template on computer media between inspection sessions, obtain the template from a measurement of a reference item for each verification campaign.
- Closely monitor the storage of reference items between inspection sessions to assure the inspectors that the items are not changed.

The benefits of this approach should be investigated.

8.4 LIMITED MEASUREMENTS WITH COMBINATION OF NMIS AND GAMMA RAY SPECTROMETRY

Simultaneous NMIS with a ^{252}Cf neutron source and gamma ray spectrometry measurements have been performed with HEU in 1997 at Y-12 and a photograph of the experimental setup is shown in Fig. 8.4.1. The HPGe detector from PNNL is shown on the left with a ^{252}Cf source on the right of the container. On the left side is shown a vertical array of NMIS detectors. A slight modification of the MCA software was made to minimize the upload of gamma ray results with essentially no time increase in the time for simultaneous operation over purely NMIS operation.

8.5 SCHEDULE

A passive combined NMIS-gamma ray spectrometry system can be built from COTS components and tested in six months. An active system which incorporates a DT generator and transmission detector can be assembled and tested in eight months. Using existing components, the combined active-passive system could be assembled with six months or less for testing.

8.6 RADIATION EXPOSURE

The active NMIS measurements with fissile material in containers requires the use of a DT neutron generator which produces 14.1 MeV neutrons. The generator for this application is of neutron intensity of 1×10^7 n/sec. The radiation dose from this source unshielded at 1 meter is ~15 mrem/hr. When this source is producing 14.1 MeV neutrons, there is no need

for personnel to be as close as one meter. Dose at 3 meters where personnel might occasionally be is as low as ~1.5 mrem/hr. This source can be turned off when not in use and presents no radiation concerns for storage. This source does contain tritium (2 curies) as a metallic tritide (ZrT) and the metallic tritide is contained in the sealed tube neutron generator.

8.7 SPACE REQUIREMENTS

The space requirements for NMIS are such that usually no modifications of the facility are required. Access is needed to two sides of the container for a passive system so the container could pass between the detectors for measurements. Two large scintillation detectors with photomultiplier tubes would be on opposite sides of the container. For an active system, a smaller proton recoil detector and DT source would be located on the other two sides with ~1 in. clearance around the container. A standard 19-in.-rack would contain the NMIS processor and gamma ray spectrometry processor, electronics, power supplies, computer, etc. The rack size would be 5-6 ft. high. At the top of the rack the red light green light display could be mounted. For the initial implementation for passive measurements, only the large proton recoil scintillators and gamma ray detector would be used.

Fig. 8.4.1. Photograph of Combined NMIS-Gamma Ray Spectrometry Measurement

9. CONCLUSIONS

A Multiple Attribute System can be configured to meet all needs for monitoring fissile material as well as HE and chemical agents. This system, which combines the Nuclear Materials Identification System (NMIS) methodology with gamma ray spectrometry, can be used to measure the following attributes of fissile material: Pu presence, Pu mass, 240/239 ratio, Pu time since reprocessing, Pu metal/non-metal, Pu geometry, U presence, U mass, U enrichment, U geometry, U metal/non-metal. Some of these attributes can be obtained by multiple data analysis methods and the system hardware and software could be configured to measure a selected subset. In addition, signatures from this system can be used for template matching such as has been implemented for confirmation of inventories and receipts for weapons components at the Y-12 National Security Complex in Oak Ridge since 1996. The active system employs a small, portable DT neutron generator, (10^7 n/s), two large proton recoil scintillators, a small proton recoil scintillator for transmission measurements, and an HPGe detector and multichannel analyzer for gamma ray spectrometry.

In six months,[30] a system for passive attribute measurements with Pu only could be completed. The passive system would measure Pu presence, Pu mass, 240/239 ratio, and Pu metal/non-metal. System would take six months to build and test prior to measurements with fissile material. Initial fissile material tests could take place with existing NMIS and gamma ray spectrometry systems. Upgrade to an active system for HEU could occur in two additional months.

Initially one passive system could be built and later upgraded to active measurements for HEU by adding the DT neutron generator and transmission detection.

The advantages of this passive system for Pu are as follows:

- Previous experience and knowledge of the system for attributes
- Can be built from commercial off-the-shelf components and tested
- Can also be used for template matching
- Inexpensive hardware costs
- Minimum facility modifications necessary because of small system size
- Expandable to HEU.

An active system adds an additional small proton recoil scintillation detector and a small, lightweight, portable DT neutron generator to the passive system. In addition to the advantages given above, the additional advantages of the active system are as follows:

- Obtains both Pu and HEU attributes
- May not require gamma ray spectrometry
- Radiation dose only ~10 mrem/hr at 1 meter from unshielded neutron source (The source is turned off when not in use.)
- Inexpensive hardware costs over passive system
- HE and chemical agent detection

[30] Estimates are times to perform technical work.

This one system is useful for a variety of nuclear material control and accountability applications. Incorporation of active gamma ray spectrometry with HPGe will allow detection of HE, chemical agents, and, in some cases, drugs.

APPENDIX A
PREVIOUS EXPERIENCE IN PLUTONIUM ATTRIBUTE ESTIMATION

PREVIOUS EXPERIENCE IN PLUTONIUM ATTRIBUTE ESTIMATION

Aside from evaluations of NMIS capabilities at the PANTEX Plant, the most relevant application of NMIS for plutonium attribute estimation stems from measurements performed jointly by Oak Ridge National Laboratory (ORNL) and Russian Federal Nuclear Center, All-Russia Scientific Research Institute of Experimental Physics (RFNC-VNIIEF) personnel at RFNC-VNIIEF facilities in Sarov, Russia in June and July 2000.

During these measurements at VNIIEF, NMIS was applied in its passive mode to eight unclassified plutonium spherical shells.[31] The shells' properties spanned the following ranges:

- Composition: δ-phase plutonium-metal, constant
- Relative ^{240}Pu-content (f_{240Pu}): f_{240Pu} = 1.77% {g ^{240}Pu / g Pu}, constant
- Inner radius (r_1): 10.0 mm ≤ r_1 ≤ 53.5 mm, mean r_1 = 33.5 mm
- Outer radius (r_2): 31.5 mm ≤ r_2 ≤ 60.0 mm, mean r_2 = 46.6 mm
- Radial thickness (Δr): 6.4 mm ≤ Δr ≤ 30.2 mm, mean Δr = 13.1 mm
- Plutonium mass (m_{Pu}): 1829 g ≤ m_{Pu} ≤ 4468 g, mean m_{Pu} = 3265 g

The features of these measurements were analyzed to extract the attributes of each plutonium shell. Given that the samples measured were of constant composition, geometry, and relative ^{240}Pu-content,[32] each shell is completely described by any two of the following four properties:

- Inner radius r_1
- Outer radius r_2
- Mass m, one of ^{239}Pu mass m_{239Pu}, ^{240}Pu mass m_{240Pu}, or total Pu mass m_{Pu} [33]
- Radial thickness Δr

Of these, generally only *mass* is acknowledged as an attribute of interest; the second property (whichever is chosen) can be considered to be a parameter of the attribute-estimation procedure, much as multiplication is a parameter necessary to accurately estimate fissile mass via most neutron measurements.

In fact, multiplication is intimately related to thickness. One result of neutron diffusion theory dictates that the effective multiplication factor, k_{eff}, of a spherical shell is controlled by the fissile mass m and geometric buckling B according to

---

[31] Only 1. 77%-^{240}Pu shells were available at RFNC-VNIIEF. This is unfortunate because without variation in the relative ^{240}Pu-content, it cannot be demonstrated from these measurements alone that NMIS is capable of extracting this attribute. Future joint measurements planned between the Russian Federal Nuclear Center, All-Russia Scientific Research Institute of Theoretical Physics (RFNC-VNIITF), RFNC-VNIIEF, and ORNL will demonstrate this capability through additional measurements of 10%-^{240}Pu shells at RFNC-VNIITF in Snezhinsk, Russia.

[32] These configurations are described in References 12 and 13: V. V. Gurov, M. I. Kuvshinov, V. A. Popov, V. P. Dubinin, J. K. Mattingly, and J. T. Mihalczo, "VNIIEF-ORNL Joint Plutonium Measurements with NMIS and Results of Plutonium Attributes Preliminary Evaluations," July 2001.

[33] Due to the constant relative ^{240}Pu-content of the VNIIEF shells, ^{239}Pu mass, ^{240}Pu mass, and total plutonium mass, are directly proportional to each other and are therefore equivalent measures of mass for attribute estimation purposes.

$$k_{eff} \sim \frac{m}{B^2}$$

("~" denotes "follows") where the geometric buckling B is proportional to the inverse of the radial thickness Δr:

$$B = \frac{\pi}{\Delta r}$$

In other words, for any particular mass, the thicker a spherical shell, the greater its multiplication M since:

$$M = \frac{1}{1 - k_{eff}}$$

In order to account for multiplication effects in the interpretation of the passive NMIS measurements, Monte Carlo calculations were performed to estimate the unreflected k_{eff} of the spherical shells listed in Table A-1. The dimensions given on the diagonal are those of the existing spherical shells; the off-diagonal dimensions include all hypothetical nestings of those shells. The span of relative ^{240}Pu-content was chosen to encompass both the VNIIEF (~ 2%-^{240}Pu) and the VNIITF (~ 10%-^{240}Pu) shells. The approximate relationship between total plutonium mass m_{Pu}, radial thickness Δr, relative ^{240}Pu-content f_{240Pu}, and multiplication factor k_{eff} was estimated via least-squares regression as:

$$k_{eff} \approx 0.034 \frac{m_{Pu}^{0.156} \Delta r^{0.465}}{f_{240Pu}^{0.015}}$$

Figure A-1 compares this model for the effective multiplication factor to the calculated values listed in Table A-1. Observe that over the span $1\% \leq f_{240Pu} \leq 20\%$ the effect on k_{eff} of relative ^{240}Pu-content is almost negligible. In other words, over this span multiplication is almost entirely dominated by mass and radial thickness.

Figure A-2 shows a typical distribution of real coincidence acquired from a passive NMIS measurement of one of the VNIIEF plutonium-metal spherical shells. Recall that the coincidence-distribution reflects the rate of real coincidence (i.e., total coincidence less accidental coincidence) as a function of the time-delay between detectors (at zero-delay, the detectors signals are synchronized). The narrow peak about the origin arises due to coincident pairs of gammas spawned by the same chain-reaction. The broader underlying bimodal distribution arises from coincident pairs composed of one gamma and one neutron created during the same chain-reaction. Not visible in the figure is a third single-mode distribution arising from coincident pairs of neutrons spawned by the same chain-reaction; it's amplitude is such that it lies beneath the preceding two distributions.

The features of the coincidence-distribution used to estimate the multiplication parameter and mass attribute are derived from its width and integral. For this study, the distribution width was taken as its full-width at tenth-maximum, or FWTM. The distribution's integral was taken as the area spanning the FWTM, a.k.a., its FWTM area. Subsequently, the distributions width and integral will respectively be denoted by W_{FWTM} and A_{FWTM}.

Table A-1. Monte Carlo Estimates of the Effective Multiplication Factor k_{eff} for Unreflected δ-Phase Plutonium-Metal Spherical Shells

Outer Radius (mm)	Relative ^{240}Pu-content (g ^{240}Pu / g Pu)	Inner Radius (mm) 10.0	14.0	31.5	40.2	46.6	53.5
14.0	1%	0.143					
	2%	0.142					
	5%	0.141					
	10%	0.139					
	15%	0.138					
	20%	0.136					
31.5	1%	0.492	0.455				
	2%	0.490	0.454				
	5%	0.487	0.451				
	10%	0.481	0.445				
	15%	0.475	0.440				
	20%	0.469	0.434				
40.2	1%	0.634	0.610	0.331			
	2%	0.633	0.608	0.331			
	5%	0.628	0.604	0.328			
	10%	0.621	0.596	0.324			
	15%	0.613	0.589	0.320			
	20%	0.605	0.581	0.315			
46.6	1%	0.734	0.715	0.495	0.277		
	2%	0.732	0.713	0.494	0.276		
	5%	0.727	0.709	0.490	0.274		
	10%	0.718	0.699	0.484	0.270		
	15%	0.709	0.691	0.478	0.267		
	20%	0.700	0.682	0.472	0.264		
53.5	1%	0.836	0.821	0.646	0.477	0.301	
	2%	0.834	0.819	0.645	0.475	0.301	
	5%	0.829	0.813	0.639	0.472	0.299	
	10%	0.819	0.804	0.632	0.466	0.295	
	15%	0.809	0.794	0.624	0.460	0.291	
	20%	0.798	0.784	0.616	0.454	0.287	
60.0	1%	0.928	0.916	0.771	0.632	0.493	0.295
	2%	0.925	0.915	0.769	0.631	0.493	0.295
	5%	0.920	0.908	0.764	0.626	0.489	0.292
	10%	0.909	0.897	0.756	0.619	0.483	0.289
	15%	0.899	0.886	0.745	0.611	0.477	0.285
	20%	0.886	0.875	0.737	0.604	0.471	0.282

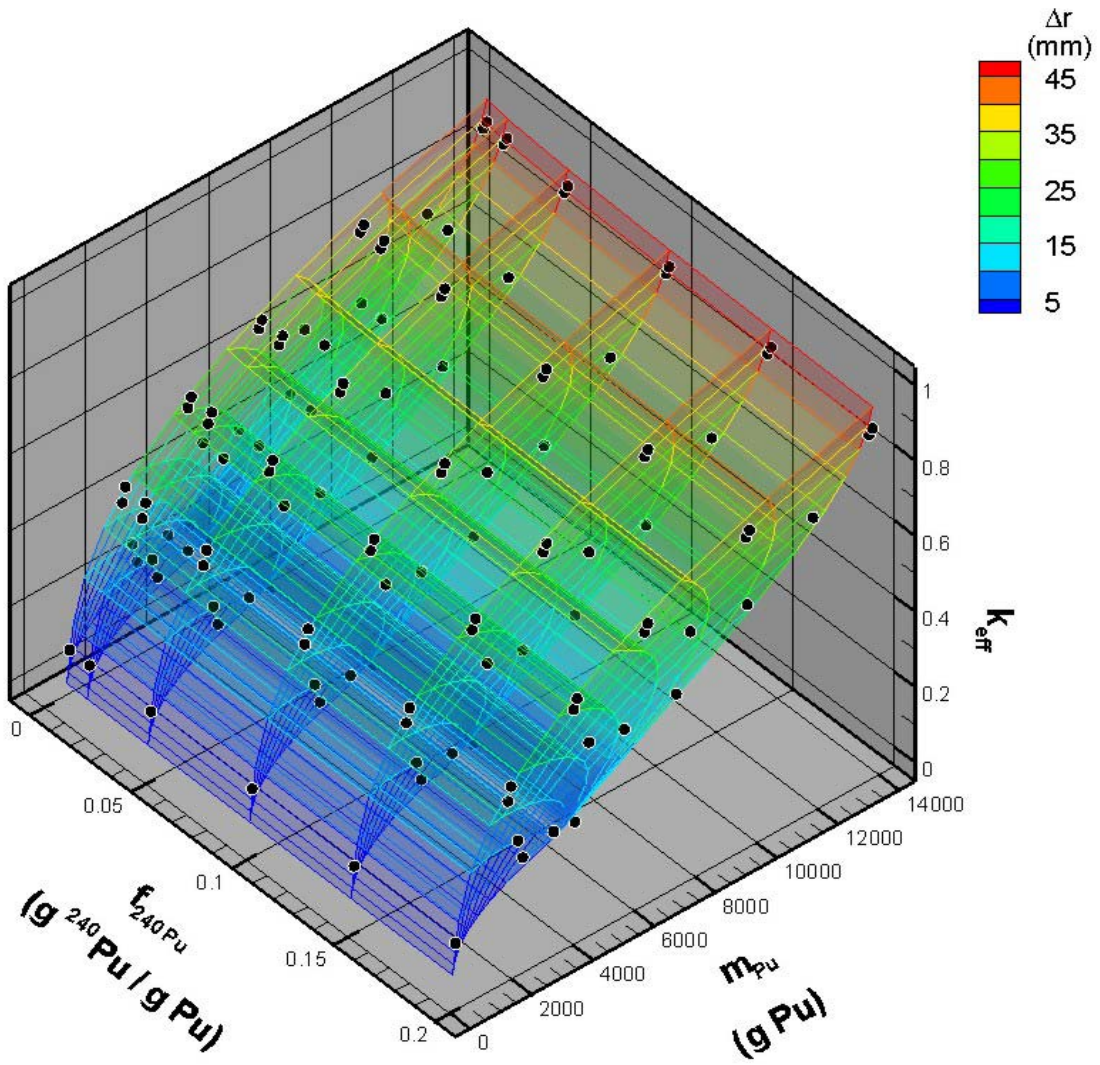


Fig. A-1. Comparison of k_{eff} Estimates From the Monte Carlo Calculations Listed in Table A-1 to a Least-Squares Regression Power Model. The black dots represent the Monte Carlo estimates; the isosurfaces versus total plutonium mass m_{Pu}, radial thickness Δr, relative ^{240}Pu-content f_{240Pu} represent the regression model.

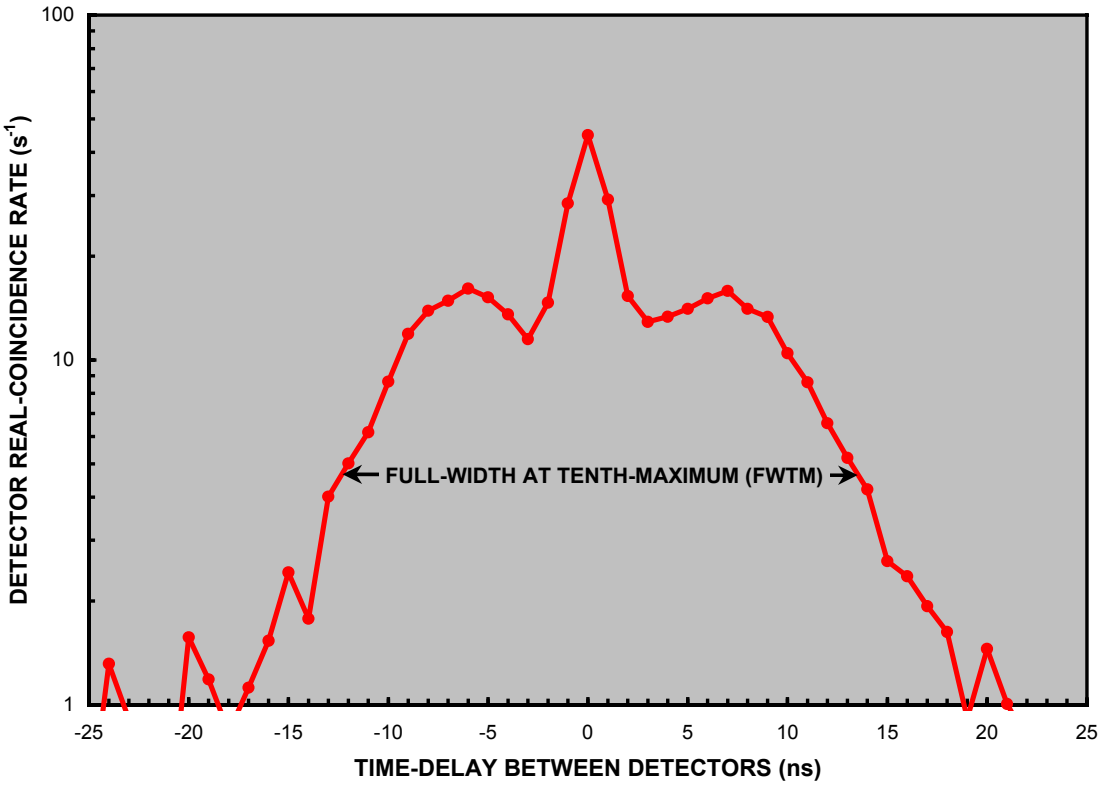


Fig. A-2. A Typical Coincidence-Distribution Acquired From a Passive NMIS Measurement of a VNIIEF Plutonium-Metal Spherical Shell. The plutonium mass and radial thickness of the specific shell measured to obtain this distribution was 3316 g and 6.9 mm, respectively (inner and outer radius: 46.6 mm and 53.5 mm, respectively). The features used for attribute-estimation were the distribution's full-width at tenth-maximum (FWTM) and it's FWTM area, i.e., the integral spanning the FWTM.

Figures A-3 and A-4 illustrate the dependence of the width and integral features upon multiplication and mass. The width tends to grow with increasing multiplication following a logarithmic trend, and the integral tends to grow with the product of multiplication and mass following a power rule. Least-squares regression quantifies these observations. The width feature is approximately proportional to multiplication according to

$$W_{FWTM} \approx 12.53 \ln M + 22.13 \tag{1}$$

and the integral feature is approximately proportional to the product of multiplication and mass according to

$$A_{FWTM} \approx 2.37 \left(M \cdot m_{240Pu}\right)^{1.14} \tag{2}$$

This *forward* model ((1) and (2)) is readily solved to yield the *inverse* model that estimates multiplication and mass given the width and integral features:

$$M \approx \exp\left(\frac{W_{FWTM} - 22.13}{12.53}\right) \tag{3a}$$

$$m_{240Pu} \approx \left(\frac{A_{FWTM}}{2.37}\right)^{1/1.14} \exp\left(-\frac{W_{FWTM} - 22.13}{12.53}\right) \tag{3b}$$

Substituting an observed width and integral feature into the preceding inverse model yields an estimate of multiplication and mass. The inverse model applied to the width and integral features observed for all the VNIIEF shells measured yields the estimates shown in Figs. 5 and 6. Note that in a root-mean-square (RMS) sense, the inverse model estimates multiplication to within 5% of its actual value and mass to within 7% of its actual value.

In its application to an unknown plutonium-metal spherical shell, the FWTM and FWTM area of the coincidence-distribution acquired from a passive NMIS measurement would be substituted into the inverse model to obtain an estimate of the ^{240}Pu mass according to (3b). If the unknown shell falls within the span of multiplication and mass shown in Figs. A-5 and A-6 then the relative error of the estimated ^{240}Pu mass will be on the order of 10%. With continued development of these methods, the error may be reduced.

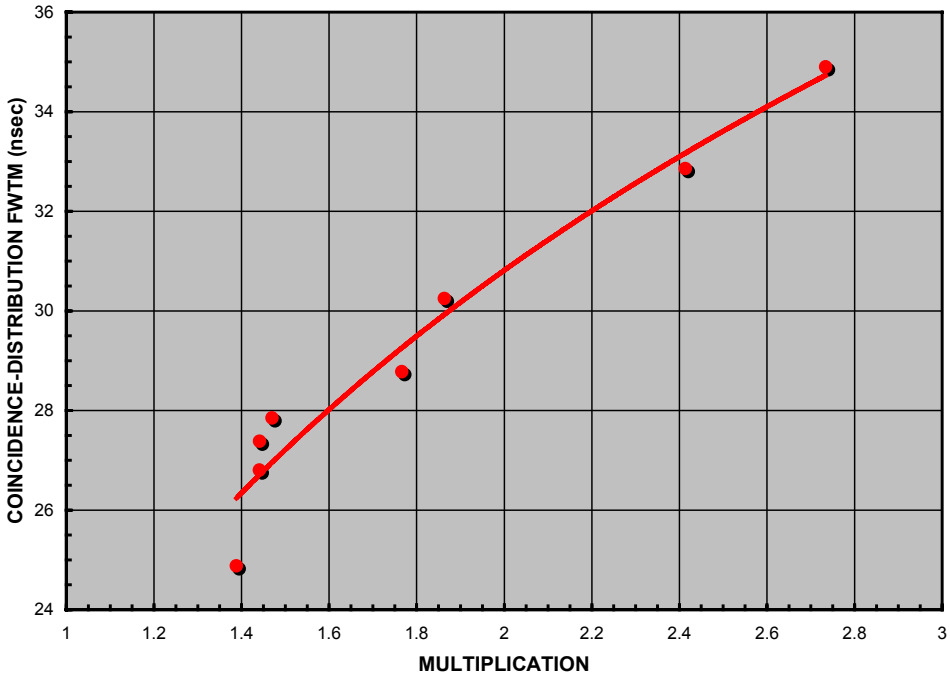


Fig. A-3. Dependence of the Width Feature Upon Multiplication

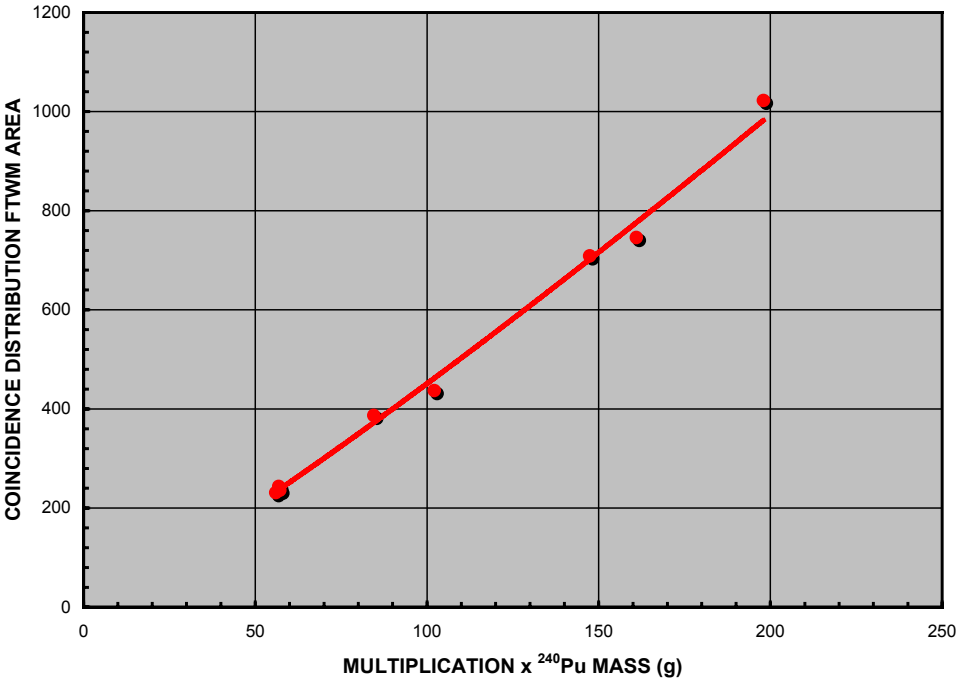


Fig. A-4. Dependence of the Integral Feature on the Product of Multiplication and Mass

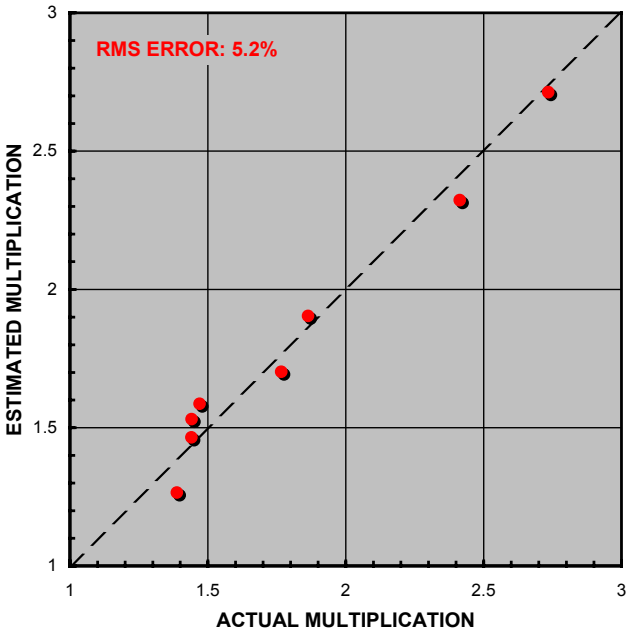


Fig. A-5. Comparison of Multiplication Estimates Obtained From the Inverse Model to the Actual (i.e., Monte Carlo Calculated) Multiplication of Each VNIIEF Shell Measured

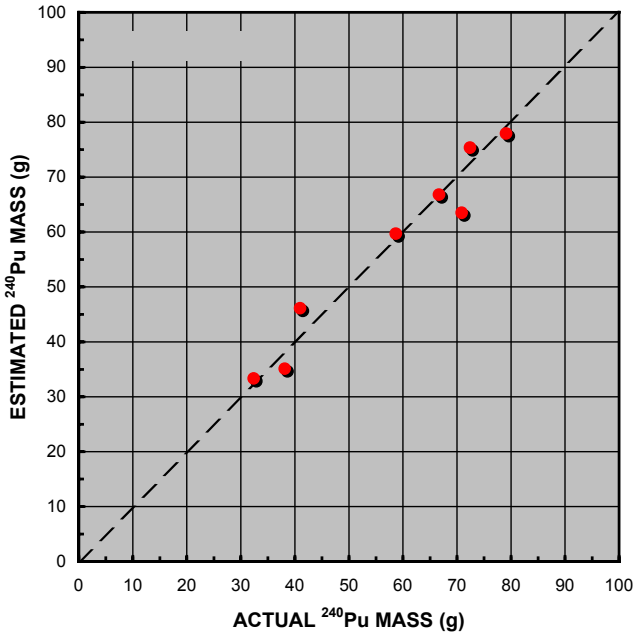


Fig. A-6. Comparison of ^{240}Pu Mass Estimates Obtained From the Inverse Model to the Actual ^{240}Pu Mass of Each VNIIEF Shell Measured

APPENDIX B
RUSSIAN WORK ON PU ATTRIBUTES FROM NMIS DATA

VNIIEF-ORNL Joint Plutonium Measurements with NMIS and Results of Plutonium Attributes Preliminary Evaluations

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ABSTRACT

Within the frameworks of TO № 007 between ORNL and VNIIEF on Nuclear Materials Identification System (NMIS) mastering at VNIIEF in July 2000 there had been finalized joint measurements, in which NMIS-technique equipment was used that had been placed at VNIIEF's disposal by ORNL, as well as VNIIEF-produced unclassified samples of fissile materials. In the report there are presented results of experimental data preliminary processing to obtain absolute values of some attributes used in plutonium shells measurements: values of their mass and thickness. Possibility of fissile materials parameters absolute values obtaining from measurement data essentially widens NMIS applicability to the tasks relevant to these materials inspections.

1. Parameters of plutonium shells under study.

The plutonium shells mass-geometric parameters (δ-phase, ^{240}Pu percentage is 1,8) are presented in Table 1.

Table 1. Parameters of the plutonium assemblies

assembly number	$R_{int.}$, cm	$R_{ext.}$, cm	Mass, g	Spherical components dimensions		Neutron multiplication, q
				$R_{int.}$, cm	$R_{ext.}$, cm	
1	1,00	4,02	4091,0	1,00	1,40	2,26
				1,40	3,15	
				3,15	4,02	
2	1,40	4,02	3768,0	1,40	3,15 *)	2,07
				3,15	4,02 **)	
3	3,15	4,66	4468,3	3,15	4,02	1,46
				4,02	4,66	
4	5,35	6,00	4004,4	-	-	1,16
5	1,40	3,15	1829,0	-	-	1,51
6	4,02	4,66	2315,1	-	-	1,12
7	4,66	5,35	3316,1	-	-	1,18
10	3,15	4,02	2153,2	-	-	1,22

*) without a stopper in the upper hemisphere

**) without stoppers in either of the hemispheres, stopper diameter ~ 2,20 cm.

For the above plutonium assemblies both active and passive measurements have been fulfilled for "bare" assemblies as well as for the same assemblies containerized into AT 400.

Radiation signatures of the studied system obtained in NMIS-measurements present an objective representation of its gamma-neutron characteristics and can be used to get diverse information relevant to the system. The way to deal with the measured signatures is different for classified and unclassified objects.

To obtain numerical values of such attributes of an object under study as, for example, values of fissile substance mass and dimension, absolute values of the measured signatures need to be used. Such analysis can be applied only to unclassified objects measurements.

Results of the performed measurements can be also presented in non-intrusive form – as a ratio of signatures of an inspected object and a "reference" one. Unclassified data obtained in such way may be useful for containerized classified objects safety (identity) non-intrusive inspecting. NMIS-measurements application to such tasks has been described earlier in ORNL reports, e.g. see [1] and[2].

2. Determining plutonium spherical shells mass and thickness.

2.1. Measurements with "bare" assemblies.

The signatures of plutonium spherical shells obtained in the measurements can be employed to find mass and thickness of "unknown" assemblies under certain subsidiary conditions. Videlicet, one need to be convinced that all the assemblies, both investigated and "unknown", are spherical shells produced of δ-phase metallic plutonium, and all assemblies have the same isotopic composition. Such requirements being met, the signatures of NMIS-measurements depend only on two assemblies parameters – shells mass and thickness, for example.

An idea of shell mass and thickness values deriving from the measurements data consists in following. First, the results of measurements of several known plutonium shells are used to obtain dependencies that associate measured signatures peculiarities with shells parameters. The obtained dependencies are then used to determine other shells parameters from results of their signatures measuring.

As spherical shells are completely characterized by two parameters, two independent equations need to be derived, associating any pair of independent characteristics of the measured signatures with the shells mass and thickness (regression equations). First estimation of such possibility was carried out by one of the joint measurements participants – J. Mattingly (ORNL) in the course of measurements in summer 2000. To obtain the sought equations he used the results of measurements in seven assemblies, the values of mass and thickness for the eighth assembly (assembly № 10 from Table 1) were then derived from its signatures measurement results and the regression equations. Comparison of the calculated values with the factual ones characterizes an accuracy degree of such procedure of shell mass and thickness determining. In one variant of such analysis J. Mattingly simultaneously used the results of both active and passive measurements. The below characteristics of the measured signatures have been obtained in active measurements, where distance between a californium chamber and detectors face surface constituted 18,7 cm, and for the passive measurements at a distance of 13 cm between the "bare"

assembly center and the detectors face surface. At in-container measurements, the distance was about 25 cm.

As F_1 – first characteristic of the measured signatures – there was used a sum of values of cross-correlation functions between the detectors pairs within the limits of delay time values $\tau = \pm 50$ ns, obtained using the passive measurements data, divided by the total measurement time T. Summation over all the detector pairs is carried out to improve the results statistical accuracy.

$$F_1(M,\Delta) = \frac{1}{T}\sum_{i\neq j}\sum_{-50ns}^{50ns} CC_{ij}(\tau), \frac{1}{\text{sec}} \qquad (1)$$

Here $CC_{ij}(\tau)$ is a value of the cross-correlation function between detector i and detector j at inter-signal delay time τ. T is the total measuring time = N×512×1 nsec, where N is the number of data blocks, employed to calculate $CC_{ij}(\tau)$ values. F_1 value is an average count rate of correlated events for all the detectors pairs, obtained at measurements with plutonium shell of mass M and thickness Δ. In linear regression representation $F_1(M,\Delta)$ function is approximated by a linear dependence on M and Δ:

$$F_1(M, \Delta) \approx A_{1M} \times M + B_{1\Delta} \times \Delta + C_1 \qquad (2)$$

Here M is the shell mass, Δ is the shell thickness, A_{1M}, $B_{1\Delta}$ and C_1 are the regression equation coefficients to be found from a condition of best fit for describing results of measuring seven assemblies selected as reference ones.

As F_2 – second characteristic of the measured signatures – there were used active measurements results: a sum of values of cross-correlation function between the californium chamber and the detector in the time interval corresponding to induced fission neutrons recording divided by the total measuring time T. This portion beginning corresponds to californium source neutrons recording cut-off due to the selected discrimination threshold. For the performed measurements conditions it corresponds to the point of time ~ 11 nsec:

$$F_2(M,\Delta) = \frac{1}{T}\sum_{-11ns}^{50ns}[CC_{12}(\tau) + CC_{13}(\tau)] \approx A_{2M} \times M + B_{2\Delta} \times \Delta + C_2 \qquad (3)$$

$$[F_2] = 1/\text{sec}$$

where $CC_{12}(\tau)$ and $CC_{13}(\tau)$ are cross-correlation functions between the chamber and detectors № 2 and № 3 according to the active measurements data. Like the previous case, signatures summation improves statistical accuracy of the results.

Making use of linear regression simulating procedure available in Microsoft Excel applied to the results of measurements with seven samples, coefficients A, B and C have been obtained, which being used in the regression equations give the best description of the experimental data. If the mass M is expressed in grams, and the thickness Δ is expressed in millimeters, then the regression equations obtained in such a way look like:

$$F_1 \approx 0{,}19844 \times M + 20{,}158 \times \Delta - 399 \qquad (4)$$

$$F_2 \approx 0{,}670318 \times M - 105{,}345 \times \Delta + 5443{,}94 \qquad (5)$$

In Table 2 there are presented results of mass and thickness computation for the eighth (reference) sample, which was not involved into the regression equation elaboration.

Table 2

Mass, gram			Thickness, mm		
M_{fact}	$M_{computed}$	Error	Δ_{fact}	$\Delta_{computed}$	Error
2153,2	2461,8	14,3%	8,70	8,0	7,8%

There has been undertaken an attempt to construct regression equations using some other characteristics of the measured signatures, F_3 (F_4) in particular – a characteristic, representing the first gamma-peak square using active measurements data.

As the first gamma-peak square depends mainly on the shell thickness, at first, there has been tested one-parameter – exponential and power – dependence of the square upon the shell thickness.

$$F_3(\Delta) = \frac{1}{T}\sum_{\sim 0ns}^{\sim 3ns}[CC_{12}(\tau) + CC_{13}(\tau)] \approx A_{2\Delta} \times e^{-B_{3\Delta}\times\Delta} \qquad [F_3] = 1/\text{sec} \quad (6)$$

$$F_4(\Delta) = \frac{1}{T}\sum_{\sim 0нсек}^{\sim 3нсек}[CC_{12}(\tau) + CC_{13}(\tau)] \approx A_{4\Delta} \times \Delta^{-B_{4\Delta}\times\Delta} \qquad [F_4] = 1/\text{sec} \quad (7)$$

In Fig. 1 there are shown a graph and equation of the exponential regression, built (like the above) upon the results of seven assemblies measuring.

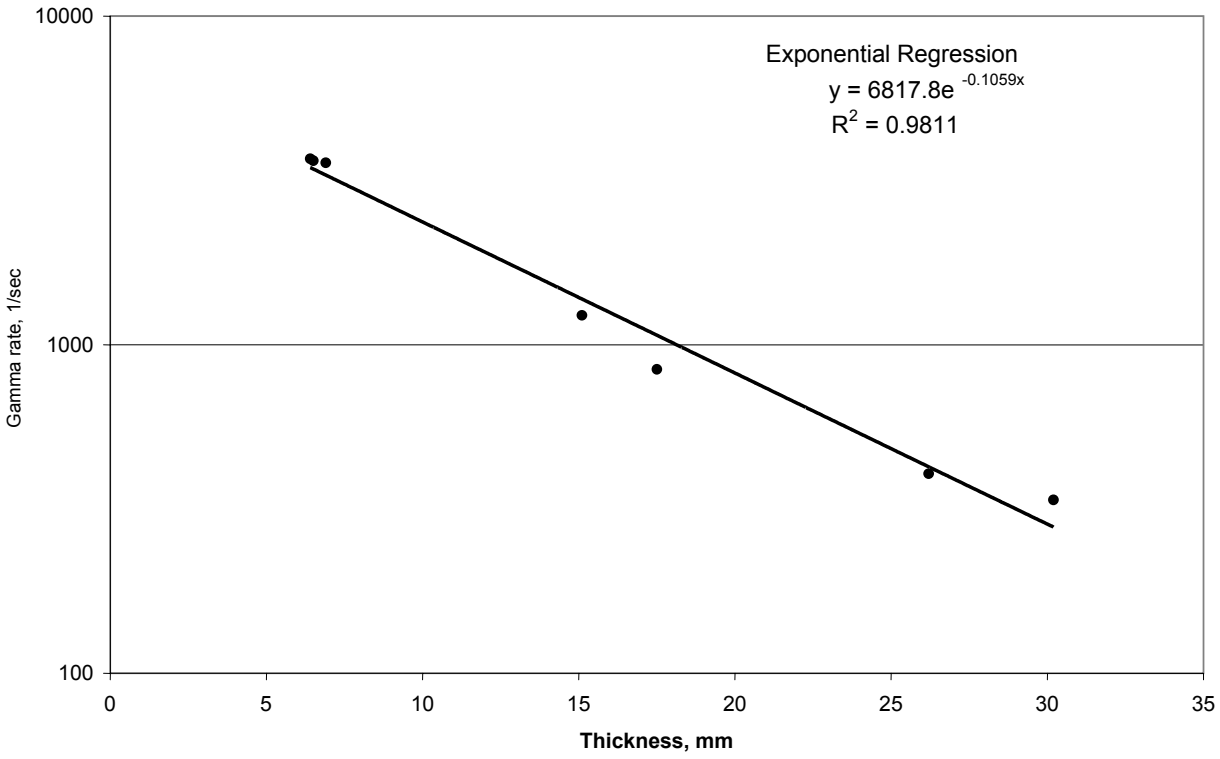

Fig. 1. Exponential regression for average count rate of the source gamma-rays, built on results of measurements of seven "reference" samples (assemblies).

In terms of the above designations the exponential regression equation for the shell thickness is given by:

$$F_3(\Delta) = 6817{,}8 \exp(-0{,}1055\Delta) \ , [\Delta] = \text{mm}, \qquad (8)$$

As it is seen from the experimental data marked in Fig.1 with separate points, gamma-ray count rate dependence on the sample thickness differs from the simple exponent, particularly at large values of the thickness. It seems to be due to the fact that exponential dependence is more suitable under "good" geometry, when multiple scattering quanta contribution to the signal value is absent, but that was not fulfilled in our measurements.

In Fig.2 it is presented a power regression of gamma-ray counting dependence on the sample thickness constructed using the same seven experimental points. It is seen to approximate the experimental data rather better than an exponent does.

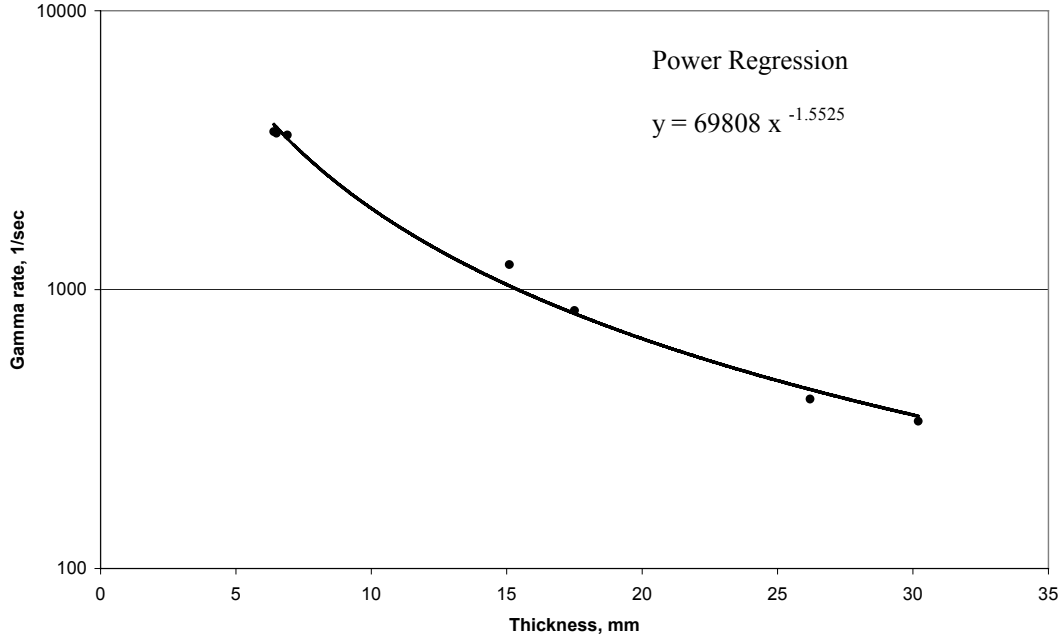

Fig. 2. Power regression for average count rate of the source gamma-rays, constructed to fit seven experimental points

In terms of the above designations power regression for the shell thickness is given by:

$$F_4(\Delta) = 69808 \ \Delta^{-1{,}5525} \ , [\Delta] = \text{mm}. \qquad (9)$$

In Table 3 there are presented values of mass and thickness of the eighth (reference) sample, which was not involved into the regression construction, computed using measured value of the correlated gamma-peak square from the equations of exponential and power regression:

Table 3

Factual thickness, mm	Exponential regression		Power regression	
	Calculation	Error	Calculation	Error
8,70	9,64	10,8%	8,64	0,7%

It is seen that power type regression being used gives much better approximation of the computed value to the factual thickness value.

In addition to this there has been considered two-parameter linear dependence of logarithm of the first gamma-peak square as a function of the shell mass and thickness.

$$F_5(M,\Delta) = \frac{1}{T}\sum_{\sim 0ns}^{\sim 3ns}[CC_{12}(\tau) + CC_{13}(\tau)]$$

$$Ln[F_5(M,\Delta)] \approx A_{5M} \times M + B_{5\Delta} \times \Delta + C_5 \qquad [F_5] = 1/\text{sec} \qquad (10)$$

Regression constructed using data for the same seven assemblies looks like:

$$LnF_5 \approx 6{,}1625\times10^{-5}\times M - 0{,}107586 \times \Delta + 8{,}644 \qquad (11)$$

In Table 4 there are presented values of mass and thickness of the eighth (reference) assembly, which was not involved into the regression construction, computed using diverse combinations of the measured signatures characteristics (the factual value of this assembly thickness constitutes 8,7 mm, value of mass is 2153 g).

Table 4

Signature characteristic to construct regression	Shell thickness		Shell mass	
	Calculation	Error	Calculation	Error
F_2, F_1	8,0	7,8%	2461,8	14,3%
F_3, F_1	9,64	10,8%	2297,6	6,7%
F_3, F_2	9,64	10,8%	2715,7	26,1%
F_4, F_1	8,64	0,7%	2399,6	11,4%
F_4, F_2	8,64	0,7%	2558,0	18,8%
F_5, F_1	9,1	5,0%	2349,0	9,1%

In the described investigations of a possibility of sample mass and thickness predicting either signatures of active measurements only were used, or signatures of both active and passive types together. When only passive signatures are used, the equation (1) can be taken as one of the equations and the second one can be obtained by constructing a linear regression for the detector signal autocorrelation function

$$F_6(M,\Delta) = \frac{1}{T}\sum_{i=1}^{i=4}\sum_{\tau=-50}^{\tau=50} AC_i(\tau) \approx A_{6M}\cdot M + B_{6\Delta}\cdot\Delta + C_6 \qquad (12)$$

Here $AC_i(\tau)$ is an autocorrelation function of i'th detector at the passive measurements.
The equations set for reference sample mass and thickness predicting has been obtained by results of measurements with six assemblies. Assembly № 2 has been excluded from consideration, as it has slightly broken spherical symmetry. In this case coefficients of the regression equation F_1, obtained earlier, when results for seven assemblies were used, slightly change (see (4)):

$$F_1' \approx 0{,}19999 \times M + 22{,}011 \times \Delta - 418 \qquad (4')$$

$$F_6 \approx 57{,}432 \times M - 6770 \times \Delta + 163945 \qquad (13)$$

In Table 5 there are presented results of mass and thickness computation of the seventh reference sample, which was not involved into the regression equation elaboration:

Table 5

Signature characteristic to construct regression	Shell thickness		Shell mass	
	Calculation	Error	Calculation	Error
F_6, F'_1	9,95	14,4%	2253,1	4,6%

Comparison between data from the Tables 3…5 shows that accuracy of the shell mass prognosis based on the passive measurements data only is ~ 2 times better, than that obtained using mixed data of active and passive measurements, and accuracy of the shell thickness prognosis on the contrary is much worse. The best accuracy of the shell thickness prognosis is achieved when data on the prompt source gamma-quanta passage in active measurements are used.

1.1. Containerized assemblies measuring.

In Table 6 there are presented results of mass and thickness evaluations based on the available data of passive measurements with assemblies placed inside AT400 container. To construct the regression equations the measured values of the detectors autocorrelation functions and cross-correlation functions between the detectors pairs have been used.

Table 6

Shell parameter	Factual values	Computed values	Error
Mass, g	2153,2	2996,3	39,2%
Thickness, mm	8,7	6,54	-24,8%

An accuracy of sample mass and thickness predicting for containerized assemblies is seen to be essentially worse than that for "bare" assemblies. Partially it may be due to reduction of number of reference points used to select coefficients for the regression equations (in the passive measurements assemblies № 6 and 7 from the list presented in Table 1 were not included into a container), partially to design of the AT400 container, having in its composition materials with considerable content of light elements (wood, polyethylene doped with boron), which results in noticeable neutron weakening and worsening experimental data statistical accuracy. Calculations carried out in ORNL [3] have shown that to improve measurements accuracy for strongly protected containers and to reduce exposition time large plastic scintillation detectors 50x50x10cm placed on the container opposite sides need to be employed in the passive measurements.

Above there are presented results of the simplest approach to parameters evaluation of plutonium assemblies to be studied (inspected), built upon the regression equations coefficients selection. For the same purposes more complicated mathematical approaches may be used, e.g. based on the neuron-net method. At VNIIEF the first attempt has been made to apply this method to evaluate parameters of plutonium assemblies the same as presented in the report and on the base of results of the same measurements. This work was encouraged by the results obtained in the report [4].

It may be mentioned that, when experimental data obtained for the same "bare" assemblies both in active and passive measurements are used to "train" a neuron net, accuracy of "unknown"

assembly parameters predicting may constitute 1-2 %, what is essentially better, than that provided by the regression approach.

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APPENDIX C
MONTE CARLO CALCULATIONS
FOR PASSIVE NMIS MEASUREMENTS OF PU IN AT400 CONTAINERS

MONTE CARLO CALCULATIONS
FOR PASSIVE NMIS MEASUREMENTS OF PU IN AT400 CONTAINERS

In addition to the measurements described in Appendix A, Monte Carlo neutron and gamma transport theory calculations were performed for a pair of delta phase Pu (6% ^{240}Pu) metal spheres with masses of 0.5, 1.0, 1.5, and 2.0 kg.[19] In one additional calculation, one Pu metal sphere of 2 kg and one Pu metal sphere of 1.5 kg. The spheres were in AT400-R containers. Detectors were plastic scintillators (sensitive to both neutrons and gamma rays) which were 50 cm × 50 cm × 10 cm-thick on each side of the container. The total correlated counts between the pair of detectors are plotted vs. Pu mass in Fig. C-1. From this curve, it is obvious if the lowest mass spheres (0.5 kg each) were used as a calibration standard, the total correlated count rate above that for these 0.5 kg spheres could be used for Pu metal mass above threshold determination. For two 2 kg spheres in the container, the correlated count rate is ~3600 counts per minute. Thus, if 10,000 total correlated counts were adequate for this determination, the verification for pairs of 2 kg spheres could be obtained from a few minutes of data while that for pairs of 1 kg spheres would require 10 minutes of measurement time. Since these plastic scintillation detectors are sensitive to both neutrons and gamma rays, these enhanced correlated count rates are achievable since gamma rays penetrate out of the AT400 containers more easily than neutrons.

The conclusion of these Monte Carlo calculations is that passive NMIS measurements can determine Pu mass above threshold with two plastic scintillation detectors (50 cm × 50 cm × 10 cm-thick) on opposite sides of the container in measurement time less than 10 minutes.

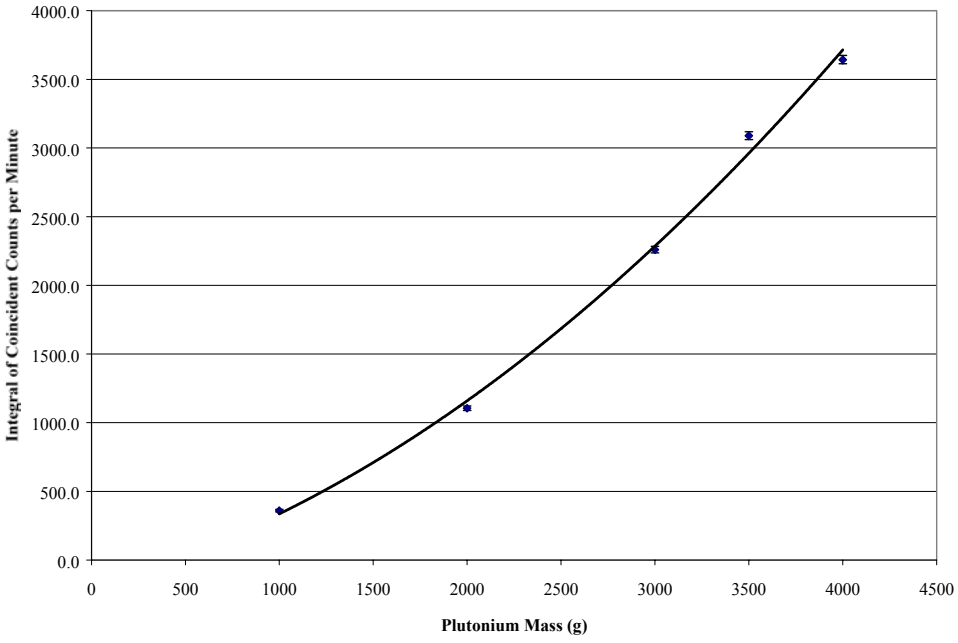

Fig. C.1. Total Coincident Counts vs. Pu Mass in an AT400R Container