MONTE CARLO EVALUATION OF PASSIVE CORRELATION MEASUREMENTS ON CONTAINERIZED PLUTONIUM SHELLS

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ABSTRACT

In previous studies the use of the Nuclear Materials Identification System (NMIS) for passive measurements on plutonium metal samples was investigated. The technique is based on the detection of correlated particles from the spontaneous fission of Pu-240 and from the subsequent induced fission on Pu-239. Past measurements and analyses showed the sensitivity of the acquired signature to the emission, attenuation, and multiplication properties of the shells. In particular, measurements were performed at VNIIEF for a variety of shell thicknesses ranging from 6 to 30 mm, and masses varying from 1829 to 4468 g.

In the present paper, the effect of placing the fissile material being investigated inside a specifically designed container (AT-400R) is investigated. The container is designed to shield the radiation emitted by the fissile sample, and is therefore expected to reduce the correlated signal greatly. A number of simulations were performed with the MCNP-PoliMi code to quantify this reduction and to assess the feasibility of conducting measurements using large liquid scintillators on the plutonium samples placed inside the AT-400R container.

INTRODUCTION

Among other signatures, the Nuclear Materials Identification System (NMIS) acquires the time-dependent coincidence distribution between two or more detectors, with a 1 ns time resolution [1]. Previous studies and measurements [2-6] have demonstrated the sensitivity of this and other related signatures to fissile mass and thickness in passive measurements at the All-Russian Institute of Experimental Physics (VNIIEF).

In this study, the attenuating properties of the Russian-built AT-400R fissile storage container are evaluated. In particular, the possibility of conducting NMIS passive measurements using a pair of large liquid scintillation detectors placed on opposite sides of the container is investigated. The Monte Carlo approach is validated with experimental results with a Cf-252 source as a surrogate for plutonium. In the experiment, the Cf-252 source is placed at the center of the AT-400R container with polyethylene as the inner moderator.

MONTE CARLO SIMULATION OF PLUTONIUM SHELL

The plutonium shell (98 wt% Pu-239) in this study had an inner radius of 5.35 cm and outer radius of 6.0 cm, and mass of approximately 2.5 kg. The geometry of the MCNP-PoliMi [7] simulations is shown in Figure 1. The shell is placed at the center of the container, enclosed by moderating material. The outer region consists of polyurethane

foam. Two large plastic scintillators (50 by 50 by 8 cm) are placed on opposite sides of the AT-400R container. The detectors are sensitive to gamma rays and fast neutrons, with the detection threshold being typically set at 110 keVee (keV electron equivalent) corresponding to an energy threshold of 110 keV for the gamma rays and 0.8 MeV for the neutrons, approximately.

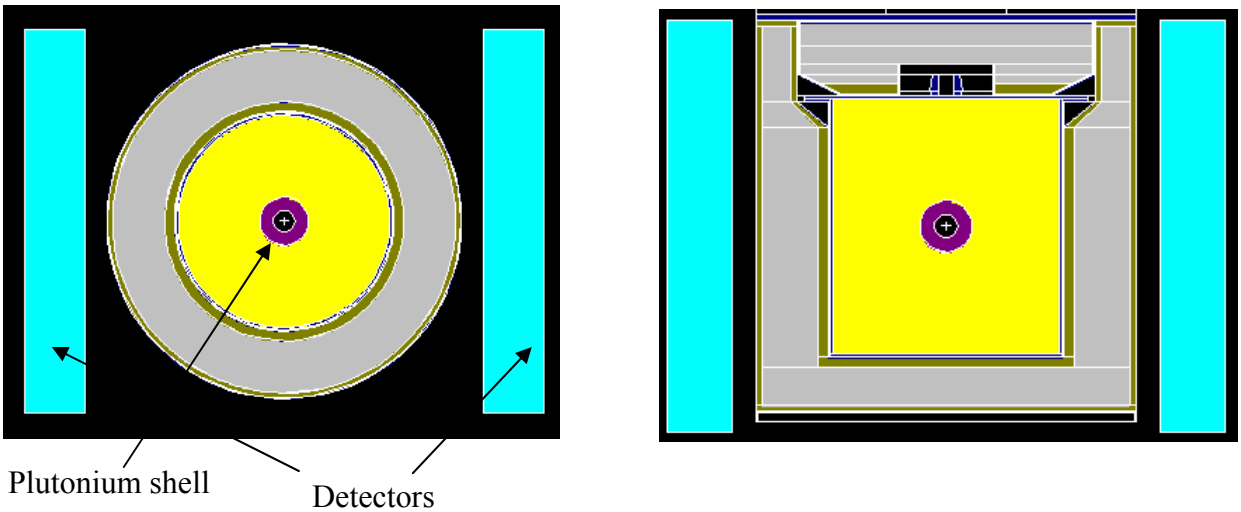

Figure 1. Schematic diagram of the MCNP-PoliMi simulations.

To evaluate the attenuating properties of the container, the simulation was performed for the bare shell and the shell placed inside the container. In both cases the number of Pu-240 spontaneous fissions modeled was equal to 10^6, uniformly distributed in the sample (1 kg of 1.77 wt% Pu-240 produces 10^6 spontaneous fissions in approximately 100 sec). The distance between the center of the shell and the face of each detector was kept constant. Figure 2 shows the cross correlation between the signals from the two detectors in the case of the bare plutonium shell, and in the case of the plutonium shell placed inside the AT-400R container. As expected, the signal from the containerized shell is attenuated by the presence of the container.

Figures 3 and 4 show the signatures given in Figure 2 subdivided into the particle types that comprise them. The possible pairs that contribute to the cross-correlation function are neutron-neutron, photon-photon, neutron-photon, and photon-neutron. Time of flight considerations can be applied to justify the time lags at which these contributions appear in the signature. The photon-photon contribution is symmetric and peaked at time lag equal to zero. Similarly, the neutron-neutron contribution is symmetric and centered at time lag zero, but has a broader distribution in time, which is consistent with the distribution in energy of the fission neutrons. The neutron-photon and photon-neutron contributions form the two distributions centered at time lag equal to 15 and -15 ns, approximately. Because the geometry is symmetric, these distributions are also

symmetric about the origin. Otherwise, e.g. a misplaced plutonium sample, the asymmetry of the measured signatures would reveal an anomaly.

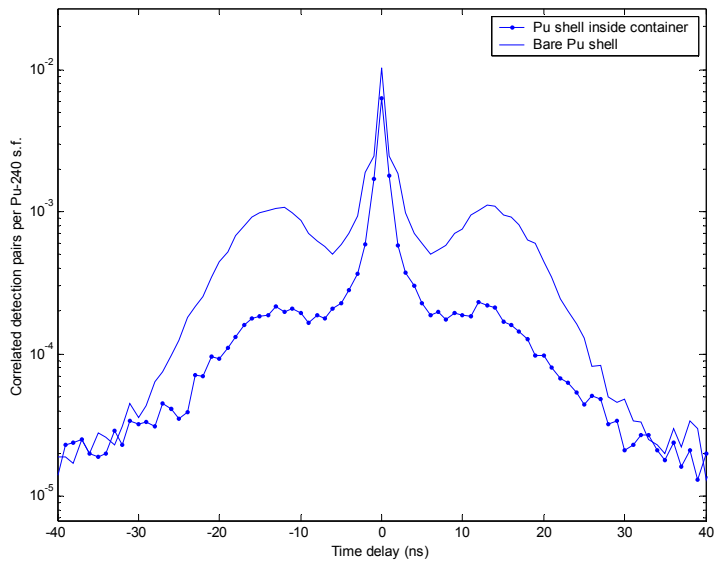

Figure 2. Cross-correlation between detectors for a plutonium shell inside the AT-400R container and a bare plutonium shell.

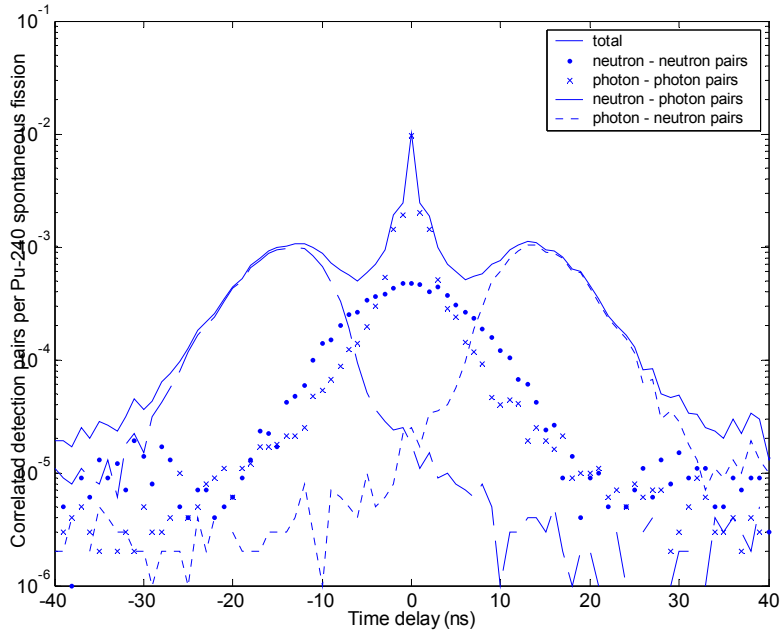

Figure 3. Cross-correlation between detectors for a bare plutonium shell, signature is subdivided into neutron-neutron, photon-photon, neutron-photon, and photon-neutron pairs.

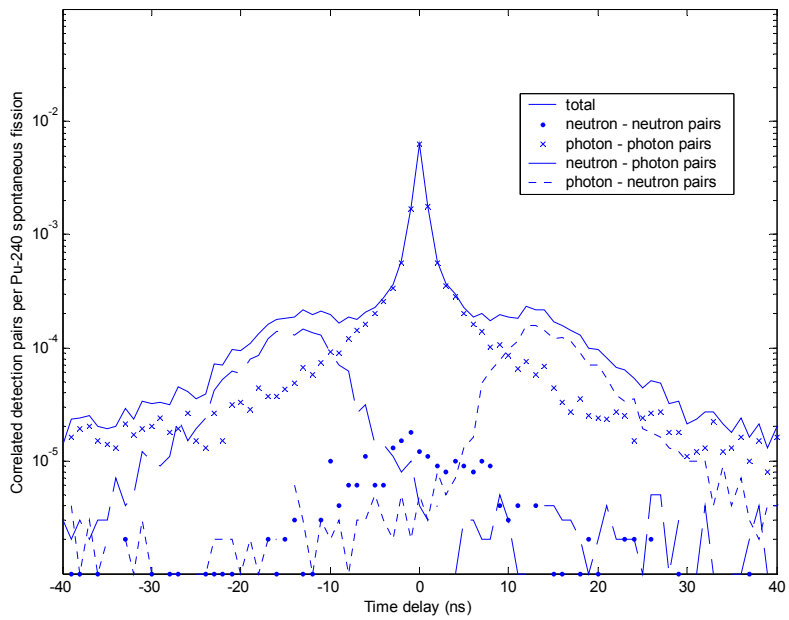

Figure 4. Cross-correlation between detectors for plutonium shell inside AT-400 container, signature is subdivided into neutron-neutron, photon-photon, neutron-photon, and photon-neutron pairs.

Comparison of Figures 3 and 4 shows that the effect of the container is to reduce the photon-photon correlations by 30%, the neutron-photon correlations by a factor 20, and the neutron-neutron correlations by a factor 50, approximately.

PLUTONIUM SHELLS OF VARYING MASS

The simulations were also performed for plutonium shells of mass varying from 1.1 to 3.6 kg, approximately [8]. The characteristics of the shells are reported in Table 1. For all samples, the composition was equal to 98 wt% Pu-239.

Table 1. Characteristics of plutonium metal shells

Radius (cm)		Mass Pu-239 (kg)
Inner	Outer	
1.4	3.15	1.1149
3.15	4.02	1.3225
4.02	4.66	1.4214
4.66	5.35	2.0376
5.35	6.0	2.4666
6.0	6.75	3.5918

Figure 5 shows the total number of correlated detections in the two detectors as a function of the sample's Pu-239 mass. The data were fit to a quadratic equation

$$N = 5.4 \cdot 10^{-5} m^2 + 0.48m - 1.5 \cdot 10^2 \tag{1}$$

where N is the number of correlated counts per second, and *m* is the mass of Pu-239. The mass of Pu-239 can be determined if the number of correlated counts is known for a known isotopic composition, and for the range of plutonium shells under analysis.

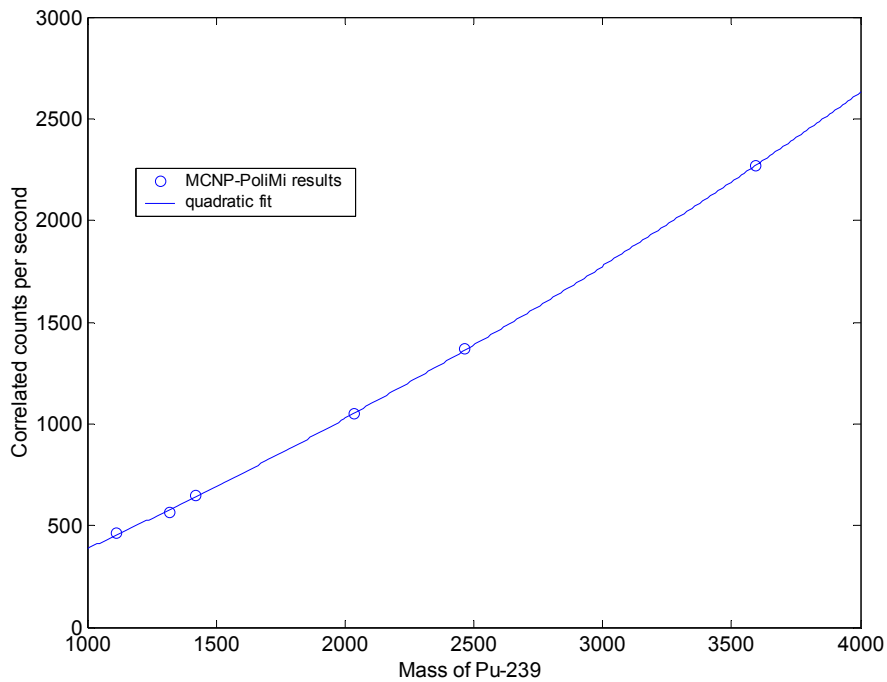

Figure 5. Total correlated counts as a function of Pu-239 mass for containerized plutonium shells.

The use of liquid scintillators in place of the plastic scintillators leads to the possibility of subdividing the signature on the basis of the particle type that generated the detection, as shown in Figures 3 and 4. The additional information can be used to select features from the signatures that are more sensitive to the attributes of the fissile samples that are to be determined. As an example, consider feature F, defined as the product of the neutron-neutron contribution to the cross-correlation (*nn*) and the photon-photon contribution to the cross-correlation (*pp*)

$$F = nn * pp \tag{2}$$

This feature as a function of Pu-239 mass is shown in Figure 6.

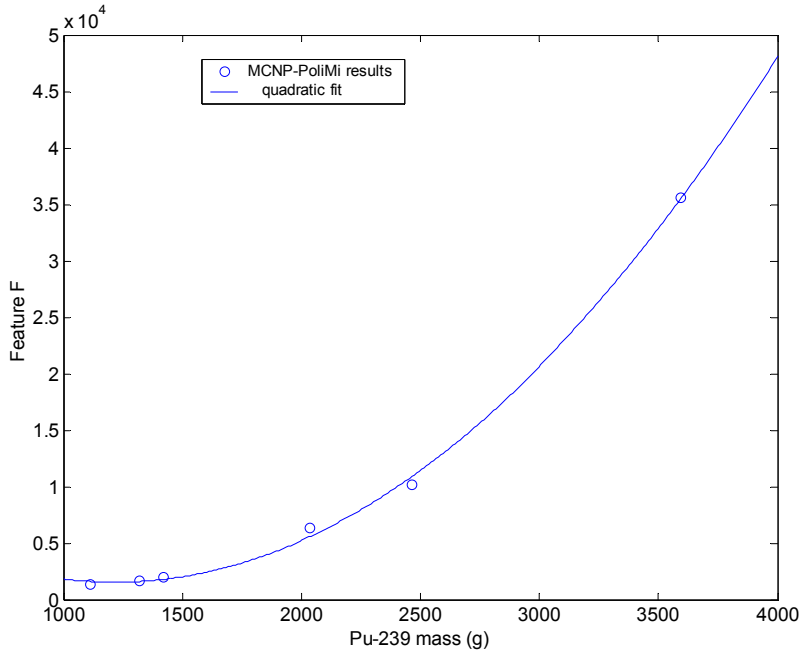

Figure 6. Feature F (defined in Eq. 2) as a function of Pu-239 mass for containerized plutonium shells.

The sensitivity of feature F to Pu-239 mass is approximately a factor of 3.6 greater than that of the total counts shown in Figure 5. Feature F is more sensitive to the upper mass range for the plutonium samples considered. Other features with higher sensitivity could be determined by conventional or artificial intelligence methods [9].

EXPERIMENTAL RESULTS

A series of experiments were performed with NMIS at Oak Ridge National Laboratory. Two arrays of large liquid scintillators and an AT-400R container were placed in the configuration given in Figure 1. The available container did not have the inner layers of moderating material, so a polyethylene cylindrical annulus having inner radius 2.54 cm and outer radius 10.16 cm was used. A Cf-252 spontaneous fission source was used in place of the plutonium sample, and was centered inside the container. The same measurement was performed with the Cf-252 source without the container. The results are shown in Figure 7.

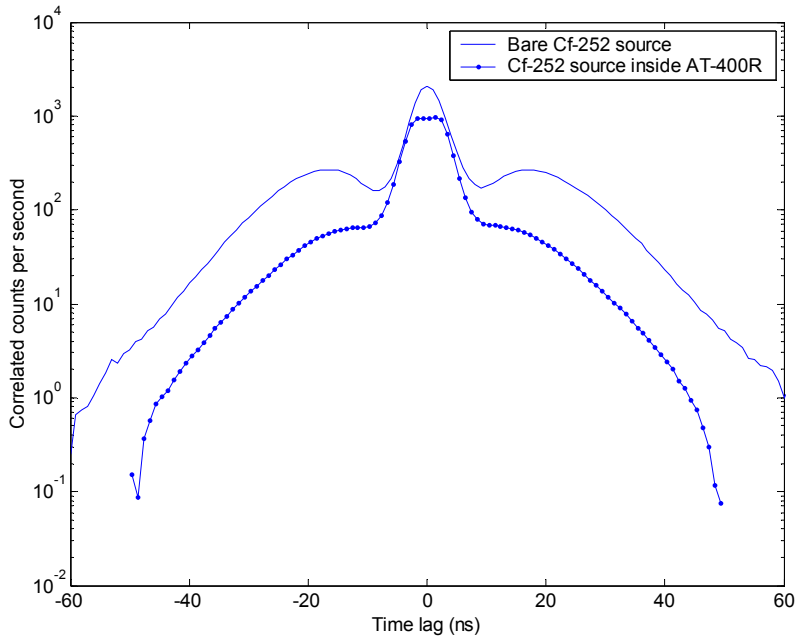

Figure 7. Measured cross-correlation between detectors for Cf-252 source outside and inside an AT-400R container.

The photon-photon peak is attenuated by a factor 2.2, and the photon-neutron peaks by a factor 10, approximately, by the presence of the container. Comparison of Figures 2 and 7 shows that the experimental results have a lower resolution than the Monte Carlo simulations: the photon-photon peak has a full width at tenth maximum of 14 ns and 5 ns in the experiment and simulation, respectively. A possible explanation of the lower resolution of the experimental result is in the process of light collection in the large liquid scintillators. Work is in progress to evaluate this effect.

CONCLUSIONS

In this paper we presented the results of a Monte Carlo analysis of passive measurements on containerized plutonium metal shells. The simulations showed that the AT-400R container effectively attenuates neutrons from the spontaneous and induced fission in the plutonium sample. Gamma rays more readily escape the container. This was confirmed by the experimental data acquired with NMIS using a Cf-252 source as a surrogate for plutonium. The measurement time required to acquire 10,000 coincidences in the photon-photon peak of the cross-correlations is less than one minute for the lowest mass plutonium shell in the AT-400R container. Combinations of neutron and gamma ray features from the cross-correlations were related to the mass of the shells, suggesting that an empirical calibration could be used to determine plutonium mass.

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