

BWXT



A BWXT/Bechtel Enterprise

Report No: Y/LB-16,131

**MCNP-POLIMI Evaluation of Time Dependent Coincidence
Between Detectors for Fissile Metal Vs. Oxide Determination**

**Sara A. Pozzi
Enrico Padovani*
John T. Mihalcz**

*** Department of Nuclear Engineering
Polytechnic of Milan
Via Ponzio 34/3, 20133 Milan, Italy**

**Y-12
National
Security
Complex**

**Nuclear Materials Management and
Storage Program Office**

May 23, 2002

**Prepared by the
Y-12 National Security Complex
Oak Ridge, Tennessee 37831
managed by
BWXT Y-12, L.L.C.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22800**

**MANAGED BY
BWXT Y-12, L.L.C.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY**

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MCNP-POLIMI EVALUATION OF TIME DEPENDENT COINCIDENCE BETWEEN DETECTORS FOR FISSILE METAL VS. OXIDE DETERMINATION

Sara A. Pozzi, Enrico Padovani*, John K. Mattingly, and John T. Mihalczo
Oak Ridge National Laboratory
Oak Ridge, TN 37831-6010 PO Box 2008 Ms6010

* Department of Nuclear Engineering
Polytechnic of Milan
Via Ponzio 34/3, 20133 Milan, Italy

ABSTRACT

In the past, passive Nuclear Materials Identification System (NMIS) measurements on plutonium metal shells at VNIIEF have shown the sensitivity of the acquired covariance functions to shell mass and thickness for a variety of shell thicknesses from 6 to 30 mm and masses varying from 1829 to 4468g. The technique acquires the time-dependent coincidence distribution between plastic scintillators detecting radiation from the Pu. The measurements showed the sensitivity of the acquired signature to the different spontaneous emission, attenuation, and multiplication properties of the shells.

In this work, the MCNP-POLIMI neutron and photon transport code was used to simulate passive measurements on plutonium metal and oxide. The code is a modified version of MCNP, which attempts to calculate more correctly quantities that depend on the second moment of the neutron and gamma distributions, and attempts to model detector pulses as closely as possible. MCNP-POLIMI, together with a post-processing code, can simulate all the time-dependent coincidence distributions measured by NMIS. In particular, the simulations evaluate the time-dependent coincidence distributions between detectors for plutonium samples having mass 2 and 4 kg, in metal and oxide form.

This work shows that the time-dependent coincidence distributions between two scintillators measured by NMIS can be used to distinguish metal from oxide.

INTRODUCTION

In the Nuclear Materials Identification System (NMIS), the time-dependent coincidence distribution between two or more detectors is one of the signatures acquired, with a 1 ns time resolution [1]. Previous studies and measurements have demonstrated the sensitivity of this and other related signatures to Pu fissile mass and thickness in active [2, 3] and passive [4 - 6] measurements at VNIIEF [4]. In this paper the passive time-dependent coincidence method for the assay of nuclear materials is evaluated for distinction between oxide and metal forms of Pu. This passive signature can also be obtained in an active NMIS measurement by separating out the source induced part of this signature and that from inherent source fission.

A variety of other methods are useful for distinctions of Pu oxide from metal. A few of them are: (1) enhanced singles over doubles, (2) gamma ray spectrometry detection of gamma rays from the (α ,n) reaction with oxygen [7], (3) transmission measurement with an active source [8], (4) gamma rays from inelastically scattered source neutrons (14.1 MeV from a DT generator) on

oxygen, [9] and (5) time-dependent coincidence measurements evaluated in this paper and (6) non nuclear methods such as the use of electromagnetic coils for oxide distinction.

The Monte Carlo simulation of time-dependent coincidence measurements that rely on the detection of fast neutrons and photons from fission requires that particle interactions in each history be described as closely as possible. The MCNP-POLIMI code [10, 11] was developed from the standard MCNP code to simulate each neutron-nucleus interaction as closely as possible. In particular, neutron interaction and photon production are made correlated and correct neutron and photon fission multiplicities have been implemented. At each collision, relevant information on neutron and gamma collisions is recorded, for example reaction type, target nucleus, energy deposited, and position. A post-processing code has also been developed, and can be tailored to model specific detector characteristics. These features make MCNP-POLIMI a versatile tool to simulate particle interactions and detection processes.

The paper is organized as follows: the simulation geometry is given in Section 2; Section 3 briefly describes the MCNP-POLIMI output file; finally, the results and analysis of the simulations performed for plutonium spheres and cylinders in metal and oxide form are given in Sections 4 and 5.

SIMULATION GEOMETRY

MCNP-POLIMI simulations performed for passive measurements on plutonium spheres and cylinders (cylinders with height equal to diameter, and height equal to twice the diameter were modeled). A sketch of the simulation geometry is shown in Figure 1. In this configuration the item to be inspected is placed between two fast plastic scintillation detectors. The plutonium metal samples had total mass equal to 2 and 4 kg, and a composition of 6wt% Pu-240 and 94wt% Pu-239. Plutonium metal and plutonium oxide were simulated, with densities 15.92 g/cm^3 and 11.46 g/cm^3 , respectively. The plutonium mass was the same in the metal and oxide samples. The distance between the center of the sample and the detectors was set to 25 cm.

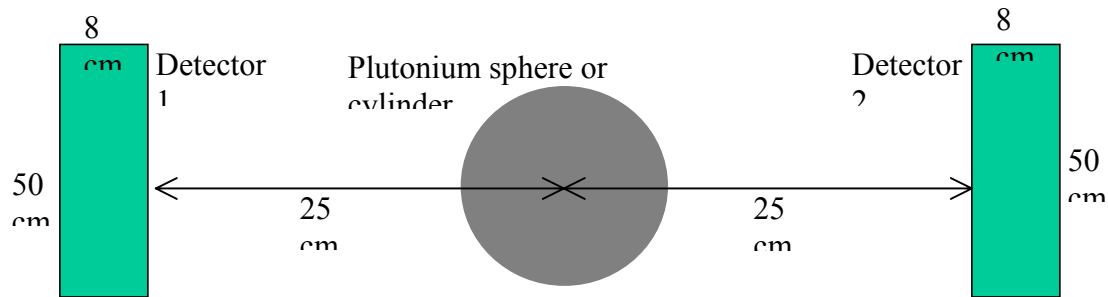


Figure 1. Top view of the geometry used in the MCNP-POLIMI simulations. Sketch not to scale.

The MCNP-POLIMI code simulates the spontaneous fission of Pu-240 by emitting neutrons and photons with correct multiplicities and energy spectra according to the appropriate distributions that can be found in literature [12-13]. The neutrons are all emitted at the time of fission (time zero). The time of photon emission in both spontaneous and induced fission takes into account the time delay of photon emission from the time of fission [14].

The MCNP-POLIMI output file consists in a detailed account of particle interactions inside the detectors. In particular, the output file contains information on each energy deposition by neutrons and photons interacting with the detectors. The data collected in the output file is post-processed using a code that takes into account the physical properties of the plastic scintillation detectors [11]. In particular, we consider the light output generated by the interactions of neutrons and photons with the hydrogen and carbon atoms of the detector, and the pulse generation time (time during which light pulses are added together to create a single impulse).

SIMULATION RESULTS

Figure 2 shows the result of the simulation for the plutonium metal cylinder, performed with $7 \cdot 10^5$ spontaneous fissions of Pu^{240} , uniformly distributed inside the cylinder. This number of spontaneous fissions occurs in approximately 9 sec in a 2 kg sample of plutonium at 6wt% Pu^{240} . All the signatures shown in the following figures have been normalized to the total number of spontaneous fissions. Therefore, the visualization of the figures does not account for the fact that in the larger samples there are more spontaneous fissions than in the smaller samples. In Figure 2, the total signature has been subdivided into four components according to the particle that generated the pulse in each of the two detectors. The possible pairs of detections are neutron-neutron, photon-photon, neutron-photon, and photon-neutron. The particles contribute to the signature on the basis of their time of flight from the fissile sample to the detectors. Because photons travel at the speed of light, they arrive on the detectors at small time lags (narrow peak centered at zero time lag).

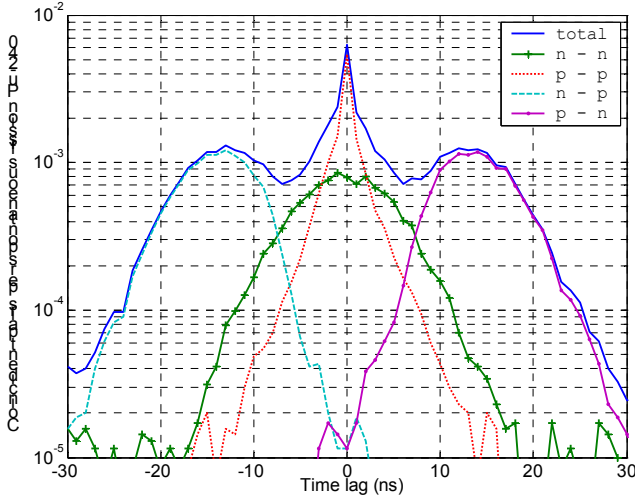


Figure 2. MCNP-POLIMI simulation of detector-detector covariance functions for 2 kg plutonium metal cylinder. The total signature is subdivided into its components given by neutron and photon contributions (neutron-neutron, photon-photon, neutron-photon, and photon-neutron).

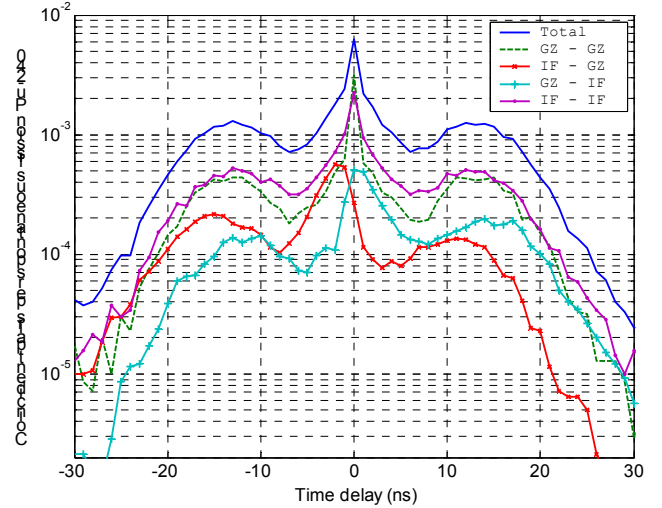


Figure 3. MCNP-POLIMI simulation of detector-detector covariance functions for 2 kg plutonium metal cylinder. The total signature is subdivided into its components given by generation zero particles (GZ) and induced fission particles (IF).

The neutron-neutron pairs are also a symmetric distribution, centered at time lag equal to zero. The speed of the neutrons depends on the plutonium fission spectrum. As a result, a broad distribution of time lags is generated. The photon-neutron and neutron-photon pairs occur when a photon reaches one detector, and a neutron, some time later, reaches the second detector.

Figure 3 shows the detector-detector covariance function for the 2 kg plutonium metal cylinder subdivided into its components according to the generation number of the particles that were detected in each of the two detectors. Generation zero particles are particles coming from the source (in this case the spontaneous fission of Pu^{240}), and particles generated by source particles in all reactions except for induced fission. Induced fission particles have generation number greater than zero, and are particles coming from fission induced inside the plutonium sample. As it can be seen, the pairs of detections given by two induced fission particles (IF – IF, line with dots in Figure 3) contribute substantially to the total signature. The pairs of detections given by generation zero particles (GZ – GZ, dashed green line in Figure 3) also contribute significantly to the total signature. Notice the difference in the shape of these two contributions: the average neutron multiplicity for the spontaneous fission of Pu^{240} is smaller than the average neutron multiplicity for Pu^{239} induced fission. Correspondingly, the total multiplicity of the particles (neutrons and photons) emitted in a spontaneous fission of Pu^{240} is smaller than the total multiplicity of particles emitted in a Pu^{239} induced fission.

The solid lines with crosses indicate the component of the signature that is given by pairs made up of one generation zero particle, and one induced fission particle (GZ – IF and IF – GZ). In this case, the originating Pu^{240} spontaneous fission generates a particle that is detected by one of the detectors, whereas the second detector detects a particle that is originated in the fission chain induced by the original Pu^{240} spontaneous fission.

Figures 4 and 5 show the result for the plutonium spheres in metal and oxide form. Figure 4 refers to the spheres having mass 2 kg, whereas Figure 5 refers to the spheres having mass 4 kg. The differences in the total signature (blue solid line with dots for the oxide sample and blue solid line for the metal sample) of the metal and oxide samples can be attributed to the signal from the induced fissions. In fact, in both cases the contribution given by generation zero particles is approximately the same. There is a slight difference in the gamma-gamma pairs at time lag zero, presumably given by the different attenuation properties of the metal and oxide samples. The difference in the contribution given by induced fission particles is in agreement with the fact that metal spheres have a greater multiplication than the oxide samples (for the 2 kg samples, the multiplication in the case of the metal is 1.99, whereas in the case of the oxide the multiplication is 1.68). Correspondingly, the total signature is greater in the case of the metal for both masses. The increase in the signature is attributed to the neutron-neutron, photon-neutron, and neutron-photon pairs. See also Figure 2.

Figure 5 shows the simulation result for the 4 kg plutonium spheres, in metal and oxide form. In this case, the difference between metal and oxide is greater than in the case of the 2 kg spheres. The multiplication is 2.64 for the metal sphere and 2.05 for the oxide sphere.

Figure 6 shows the result for the 4 kg plutonium cylinder in metal and oxide form. As it can be seen, it is possible to distinguish between the metal and oxide cylinders on the basis of the total signature. Once again the difference in the total signature is essentially given by the induced fission signal. In the case of the metal, the induced fission signal (IF – IF) is considerably greater than the signal coming from the spontaneous fission source (GZ – GZ).

ANALYSIS

Discrimination between metal and oxide plutonium can be achieved by selecting relevant features from the acquired signature. In this application, our goal was to distinguish between the metal and oxide without knowledge of the mass or geometry of the plutonium sample. The area of the neutron-neutron contribution to the covariance and the second central moment of that same distribution are two features from the signatures that are sensitive to metal vs. oxide composition. The choice of these two features is empirical, and based on the observation of the acquired signatures for all the cases considered. Figure 8 shows a plot of the area vs. the second central moment of the distribution for all the cases under investigation. As it can be seen, the oxide cases are easily distinguished from the metal cases.

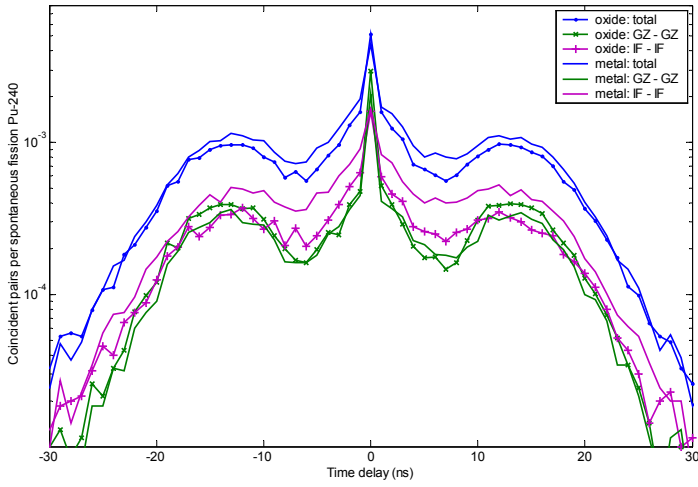


Figure 4. MCNP-POLIMI simulation of detector-detector covariance functions for 2 kg plutonium oxide and metal sphere. The total signature is shown in blue for the oxide (solid line with dots) and metal (solid line) samples. Also shown are the components given by generation zero particles (GZ-GZ) and induced fission particles (IF-IF). See also Figure 3 and description in text.

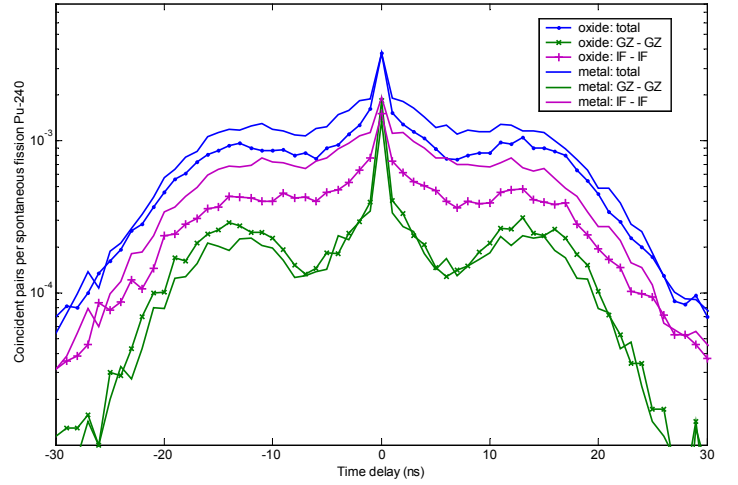


Figure 5. MCNP-POLIMI simulation of detector-detector covariance functions for 4 kg plutonium oxide and metal sphere. The total signature is shown in blue for the oxide (solid line with dots) and metal (solid line) samples. Also shown are the components given by generation zero particles (GZ-GZ) and induced fission particles (IF-IF). See also Figure 3 and description in text.

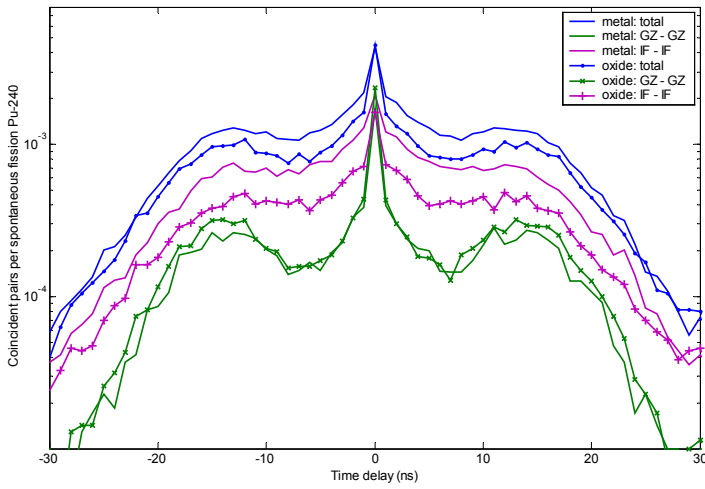


Figure 6. MCNP-POLIMI simulation of detector-detector covariance functions for 4 kg plutonium oxide and metal cylinder. The total signature is shown in blue for the oxide (solid line with dots) and metal (solid line) samples. Also shown are the components given by generation zero particles (GZ – GZ) and induced fission particles (IF – IF).

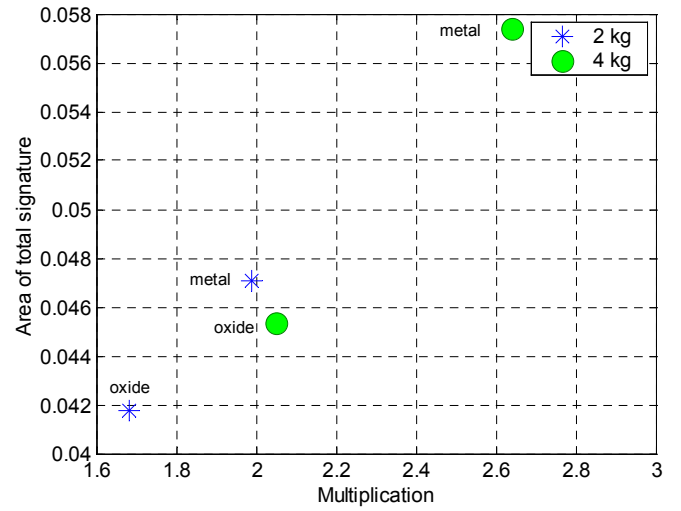


Figure 7. Area of the total signature as a function of multiplication for the plutonium cylinders of mass 2 and 4 kg.

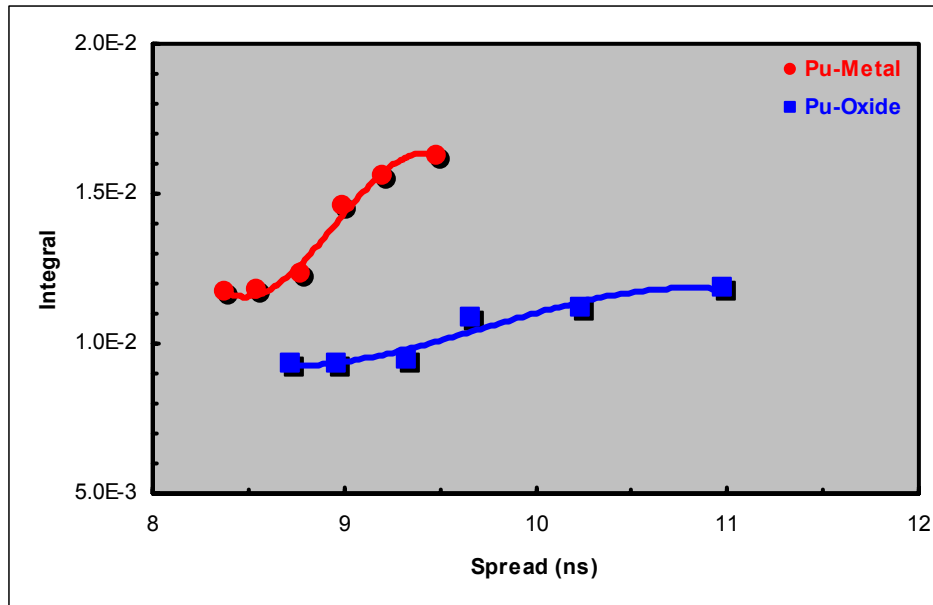


Figure 8. Integral of the neutron-neutron contribution to the signature as a function of the spread (second central moment) of the neutron-neutron contribution to the signature. Lines have been drawn to guide the eye.

CONCLUSIONS

In this paper passive time-correlation measurements of neutrons and photons from plutonium samples were evaluated for distinguishing Pu metal from oxide. Simulations performed using the MCNP-POLIMI code provided the estimate of the time-dependent coincidence distribution between detectors in a passive NMIS measurement. On the basis of two selected features of the neutron-neutron contribution to the signature, it is feasible to distinguish plutonium in its metal and oxide form, for all the spherical and cylindrical samples considered. This was done without taking into account the shape or mass of the sample in the range of cases examined. The simulations also showed that the differences in the signature increase with the multiplication in the fissile sample.

REFERENCES

1. J.T. Mihalcz, 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 Section A*, 450 (August 2000) 531.
2. S.A. Pozzi and F.J. Segovia, "²⁵²Cf-Source-Correlated Transmission Measurements and Genetic Programming for Nuclear Safeguards", accepted for publication on *Nuclear Instruments and Methods in Physics Research Section A*.
3. J.K. Mattingly, J.T. Mihalcz, L.G. Chiang, and J.S. Neal, "Preliminary Analysis of Joint RFNC-VNIIEF/ORNL Measurements Performed In Year 2000", Y/LB-16,097, Y-12 National Security Complex, September 2001.
4. J.K. Mattingly, J. Neal, J.T. Mihalcz, "NMIS Passive Time-Dependent Coincidence Measurement for Plutonium Mass and Multiplication", presented at the *Institute of Nuclear Materials Management 43rd annual meeting*, June 23-27, 2002, Orlando, Florida.
5. T.E. Valentine, L.G. Chiang, and J.T. Mihalcz, "Monte Carlo Evaluation of Passive NMIS for Assay of Plutonium in Shielded Containers", *proceedings of the Institute of Nuclear Materials Management 41st annual meeting*, July 16-20, 2000, New Orleans, Louisiana.
6. L.G. Chiang, "NMIS Time Correlations for determining the Shape of Plutonium Using Second Order Statistics" *proceedings of the Institute of Nuclear Materials Management 42nd annual meeting*, July 12-19, 2001, Indian Wells, California.
7. 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).
8. J. T. Mihalcz, J. K. Mattingly, J. S. Neal, and J. A. Mullens, "NMIS Plus Gamma Spectroscopy For Attributes of HEU, Pu, And HE Detection," *Institute of Nuclear Materials Management annual meeting*, June 23-27, 2002, Orlando, Florida.
9. A. J. Caffrey, personal communication, 2002.
10. M. Marseguerra, E. Padovani, and S.A. Pozzi, "Simulating the wrong physics can yield correct results", *proceedings of the 3rd IMACS Seminar on Monte Carlo Methods MCM2001*, September 10-14, 2001, Salzburg, Austria.
11. M. Marseguerra, E. Padovani, and S.A. Pozzi, "Use of the MCNP-POLIMI Code for Time-Correlation Safeguards Measurements", *SMORN: Symposium on Nuclear Reactor Surveillance and Diagnostics*, May 27-31, 2002, Goteborg, Sweden.

12. T.E. Valentine, "Evaluation of prompt fission gamma rays for use in simulating nuclear safeguard measurements", *Annals of Nuclear Energy* 28, February 2001, pp. 191-201.
13. T.E. Valentine, MCNP-DSP Users Manual, ORNL/TM-13334, Oak Ridge National Laboratory (1997).
14. R. Vandenbosch and J.R. Huizenga, *Nuclear Fission*, Academic Press, New York, 1973.