

NMIS PLUS GAMMA SPECTROSCOPY FOR ATTRIBUTES OF HEU, PU, AND HE DETECTION

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ABSTRACT

A combined Nuclear Materials Identification System (NMIS)-gamma ray spectrometry system can be used passively to obtain the following attributes of Pu: presence, fissile mass, 240/239 ratio, and metal vs. oxide. This system can also be used with a small, portable, DT neutron generator to measure the attributes of highly enriched uranium (HEU): presence, fissile mass, enrichment, metal vs. oxide; and detect the presence of high explosives (HE). For the passive system, time-dependent coincidence distributions can be used for the presence, fissile mass, metal vs. oxide for Pu, 240/239 ratio, and gamma-ray spectrometry can also be used for 240/239 ratio and presence, allowing presence and 240/239 ratio to be confirmed by two methods. For the active system with a DT neutron generator, all relevant attributes for both Pu and HEU can be determined from various features of the time-dependent coincidence distribution measurements. Active gamma ray spectrometry would determine the presence of HE. The various features of time-dependent coincidence distributions and gamma ray spectrometry that determine these attributes are discussed with some examples from previous determinations.

INTRODUCTION

The concept for the Multiple Attribute System is a Nuclear Materials Identification System (NMIS) time-dependent coincidence processor [1] 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 combined 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. This system can be built using commercial off the shelf (COTS) components. 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. This paper describes the system and explains the attribute determination methods for fissile materials.

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. 1).

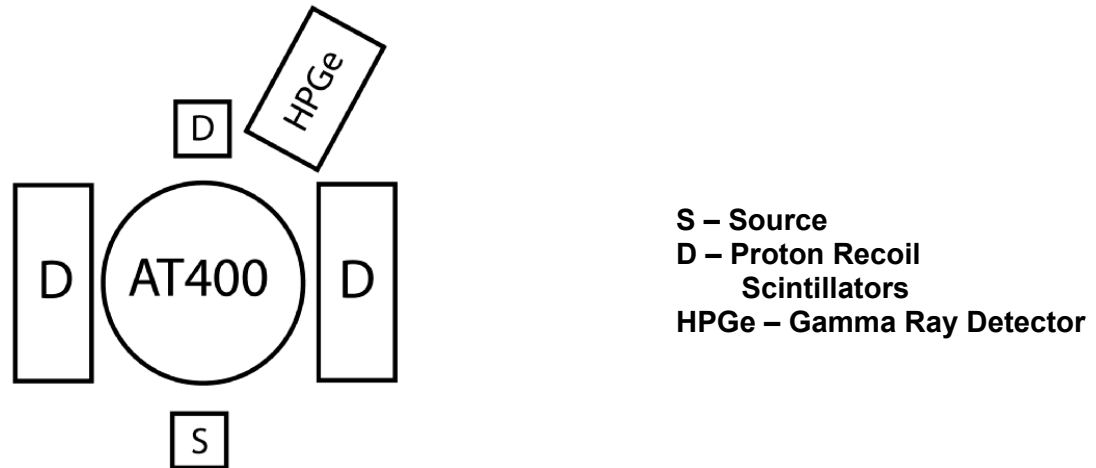


Fig. 1. Sketch of Configuration of System

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 large proton recoil scintillation detectors are stationary on each side of the container. A third smaller proton recoil scintillation 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.

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. [2] 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, using 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, [3] reduced radiation exposure, 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. It is a cylinder 40 inches long with 3 inches in diameter. The neutron tube has a built-in alpha particle detector that is used to tag 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). This system and its advantages have been described (Ref. 2). 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 is only 8.75-in.-long and could be separated from the power supply.

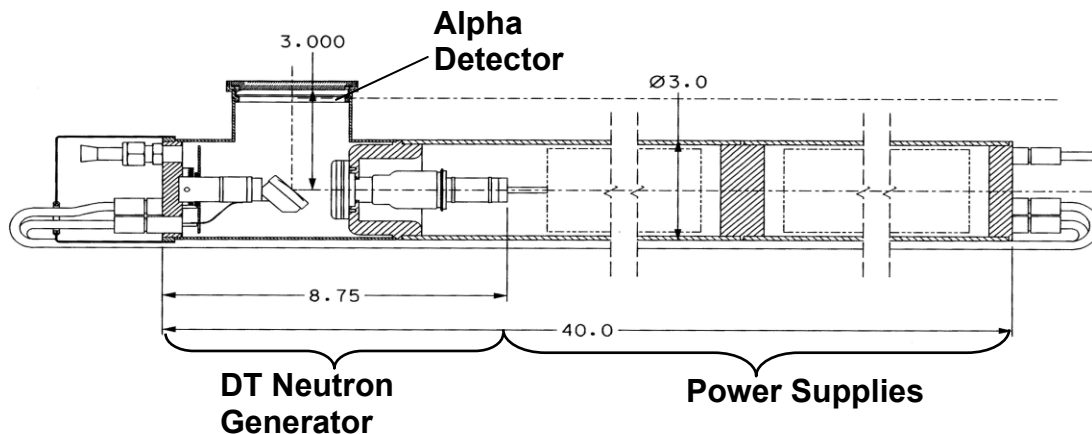


Fig. 2. Sketch of a Small, Portable, Lightweight DT Generator

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 as needed and can be built from COTS components.

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 1. Table 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. 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 and 240/239 ratio. Time dependence coincidence could also be used for Pu 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. 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 determine 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.

ATTRIBUTE DETERMINATION METHODS FOR FISSILE

Some of the attribute detection methods of Table 1 are described briefly, mainly some of those associated with the time dependent coincidence distribution methods of NMIS since those associated with gamma ray spectrometry are well known. NMIS measures the time distribution of correlations between a pair of detectors in both an active or passive measurement and also measures the time distribution of correlations between a single detector and the source in an active measurement. Sketches of these distributions are given in Figs. 3 and 4. In addition, it measures correlations between three detectors (source or detectors) as well as the number of times m pulses occur in a time interval ($m=0, 1, 2, \dots$).

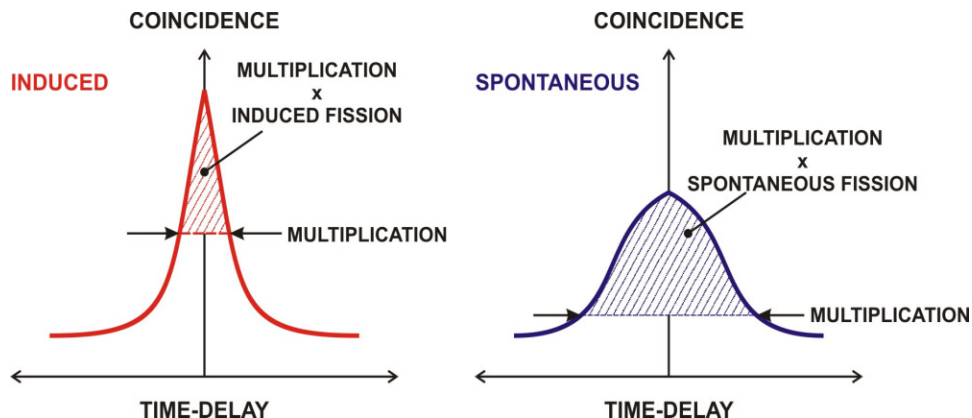


Fig. 3. Features of the Measurement Used to Infer the Presence, Relative ^{240}Pu -Content, and Fissile Mass Attributes of Plutonium Samples. (The widths of both the induced and spontaneous time-dependent coincidence-distributions are each proportional to multiplication. The integral of the induced coincidence-distribution 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.)

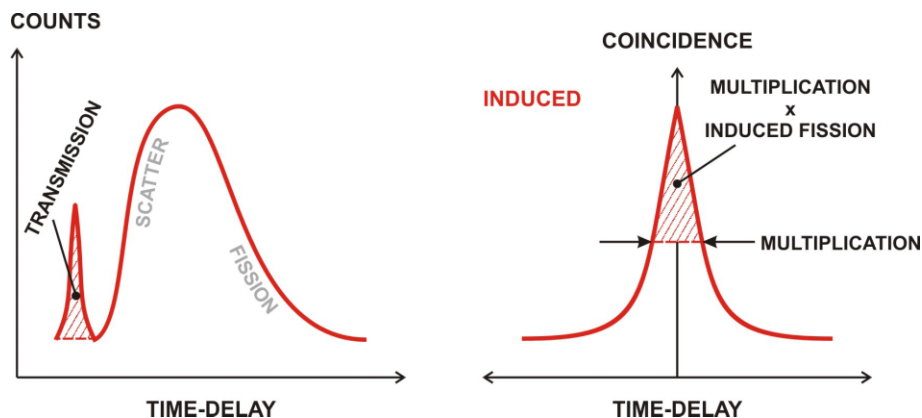


Fig. 4. Features of the Measurement Illustrated in Fig. 1.3 Used to Infer the ^{235}U -Enrichment and Fissile Mass Attributes of Uranium Samples. [The integral of the first peak in the count-distribution is proportional to the neutron transmission rate and thus total uranium. The width of the coincidence-distribution is proportional to multiplication and mass of ^{235}U , and its integral is proportional to the product of multiplication and induced fission.]

Spontaneous fission induced correlation of ^{240}Pu indicate the presence of Pu and induced fission without inherent source fission indicates the presence of HEU. From data like Fig. 3b, the spontaneous fission rate can be obtained since the integral of the distribution is related to the product of the spontaneous fission rate and the multiplication and the width of the distribution is related to the multiplication. Knowing the 240/239 ratio from gamma ray spectrometry, the mass of ^{239}Pu can be obtained. [4]

The 240/239 ratio can be obtained from an active measurement with Pu as follows. The ^{240}Pu constituent is a spontaneous source of time-dependent coincidences neutrons and gamma rays, and the ^{239}Pu content is a source of time-dependent coincidences from induced fission. The ratio of the spontaneous and induced sources, when adjusted for the effects of total mass and multiplication, is proportional to the $^{240}\text{Pu}/^{239}\text{Pu}$ mass ratio. Correlations between detectors are used to separate the induced part of the distribution from the inherent by rating the coincidence on the active source signal. Thus, this method could potentially determine the 240/239 ratio without gamma ray spectrometry. Alternatively, in a passive measurement the total coincident pairs between detectors are primarily related to ^{240}Pu mass, whereas the width of the time distribution of coincidences (1/10 maximum value) is primarily related to the ^{239}Pu mass through self multiplication and thus the ratio of the two isotopes can be obtained.

The absence of Pu or U metal can be determined for a passive measurement since the shape of the time correlation functions depend on the form of the material (say metal vs oxide), and this is discussed for Pu in another paper at this conference. [5] In addition, active gamma ray spectrometry will produce 6.129 KeV gamma rays from interaction of 14.1 MeV neutrons with oxygen or fluorine. Geometry of HEU or Pu can be obtained by scanning or using a vertically segmented detector for vertical profiles of shape

For the uranium mass attribute, the ^{235}U constituent can easily be induced to fission by the active source and consequently by measuring the induced fission rate (Fig. 4b) the ^{235}U mass can be estimated. In this same type of measurement, the neutron transmission can be used to obtain the total uranium content thus also giving the ^{235}U enrichment. This has successfully been demonstrated with HEU at Y-12 when the relative enrichment and HEU masses were defined to $\pm 1.5\%$ in five minutes. [6]

CONCLUSIONS

Such 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 age 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 with threshold displays if desired. 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. This one system, which has a small facility footprint and uses low intensity neutron sources (<15 mrem/hr at 1 meter), is a cost-effective way of determining the attributes of fissile materials. 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.

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Table 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 Passive	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	
drugs	presence	1	gamma spectrometry	ratios of N/C, H/C, and O/C	active	neutron source, HPGe ^a or BGO ^b	

^aMethods of A. J. Caffrey; see Reference 4.

^bG. Vourvopoulos 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).