

MCNP-POLIMI SIMULATION OF ACTIVE AND PASSIVE MEASUREMENTS ON URANIUM AND PLUTONIUM OBJECTS

Sara A. Pozzi, John K. Mattingly, John T. Mihalczo, Enrico Padovani*
Oak Ridge National Laboratory
P.O. Box 2008, MS-6010, Oak Ridge, TN 37831-6010

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

ABSTRACT

This paper presents the validation of the Monte Carlo code MCNP-PoliMi for the simulation of nuclear safeguards experiments with plutonium and uranium metal based on fast time-correlation measurements. A comparison is presented between experimental data acquired with the Nuclear Materials Identification System and the Monte Carlo simulations. The measurements and simulations were performed for assemblies of delta-phase plutonium metal shells of varying inner and outer diameter, in both passive and active mode, and for a highly enriched uranium annular metal casting in active mode. The simulation results are generally in good agreement with the measurement. The areas of partial disagreement with the measured data are discussed.

INTRODUCTION

Monte Carlo codes are frequently used to optimize the design and to interpret the results of measurement systems using neutron and photon analysis for nuclear nonproliferation and safeguards applications [1-2]. This work presents the validation of the MCNP-PoliMi code [3] for the simulation of correlation measurements. In particular, we consider experiments performed with the Nuclear Materials Identification System (NMIS) [4]. The technique is based on the time-correlation measurement of neutrons and gamma rays from fission on a nanosecond time scale. The validation of MCNP-PoliMi presented in this work concerns experiments on highly enriched uranium metal castings and plutonium metal shells (delta phase, 98% Pu-239). The measurements on uranium were performed in active mode, with an interrogating Cf-252 source, whereas the measurements on plutonium were performed both in active and passive mode, i.e., both with and without an interrogation source. The simulation results for the active measurements are presented, and are generally in good agreement with the experimental results. Explanations for the areas of partial disagreement with the measured data are discussed. Relevant features of the Monte Carlo code and its post-processor are discussed, for example the ability to distinguish neutron from photon detections, and to distinguish detections of source particles from induced fission particles. The latter capability is of interest in other nuclear safeguards methodologies that make use of interrogation sources to induce fission.

The capability of simulating measurements with good approximation leads to the use of the code to generate a large number of cases, for the design and analysis of the safeguards experiments. In particular, these test cases could be used in conjunction with conventional or artificial intelligence

methods to solve the inverse problem: i.e., determine the quantities of interest (fissile mass and enrichment, for example) on the basis of features extracted from the time-correlation functions [5-6].

MCNP-POLIMI OUTPUT AND POST-PROCESSING CODE

The experimental data available for the code validation discussed in this paper consist of active measurements performed with NMIS on fissile samples. In active measurements, performed on both plutonium and uranium, an external Cf-252 source was used to induce fission in the fissile isotopes. The Cf-252 source was placed inside an ionization chamber, which provides the trigger pulse for the correlation measurements. MCNP-PoliMi models the spontaneous fission of Cf-252 by emitting neutrons and gamma rays at essentially the same time. The number of neutrons and gamma rays emitted are sampled from the appropriate distributions. The simulation output is a detailed description of the interactions occurring within the detector. The output is then post-processed with a specifically designed code to obtain the detector response [1].

MCNP-POLIMI SIMULATIONS FOR MEASUREMENTS ON PLUTONIUM

In June and July 2000, a series of measurements on assemblies of plutonium metal were performed at the Russian Federal Nuclear Center, All-Russia Scientific Research Institute of Experimental Physics (RFNC-VNIIEF) in Sarov, Russia [7-10]. The experiments were performed jointly by personnel from VNIIEF and Oak Ridge National Laboratory. The mass and dimensions of some of the delta-phase nickel-plated plutonium spherical shells are listed in Table 1. Further details on the composition of the samples can be found in Reference 9, in which delayed critical experiments with various assemblies of shells are benchmarked.

Table 1. Properties of plutonium spherical shells

Mass (g)	Outer radius (mm)	Inner radius (mm)	System ID
4468.3	46.6	31.5	<i>Pu3</i>
4004.4	60.0	53.5	<i>Pu4</i>
3316.1	53.5	46.6	<i>Pu7</i>

The experiments were performed on eight plutonium metal fissile assemblies (designated *Pu1* to *Pu8*). A previous analysis [8] showed that it is possible to obtain the mass and radial thickness of the sample on the basis of features extracted from the correlation functions measured with the Nuclear Materials Identification System (NMIS).

The simulations were performed for the three shells given in Table 1. In the measurement configuration, the plutonium spherical shell was placed between the Cf-252 instrumented source and two plastic scintillators. The distance between the Cf-252 source and the face of the detectors was set to 19.8 cm. The floor and closest wall of the room where the measurement was performed were also modeled. Details of the experimental setup can be found in Reference 7.

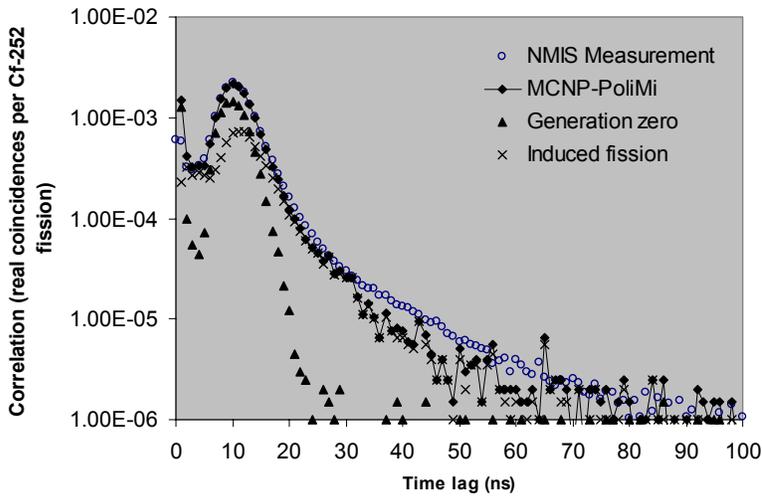
Figure 1(a) shows the comparison of the measured cross-correlation between the instrumented source and detector 1 for the 4.4 kg plutonium shell (*Pu3*), with the corresponding MCNP-PoliMi

simulation. Both measured and simulated data were normalized to the number of Cf-252 fissions. The simulation output was post-processed using appropriate parameters that are related to the detector response [2]. The signature consists of two peaks: the first peak is given by Cf-252 source gamma rays, which travel to the detector at the speed of light, and give a contribution at time lag 0.6 ns, approximately; the second peak is given by Cf-252 source neutrons, which have a broad distribution of energies, and by neutrons and gamma rays from reactions occurring in the fissile sample. As it can be seen, there is generally good agreement between the measurement and the simulation. The agreement is very good until time lags of approximately 30 ns. At greater time lags, the simulated curve underestimates the measured curve.

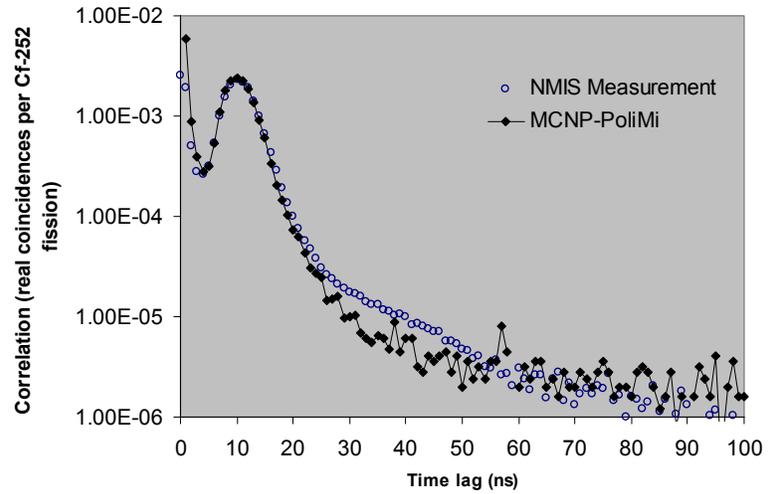
A possible explanation of the difference in the tail of the second peak is the presence of materials not modeled in the simulation in the laboratory where the experiment was performed; for example, the apparatus used to hold the source, sample and detectors, and the detector photomultiplier tube. The presence of this material could contribute to augment the experimental signature via neutron scattering. A second possible explanation is in the choice of the parameters used in the post-processing code. A previous study has shown that the calculated signature is very sensitive to the settings of these parameters [2]. In particular, this is true for neutron energies close to the detection threshold. Ideally, every detector used in the measurements should be calibrated and its calibration curve used in the post-processing of the Monte Carlo output. In this case, a calibration performed on plastic scintillators of different size than the ones present in these measurements was used in the post-processing code¹.

A feature of the post-processing code allows us to distinguish between the particles from the interrogating source (Cf-252 spontaneous fission), and the particles from the fission induced inside the sample (for the most part Pu-239 induced fission). In our nomenclature, “generation zero” particles are source particles that reach the detector uncollided and source particles that have interacted by all reactions except for nuclear fission, whereas “induced fission” particles are originated by fissions induced inside the uranium metal casting. Figure 1(a) shows the simulation result subdivided into these two components: the particles coming from the Cf-252 spontaneous fission and the particles from induced fission. As it can be seen, the first part of the signature is given for the most part by particles from the Cf-252 spontaneous fission (time lags 0 to 18 ns, approximately). At time lag 18 ns, the two components have approximately the same intensity, and at greater time lags the signal from the induced fission particles is predominant. This feature of the code allows the user to evaluate the ability of a given interrogation source to induce fission inside the sample to be analyzed. An example of this application can be found in Reference 12. Figure 1 (a-c) shows the comparison between the experiment and the simulation result for the active measurements on plutonium metal shells *Pu3*, *Pu4* and *Pu7*, respectively. In all cases the agreement is very good for delays up to 20 ns, approximately. Then, the simulated signature for the tail of the second peak is lower than the measurement. Figure 1 (d) shows the comparison for passive measurement on shell *Pu4*. Table 2 shows the percentage error for the areas of the two peaks for the cases discussed in this section, together with the root-mean-square error. The RMS percentage error for the simulated curves is 6.47% in the case of the first peak and 2.36% in the case of the second peak.

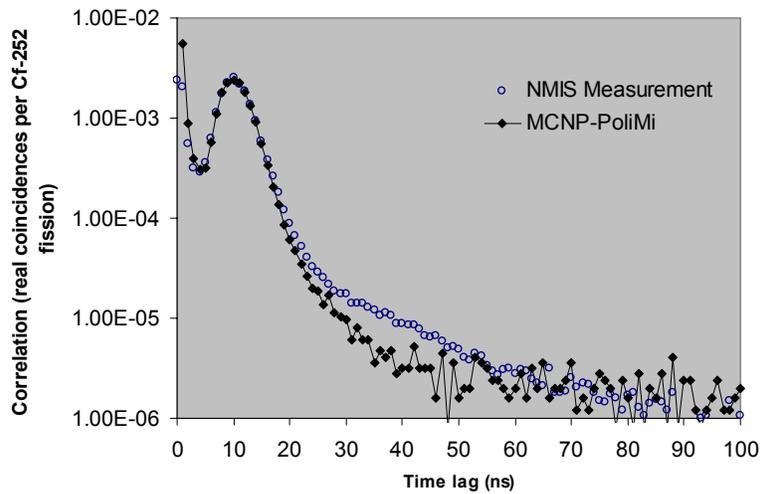
¹ 3 by 3 by 3 inch detectors were used to find the calibration curves given in Section 2. The present experiment was performed with 4 by 4 by 4 inch detectors.



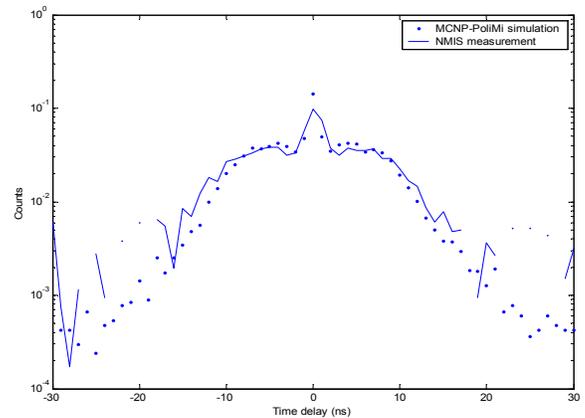
(a)



(b)



(c)



(d)

Figure 1. Source-detector cross-correlation for plutonium shell active measurements (a) *Pu3* (b) *Pu4* (c) *Pu7*. (d) Detector-detector cross-correlation for passive measurement on shell *Pu4*.

Figure 1(d) shows the comparison of the measured and simulated signature of detector pair 1-2 for plutonium sample *Pu4*. Both curves were normalized to their total area. The narrow peak about the origin is due to gamma pairs, which reach the detectors at essentially the same time. Most gamma

pairs are originated by Pu-240 spontaneous fission and Pu-239 induced fission². The underlying broader peak is due to gamma-neutron pairs and neutron-neutron pairs, also originated most probably by fission.

Table 2. Percent error in areas of peaks for MCNP-PoliMi simulation of active measurements

System ID	First peak (% error)	Second peak (% error)
<i>Pu3</i>	+2.5	+0.9
<i>Pu4</i>	+9.2	+0.8
<i>Pu7</i>	+5.9	-3.9
RMS	6.47	2.36

MCNP-POLIMI FOR THE SIMULATION OF ACTIVE MEASUREMENTS ON HIGHLY ENRICHED URANIUM METAL

In 1998, a series of measurements on uranium metal annular castings were performed at what is currently the Y-12 National Security Complex [13]. In these measurements, the Nuclear Materials Identification System (NMIS) was used in active mode, with a Cf-252 ionization chamber providing the trigger pulse.

In the experimental setup, the uranium metal casting was placed between a Cf-252 interrogation source and four plastic scintillators. The detectors, having dimensions 69.85 by 69.85 by 101.6 mm, were arranged in a 2 by 2 array. The detectors were surrounded by a 6.35 mm thick lead shield. The measurements were performed on a 6.34 mm thick carbon steel table top, approximately one meter from the concrete floor. The floor and the table were included in the MCNP-PoliMi simulation. Further details of the experimental setup can be found in Reference 13.

Many signatures were acquired in the measurements. Of these, the ones used for the code validation are the cross-correlation functions between the source and detector 1, and the cross-correlation between detectors 1 and 2. Table 3 contains the properties of the uranium metal casting.

Table 3. Properties of the uranium metal casting

Total mass (kg)	Outer diameter (mm)	Inner diameter (mm)	Height (mm)	U-235 enrichment (wt%)
~18.6	127	107.95	152.4	93.15

² Pu-240 spontaneous fissions are not a factor in the Cf-252 source and detector cross-correlations because they are not correlated with the source.

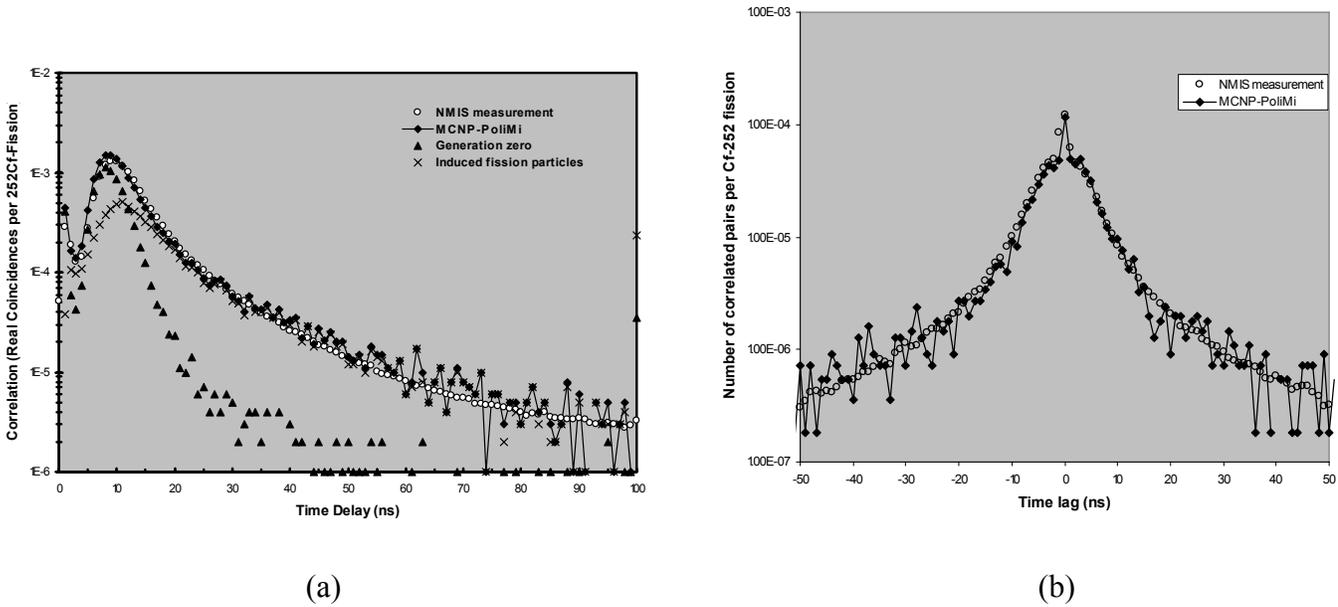


Figure 2. Uranium metal casting MCNP-PoliMi simulations (a) source – detector 1 cross-correlation subdivided into its generation zero component and induced fission particles component (b) detector 1 – detector 2 cross-correlation.

The measured and simulated source-detector cross-correlation functions are shown in Figure 2. As it can be seen, there is generally very good agreement between the MCNP-PoliMi simulation and the measurement. Using a feature of the post-processing code discussed in Section 3.3, we subdivided the signature into Cf-252 source particles (generation zero) and particles from induced fission. As it can be seen in Figure 2, the Monte Carlo simulation shows that at low time lags (up to time lag 12 ns, approximately) the signature is mostly due to generation zero particles. Starting at time lag equal to 20 ns, approximately, the tail of the signature is composed almost entirely of induced fission particles. The agreement between the measurement and the simulation is good.

Table 4 reports the percentage error for the prediction of the area of the first and second peak in the case of cross-correlation between the source and detector 1.

Table 4. Percent error in the MCNP-PoliMi simulation of measurements: areas of neutron and gamma peaks

Cross-correlation	First peak (% error)	Second peak (% error)
Source – detector 1	+0.5	+6.8

The cross-correlation between detector 1 and detector 2 was also simulated. Figure 2(b) shows the result. There is generally very good agreement between the simulation and the measurement. The

simulation does not take into account the possibility of particles generated in the uranium casting, which are uncorrelated with the initiating Cf-252 fissions. Examples of such particles are gamma rays from the decay of uranium. In particular, uranium-238 (in equilibrium with its short-life progeny Th-234 and Pa-234) decays emitting a gamma ray of energy 1 MeV, approximately. These particles could give correlated pulses in the detector as a result of cross-talk [14]. This consideration might explain the slight disagreement in the gamma ray region of the correlation: it appears that the simulated signature underestimates the measurement in this region. The error in the total area, reported in Table 5, might be reduced accordingly.

Even if the statistics are not good enough to give a definitive indication, in the region at greater time lags the simulated signature appears to be slightly higher than the experimental value. This disagreement is similar to the one outlined in the source-detector cross-correlation. A possible common explanation is in the Monte Carlo generation of prompt and delayed photons from fission. Experimental data on the time of emission of photons during fission is scarce. The implemented algorithm was based on sampling from a cumulative distribution drawn from a graph [15] and assumed valid both for Cf-252 spontaneous fissions and U-235 fissions. It is possible that this algorithm is not accurate enough, so that the resulting errors are not negligible; if this is the case, it must be revised. Work is in progress on this issue.

Table 5. Percent error in the MCNP-PoliMi simulation of measurements: total area

Cross-correlation	Total area (% error)
<i>Detector 1 – detector 2</i>	-6.3

CONCLUSIONS

This paper validated the MCNP-PoliMi code for the simulation of nuclear safeguards measurements on plutonium and uranium metal samples. The simulated cross-correlation functions were compared to the experimental data acquired with the Nuclear Materials Identification System on assemblies of plutonium shells (δ phase, 98% ^{239}Pu) of varying mass, and highly enriched uranium metal castings. Generally, there was good agreement between the simulated and measured signatures. In active measurements on plutonium the area of the first and second peaks of the cross-correlation had an RMS percentage error of 6.5% and 2.4%, respectively. The presence of additional scattering material in the room, which was not modeled in the simulation, might explain the disagreement at long times.

In the simulations of the uranium casting measurements, the error in the prediction of the area of the cross-correlation function was 0.5% for the first peak and -6.8% for the second peak. The error in the prediction of the total area of the cross-correlation between detectors was -6.3%.

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