ORNL/TM-2012/22

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February 2012

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ORNL/TM-2012/22

Global Nuclear Security Technology Division

NUCLEAR MATERIAL IDENTIFICATION SYSTEM WITH IMAGING AND GAMMA-RAY SPECTROMETRY FOR PLUTONIUM, HIGHLY ENRICHED URANIUM, HIGH EXPLOSIVES, AND OTHER MATERIALS

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Date Published: February 2012

Prepared by OAK RIDGE NATIONAL LABORATORY P.O. Box 2008 Oak Ridge, Tennessee 37831-6285 managed by UT-Battelle, LLC for the U.S. DEPARTMENT OF ENERGY under contract DE-AC05-00OR22725

NUCLEAR MATERIAL IDENTIFICATION SYSTEM WITH IMAGING AND GAMMA-RAY SPECTROMETRY FOR PLUTONIUM, HIGHLY ENRICHED URANIUM, HIGH EXPLOSIVES, AND OTHER MATERIALS*

The Nuclear Material Identification System (NMIS) has been under development at ORNL and the National Nuclear Security Administration (NNSA) Y-12 National Security Complex since 1984. In the mid-1990s, what is now the US Department of Energy (DOE) Office of Nuclear Verification (ONV) realized that it was a useful technology for future arms control treaty applications and supported further development of the system. In 2004, fast-neutron imaging was incorporated into the system. In 2007, the ONV decided to develop a fieldable version of the system, designated as FNMIS, for potential use in future treaties. The FNMIS is being developed to be compatible with the eventual incorporation of gamma-ray spectrometry and an information barrier. This report addresses how and what attributes could be determined by the FNMIS system with gamma-ray spectrometry. The NMIS is a time-dependent coincidence system^{1,2} that incorporates tomographic imaging³ (including mapping of the fission sites^{4,5}) and gamma-ray spectrometry. It utilizes a small, lightweight (30 lb), portable deuterium-tritium (DT) neutron (14.1 MeV) generator $(4 \times 10^7 \text{ neutrons/second})^6$ for active interrogation and can also perform passive interrogation. A high-purity germanium (HPGe) gamma-ray detector with multichannel analysis can be utilized in conjunction with the source for active interrogation or passively. The system uses proton recoil scintillators: 32 small $2.5 \times 2.5 \times 10.2$ -cm-thick plastic scintillators for imaging and at least two 2 \times 2 arrays of 27 \times 27 \times 10-cm-thick plastic scintillators that detect induced fission radiation. The DT generator contains an alpha detector^{7,8} that time and directionally tags a fan beam of some of the neutrons emitted and subdivides it into pixels. A fast (1 GHz) time correlation processor⁶ measures the time-dependent coincidence among all detectors in the system. A computer-controlled scanner moves the small detectors and the source appropriately for scanning a target object for imaging.

The system is based on detection of transmitted 14.1 MeV neutrons, fission neutrons, and gamma rays from spontaneous, inherent source fission of the target, fission neutrons and gamma rays induced by the external DT source, gamma rays from natural emissions of uranium and plutonium, and induced gamma-ray emission by the interaction of the 14.1 MeV neutrons from the DT source. The NMIS can and has been used with a time-tagged californium spontaneous fission source.⁹ It has also been used with pulsed interrogation sources such as LINACs,¹⁰ DT, and deuterium-deuterium (DD) sources.¹¹ This system is uniquely suited for detection of shielded highly enriched uranium (HEU), plutonium, and other special nuclear materials and detection of high explosives (HE) and chemical agents.¹²

The NMIS will be adapted to utilize a trusted processor that incorporates information barrier and authentication techniques using open software and then be useful in some international applications for materials whose characteristics may be classified. The proposed information barrier version of the NMIS system would consist of detectors and cables, the red (classified side) computer system, which processes the data, and the black (unclassified side) computer, which handles the computer interface. The system could use the "IB wrapper" concept proposed by Los Alamos National Laboratory and the software integrity (digital signatures) system proposed by Sandia.¹³ Since it is based entirely on commercially available components, the entire system, including NMIS data acquisition boards, can be built with commercial off-the-shelf components.

This system is being developed into a fieldable system (FNMIS) for potential arms control treaties by the ONV.¹⁴ The system will be modularly constructed with the RF shielded modules connected to the processor by appropriate control and signal cable in metal conduit. The FNMIS is presently being

^{*} This report is a revision and expansion of an earlier report, *NMIS with Imaging and Gamma Ray Spectrometry for Pu, HEU, HE, Chemical Agents and Drugs*, ORNL-TM-2006/76 (2006).

designed for eventual incorporation of gamma-ray spectrometry and an information barrier to protect classified information. The system hardware and software can be configured to obtain the following: plutonium presence, plutonium mass, Pu-240/239 ratio, plutonium geometry, plutonium metal vs nonmetallic (absence of metal), time (age) since processing for plutonium (or last purification), uranium presence, uranium mass, uranium enrichment, uranium geometry, uranium metal vs non-metallic compound (absence of metal), beryllium presence and mass, tritium and deuterium gas bottle presence, HE, and chemical weapons. A matrix of the quantities determined, the method of determination, whether active (external neutron source) or passive, and the measurement equipment involved is given in the Tables 1–4. Some of these attributes can be obtained by multiple data analysis methods. The gamma-ray spectrometry methods for HEU, plutonium, and HE have been developed by other laboratories, are well known, and will be incorporated.^{15, 16} This system will incorporate the gamma spectrum interpretation technology for identification of HE and chemical agents, developed for the Portable Isotopic Neutron Spectrometer of A. J. Caffrey of the Idaho National Laboratory.¹² In addition, the imaging capability allows warhead authentication and traceability of weapons part types and weapons component types through dismantlement and can be used to verify the destruction of change in form of special nuclear material, HE, and other essential non-fissile components. The imaging data will provide geometric configurations for GADRAS-like codes^{17,18} Imaging and GADRAS-like codes complement each other for material determinations since both gamma-ray spectrometry interpretations and neutron transmission depend upon materials and configuration. In addition, the data imaging measurements and MCNP-PoliMi calculations¹⁹ of the NMIS time-correlation signatures can be used to obtain fissile shape and mass without calibration. Very good initial estimates of the configuration of materials from imaging can be provided to both these codes for further refined analyses. The presence and mass of beryllium requires a time-tagged californium source that can be operated simultaneously with the imaging and the data from both the time-tagged and the DT generator separated by time correlation methods, which has been done several times in the past. System hardware and software modules may also be configured to estimate a selected subset of these attributes.

In addition, signatures for fissile material can be used for template matching such as had been implemented for confirmation of inventories and receipts for weapons components at the Y-12 National Security Complex in Oak Ridge in 1996. Y-12 personnel were trained and have been operating three NMIS systems at the Y-12 complex.

A typical measurement and analysis scenario is depicted in Fig. 1. If low- or high-energy X-ray imaging information is available, it can complement the fast-neutron imaging information. Gamma and neutron transport can be used for the offline analysis. After setup, location, and checkout of the detectors, DT generator, and data processing system, the measurement and analysis scenario steps could proceed as follows.

- 1. Perform passive gamma-ray spectrometry measurements with no object to determine background.
- 2. Perform passive NMIS time correlation measurements with no object to determine background and determine time coincidence distributions and multiplicities.
- 3. Perform baseline I_0 measurement with no object with the subsampling that will be used later for the vertical radiographic image scan and the tomographic slice image. (I_0 is the neutron flux counted when no target is present, that is, a calibration of the efficiency of the transmission detection system.)
- 4. Perform gamma-ray spectrometry measurements with the sources turned on to obtain the active background from the nearby materials.

- 5. Locate the target object appropriately and measure its location.
- 6. Perform passive gamma-ray spectrometry with object.
- 7. Perform passive time correlation measurements to see if plutonium or HEU is present.
- 8. Evaluate the passive data to make preliminary conclusions including running a GADRAS-like code.
- 9. Perform a shadowgraph (radiograph) imaging scan to determine overall shape of the object and find location of interest for more detailed imaging.
- 10. Perform detailed tomographic slice image measurements at heights of interest to determine the internal configuration. Measure at least two turntable rotary positions if possible.
- 11. Perform time-tagged gamma spectrometry at the same time as step 10 at locations of interest to assist in the identification of materials.
- 12. Reconstruct transmission data using MLEM algorithm. Use resulting images to create the initial guess for dimensions and attenuation coefficients of individual parts inside of the object.
- 13. Run TAKE to determine dimensions and attenuation coefficients of individual parts using information obtained in step 10.
- 14. Reconstruct fission site images from multiplicity singles and doubles data using MLEM algorithm.
- 15. Overlay singles and doubles reconstructions onto transmission reconstructions to identify which internal parts are composed of fissile material.
- 16. Look at the neutron scattering time-of-flight data for each pixel to identify light materials.
- 17. Look at passive and active gamma-ray spectra to determine the presence of and the relative amounts of isotopes of interest in the object. Subtraction of the passive data from the active should give the non-elastic gamma production to identify explosives and other materials.
- 18. Come up with a simple 1-D model using the results from previous steps and fit the model parameters to the passive gamma spectra using GADRAS.
- 19. Develop a MCNP model of the system based on the previous steps.
- 20. Run MCNP simulations of measured quantities (possibly including transmission, fission mapping, neutron scattering, and active gamma spectrometry).
- 21. Generate passive gamma spectrum using the GADRAS 1-D model.
- 22. Compare results of steps 20–21 with measurements and generate a goodness-of-fit value.
- 23. Check on the goodness-of-fit result for convergence. If converged, then end; otherwise, modify model dimensions and materials and return to step 20.

This system has the advantage of combining multiple technologies into a single system for a variety of applications and thus is cost-effective.

Attribute	Measurement implementation	Measurement basis	Active or passive	Measurement equipment
Identification of plutonium attributes				
Presence	Time-dependent coincidence	Detect internal spontaneous fission/multiplicities	active/passive	DT neutron source (if active), scintillation detectors, time-correlator
	Gamma spectrometry	Detect plutonium spectral lines	passive	High-resolution gamma detector, multi-channel analyzer
Age	Gamma spectrometry	Measure in-growth of ²⁴¹ Am	passive	High-resolution gamma detector, multi-channel analyzer
Metal or non-metal	Time-dependent coincidence	Measure density from neutron transmission	active	DT neutron source, scintillation detectors, time-correlator
	Time-dependent coincidence	Attenuation of gammas emitted and multiplication depending on density	active	DT neutron source, scintillation detectors, time-correlator, gamma ray detector and multichannel analyzer
	Neutron-initated gamma spectrometry	Detect 6129 keV gamma from 14.1 MeV neutron interactions with Oxygen & Florine	active	DT neutron source, high- resolution gamma detector, multi-channel analyzer
Geometry	Time-dependent coincidence	imaging	active	DT neutron source, scintillation detectors, time-correlator
Relative ²⁴⁰ Pu content	Gamma spectrometry	Compare ²⁴⁰ Pu and ²³⁹ Pu spectral lines	passive	High-resolution gamma detector, multi channel analyzer
	Time-dependent coincidence	Measure induced fissions enhanced by imaging and compare to spontaneous fissions	Active/passive	DT neutron source, scintillation detectors, time-correlator
	Time dependent coincidence	Measure time distribution of coincidences	passive	Large palastic scintillators with off-line Monte Carlo analysis
Plutonium mass	Time-dependent coincidence	Measure spontaneous fission rate	passive	Scintillation detectors, time-correlator
	Time-dependent coincidence	Transmission imaging tomography	active	DT neutron source, scintillation detectors, time-correlator
Spatial distribution	Time-dependent coincidence	Imaging plus DT source pixel correlated multiplicities for fission mapping	active	DT neutron source, scintillation detectors, time-correlator

Table 1. Attribute determination methods and measurement equipment for NMIS and FNMIS imaging with gamma spectrometry for plutonium attributes

Table 2. Attribute determination methods and measurement equipment for NMIS and FNMIS imaging with gamma spectrometry for uranium attributes

Attribute	Measurement implementation	Measurement basis	Active or passive	Measurement equipment	
Identification of uranium attributes					
Presence	Time-dependent coincidence	Detect induced fission and absence of internal spontaneous fission	active	DT neutron source, scintillation detectors, time-correlator	
	Gamma ray spectrometry	Detect 186 keV gamma for unshielded uranium	passive	Gamma ray detector, multichannel analyzer	
Metal or non-metal	Neutron-initiated gamma spectrometry	Detect 6129 keV gamma from 14.1 MeV neutron interactions with Oxygen & Florine	active	DT neutron source, high- resolution gamma detector, multi-channel analyzer	
	Time-dependent coincidence	Measure density from neutron transmission imaging	active	DT neutron source, scintillation detectors, time-correlator	
	Time-dependent coincidence	Attenuation of gamma emitted and multiplication depend on density	active	DT neutron source, scintillation detectors, time-correlator	
Geometry	Time-dependent coincidence	Imaging	active	DT neutron source, scintillation detectors, time-correlation	
²³⁵ U enrichment	Time-dependent coincidence	Compare induced fission rates and neutron transmission for simple parts	active	DT neutron source, scintillation detectors, time-correlator, HPGe	
	Gamma spectrometry corrected for imaging using field of view, volume, shielding	Gamma ray spectrometry	active	DT neutron source, scintillation detectors, time-correlator, high resolution gamma detector, multi channel analyzer	
Fissile mass	Time-dependent coincidence	Measure induced fission rate complemented by imaging and fission mapping	active	DT neutron source, scintillation detectors, time-correlator	
Spatial distribution	Time-dependent coincidence	Imaging plus DT source pixel correlated multiplicities for fission mapping	active	DT neutron source, scintillation detectors, time-correlator	

Table 3. Attribute determination methods and measurement equipment for NMIS and FNMIS imaging with gamma spectrometry for other material or component attributes

Identification of other material or component attributes					
Material or	Attributo	Measurement	Measurement	Active or	Measurement
component	Attribute	implementation	basis	passive	equipment
Beryllium	presence	Time-dependent coincidence	Transmission TOF	active	Time-tagged Cf, one plastic scintillator, time-correlator
	mass and geometry	Time-dependent coincidence	Transmission TOF	active	Same as presence with image and off- line Monte Carlo analysis
Deuterium and Tritium	presence	Time-dependent coincidence	Elastic scattering time distribution	active	DT neutron source, scintillation detectors, time correlator (Scattered peaks at a fixed angle occur at specific times)
Beryllium Deuterium and Tritium High Explosives Chemical Weapon	presence	Gamma Spectrometry	N presence and Ratios of N/C, H/C, and O/C	active	DT neutron source, HPGe or BGO
	mass and geometry	Time-dependent coincidence	Transmission imaging tomography	active	DT neutron source, scintillation detectors, time- correlator
	Separation from fissile	Time-dependent coincidence	Transmission imaging tomography	active	DT neutron source, scintillation detectors, time- correlator
	destruction of	Time-dependent coincidence	Transmission imaging tomography	active	DT neutron source, scintillation detectors, time- correlator
Chemical Weapon	presence	Time tagged gamma spectrometry	Ratios of N/C H/C, and O/C	veacti	DT neutron source, high resolution gamma ray detector or BGO

 Table 4. Attribute determination methods and measurement equipment for NMIS and FNMIS imaging with gamma spectrometry for dismantlement attributes

Identification of dismantlement attributes				
Attribute	Measurement implementation	Measurement basis	Active or passive	Measurement equipment
Warhead authentication	Time-dependent coincidence	Transmission imaging tomography and fission mapping	active	DT neutron source, scintillation detectors, time-correlator
Traceability ^a of weapons/parts	Time-dependent coincidence	Transmission imaging tomography and fission mapping	active	DT neutron source, scintillation detectors, time-correlator
Destruction of nuclear warhead parts	Time-dependent coincidence	Transmission imaging tomography and fission mapping	active	DT neutron source, scintillation detectors, time-correlator
Destruction of essential non- nuclear parts	Time-dependent coincidence	Transmission imaging tomography	active	DT neutron source, scintillation detectors, time-correlator

^{*a*}Complemented by tags and seals methods



Fig. 1. Possible measurement and analysis scenario.

REFERENCES

- J. A. Mullens, J. E. Breeding, R. B. Perez, J. T. Mihalczo, T. E. Valentine, and J. A. McEvers, "Multipurpose Processor for Arms Control and Nonproliferation and NMC&A," Institute of Nuclear Materials Management Annual Conference, July 1999.
- J. T. Mihalczo, J. K. Mattingly, J. A. Mullens, J. A. McEvers, J. S, Neal, R. B. Oberer, J. D. White, Oak Ridge Multiple Attribute System (ORMAS) for Pu, HEU, HE, Chemical Agents, and Drugs, ORNL/TM-2001/175 (2001).
- J.A. Mullens, John Neal, Sara Pozzi, and John Mihalczo, "Tomographic Image and Detector Coincidence Measurements for HEU Object Inspection," Central Chapter INMM Meeting, Oak Ridge, Tennessee, 2004.
- P. Hausladen, M. Blackston, J. A. Mullens, S. McConchie, J.Mihalczo, P. Bingham, N. Ericson, and L. Fabris, "Induced-Fission Imaging of Nuclear Material", Proceedings of the Institute of Nuclear Materials Management, Baltimore, Maryland, July 11–15, 2010.
- J. Mullens, S. McConchie, P. Hausladen, J. Mihalczo, B. Grogan, and E. Sword, "Neutron Radiography and Fission Mapping Measurements of Nuclear Materials with Varying Composition and Shielding" Proceedings of the Institute of Nuclear Materials Management, Palm Desert, California, July 17–21, 2011.
- J. T. Mihalczo, J. A. Mullens, J. K. Mattingly, and T. E. Valentine, "Physical description of nuclear materials identification system (NMIS) signatures," *Nuclear Instruments and Methods in Physics Research A* 450(2–3), 531–555 (2000).
- 7. J. S. Neal et al., "Evaluation of ZnO:Ga Alpha Particle Detectors for Use in Neutron Generators," *IEEE Nuclear Science Symposium*, Rome, Italy, October 15–22, 2004 (2004).
- 8. P.A. Hausladen, J.S. Neal, and J.T. Mihalczo, "An Alpha-particle Detector for a Portable Neutron Generator for the Nuclear Materials Identification System (NMIS)," *Nuclear Instruments and Methods in Physics Research B* **241**, 835–838 (2005).
- 9. J. T. Mihalczo, "The Use of 252Cf as a Randomly Pulsed Neutron Source for Prompt Neutron Decay Measurements," *Nucl. Sci. Eng.* **41**, 296 (1970).
- P. A. Hausladen, J. T. Mihalczo, B. W. Blackburn, S. M. Watson, J. L. Jones, A. W. Hunt, and S. Thompson, "Identifying Nuclear Material via Prompt Photo-Neutron Multiplicity Measurements," CAARI, Fort Worth, 2008.
- S. McConchie, P. Hausladen, J. Mihalczo, J. Jones, S. Watson, and B. Blackburn, "Pulsed Source Time Correlation Measurements of Shielded HEU Oxide Fuel Pins and DU," IEEE Nuclear Science Symposium, 2009.
- J. Caffrey, J. D. Cole, R. J. Gehrke, and R. C. Greenwood, "Chemical Warfare Agent and High Explosive Identification by Spectroscopy of Neutron-Induced Gamma Rays," *IEEE Transactions of Nuclear Science* 39, 1422–1426 (1992).

- D. MacArthur, M. W. Johnson, N. J. Nicholas, and R. Whiteson, "Use of Information Barriers to Protect Classified Information," Proceedings of INMM summer meeting, Naples, Florida, July 26– 30,1998 (LA-UR-98-2910).
- 14. J. E. Radle, D. E. Archer, J. A. Mullens, J. T. Mihalczo, C. L. Britton Jr, R. F. Lind, M. C. Wright, and C. R. Brittian, "Fieldable Nuclear Material Identification System (FNMIS)," INMM Meeting, 2009.
- 15. R. Gunnink et al., "MGAU: A New Analysis Code for Measuring U-235 Enrichments in Arbitrary Samples," Proceedings of the IAEA Symposium on International Safeguards, Vienna, Austria, 1994.
- S. J. Luke and D. E. Archer, "Gamma Attribute Measurements –Pu-300, Pu600, Pu900," Institute of Nuclear Materials Management, New Orleans, Louisiana, July 15–20, 2000.
- 17. D. R. Waymire and D. J. Mitchell, *GADRAS User's Manual*, Sandia National Laboratories, Albuquerque, New Mexico, 2009 [FOUO].
- D.J. Mercer, J.M. Blackadar, D.D. Dietrich, D.K. Smith, G.P. Lasche, and D.R. Waymire, "U.S. Government Capabilities to Support Analysis of Gamma Ray Data Submitted by Field Elements," INMM Annual Meeting, Phoenix, 2005.
- 19. S.A. Pozzi, E. Padovani, and M. Marseguerra, "MCNP-PoliMi: A Monte Carlo Code for Correlation Measurements," *Nuclear Instruments and Methods in Physics Research A* **513**(3), 550–558 (2003).