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VALIDATION OF THE MCNP-POLIMI CODE FOR THE SIMULATION OF NUCLEAR SAFEGUARDS EXPERIMENTS ON URANIUM AND PLUTONIUM METAL

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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. We present the simulation results, generally in good agreement with the measurements, and attempt to give an explanation for the areas of partial disagreement with the measured data.

Key Words: Monte Carlo simulation, nuclear safeguards, plastic scintillator, detection efficiency, time correlation measurements

1. INTRODUCTION

Nuclear safeguards efforts aim at determining the content of fissile material enclosed in sealed containers to prevent illicit diversion. The measurement system we consider here is the Nuclear Materials Identification System (NMIS), which is based on the detection of correlated particles from fission, and makes use of plastic scintillating detectors [1]. The design and analysis of the experiments require the use of Monte Carlo codes to simulate the behavior of neutrons and photons interacting with the fissile material, the detectors, and the surrounding environment.

The MCNP-PoliMi code was developed to improve the simulation of the above-mentioned correlation measurements [2-3]. In the standard MCNP code, photons produced by neutron collisions are sampled independently from the type of neutron collision. For instance, it is possible for a neutron in a collision with uranium to generate fission photons in the course of an elastic scattering. This technique provides correct answers when we consider the average outcome of a large number of histories but, as explicitly stated by the authors of MCNP, it fails when considering the single history [4]. This is a severe drawback in the simulation of correlation measurements in which the experimental quantity of interest is the delay between particles emitted by a single event or fission chain. The MCNP-PoliMi code restores the correlation between neutron interaction and the corresponding photon production.

The MCNP-PoliMi output consists of a detailed account of the interactions of neutrons and photons in user-specified cells (typically detectors). This information can then be post-processed to simulate the response of a variety of detectors. In this paper, we are interested in the plastic scintillators used in the NMIS system. Plastic scintillators are widely used for the detection of fast neutrons and photons. The analysis of detector response relies on the use of Monte Carlo codes for simulating the physics of the response. Here, a relevant issue is that the codes often make some approximations on the physics of the detection process. With MCNP-PoliMi and its post-processor, it is possible to simulate the single interactions inside the detector, the light output generation, and, finally, the pulse formation.

Previous studies [2-3] describe the validation of this approach for time-of-flight measurements in air performed with plastic scintillation detectors. Those works aimed at determining the detector response to neutrons and gamma rays from a Cf-252 source. The present study extends the validation of the code to time-dependent coincidence distributions measurements with fissile samples. In particular, we compared correlation measurements, performed in passive and active mode on bare, spherical plutonium assemblies and in active mode on a highly enriched uranium metal casting, with simulations.

The capability of simulating measurements with good approximation leads to the use of the code to generate a large number of cases, useful in 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].

2. MCNP-POLIMI OUTPUT AND POST-PROCESSING CODE

The experimental data available for the code validation discussed in this paper consist in passive and active measurements performed with NMIS on fissile samples. Passive measurements were performed on plutonium metal assemblies. The spontaneous fission of Pu-240 present in the fissile samples provides an inherent source of correlated neutrons and gamma rays that can be measured by placing the plastic scintillators around the fissile assembly. In active measurements, performed on both plutonium and uranium, an external Cf-252 source was used to induce fission on 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 Pu-240 (passive measurements) and Cf-252 (active measurements) by emitting neutrons and gamma rays at essentially the same time. Neutron and gamma ray multiplicities are sampled from the appropriate distributions.

2.1. Post-processing Code

Relevant information on the collision events by neutrons and gamma rays inside the detectors is recorded in an output file, which is processed with a dedicated post-processing code. The code takes into account the response of the plastic scintillation detectors used in these measurements (Bicron BC 420). A linear fit was used to relate the scintillation light output to the energy deposition by photons and neutrons in the scintillator. The fit was determined by experimental calibration. In the case of photons the relationship is

$$\text{electron light output} = 0.008 + 0.99 * E_e, \tag{1}$$

whereas for neutrons

$$\text{proton light output} = -0.0685 + 0.23 * E_p \quad (0.3<E_p<3 \text{ MeV}), \tag{2}$$

where E_e and E_p are the energies of the recoil electron and proton, respectively, in MeV. The light output is expressed in terms of MeVee (MeV electron equivalent). In the case of neutrons, the linear relationship given by equation (2) is a reasonable approximation in the range $0.3<E_p<3$ MeV, approximately. For greater energies, the light output deviates considerably from (2) and other fits, such as the Birks equation, should be used [7].

2.2 Parameters Used in the Post-processing Code for Measurements on Plutonium

The neutron energy threshold for detectors 1 and 2 was determined by a linear fit of the rising edge of the efficiency curve shown in Fig. 1 (the fit was performed for efficiency from 5 to 50 %).

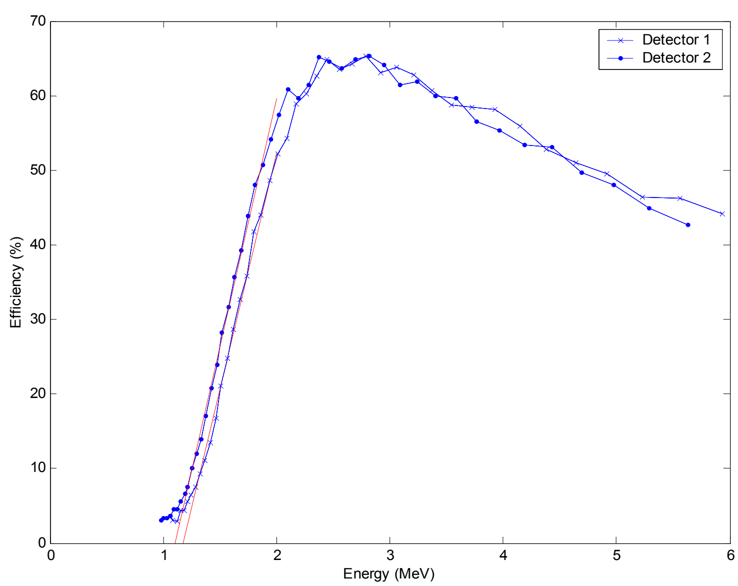

Figure 1. Experimental neutron detector efficiency for detectors 1 and 2 used in the measurements on plutonium.

The efficiency curve was measured with a time-of-flight technique using the instrumented Cf-252 source placed at a one meter distance from the detectors. The neutron energy threshold was then converted into a light output threshold by using Equation 2.

The post-processing code makes use Equations (1) and (2), together with the parameters listed in Table I. These parameters were used in the simulation of plutonium samples discussed in Section 3 of this paper. An analogous procedure was used to determine the detector energy threshold for the measurements on uranium metal discussed in Section 4 of this paper.

Table I. Parameters used in the post-processing code for the measurements on plutonium.

	Dead time (ns)	Pulse generation time (ns)	Neutron energy threshold (MeV)	Light output threshold (MeVee)
Detector 1	20	10	1.17	0.201
Detector 2	20	10	1.11	0.187

3. MCNP-POLIMI SIMULATIONS FOR PASSIVE AND ACTIVE 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 [8-11]. The experiments were performed jointly by personnel from VNIIEF and Oak Ridge National Laboratory.

3.1 MCNP-PoliMi Simulation of Passive Measurements on Plutonium

The properties of the delta-phase nickel-plated plutonium spherical shells are listed in Table II. Further details on the composition of the samples can be found in Reference 10, in which delayed critical experiments with various assemblies of shells are benchmarked.

Table II. Properties of plutonium spherical shells.

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

The experiments were performed on eight plutonium metal fissile assemblies (designated *Pu1* to *Pu8*). A previous analysis [9] 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).

Simulations were performed for three measurements similar to that shown in Fig. 2. In this configuration, the plutonium spherical shell was placed at the center of four detectors, placed with opposite pairs facing each other. The large detectors had size 15.2 by 15.2 by 10.2 cm, whereas the small detectors had size 10.2 by 10.2 by 10.2 cm. The distance between the center of the plutonium shell and the face of each detector was equal to 12.8 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 8.

The signatures acquired in the measurements and chosen for the code validation are the cross-correlation functions of the signals from detectors 1 and 2.

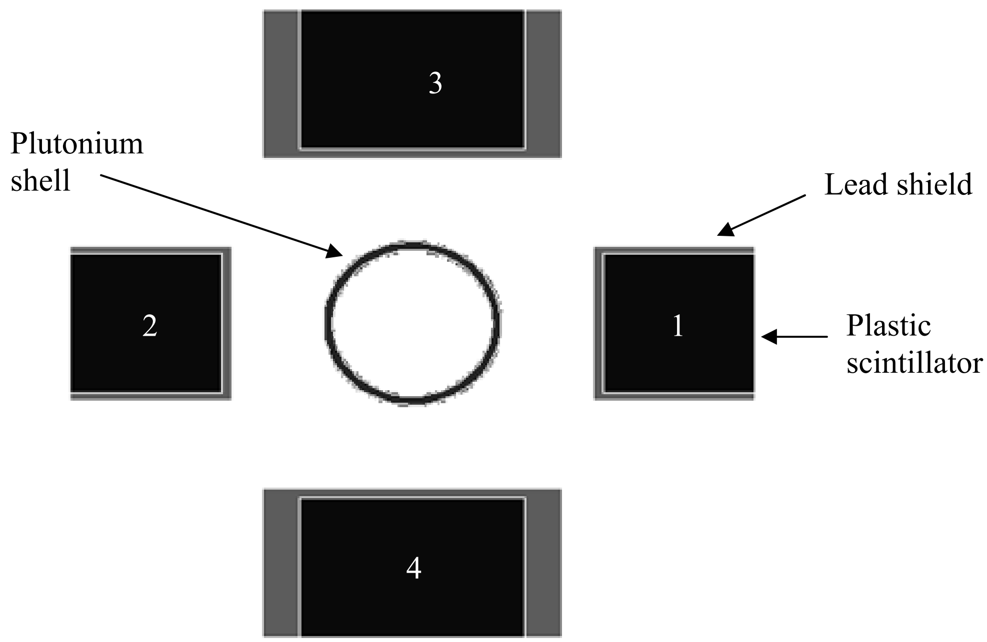

Figure 2. Sketch of the geometry of the MCNP-PoliMi simulations at the horizontal midplane. Plutonium spherical assembly in the center and four detectors (labeled 1-4) placed at 90 degree intervals. Not shown, floor and wall.

Figs. 3 and 4 show the comparison of the measured and simulated signature of detector pair 1-2 for plutonium samples *Pu7* and *Pu4,* respectively. 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 fission. The underlying broader peak is due to gamma-neutron pairs and neutron-neutron pairs, also originated most probably by fission[1].

[1] Other reactions could result in the production of correlated neutrons and gamma rays. Examples are neutron inelastic scattering and (n,xn) reactions.

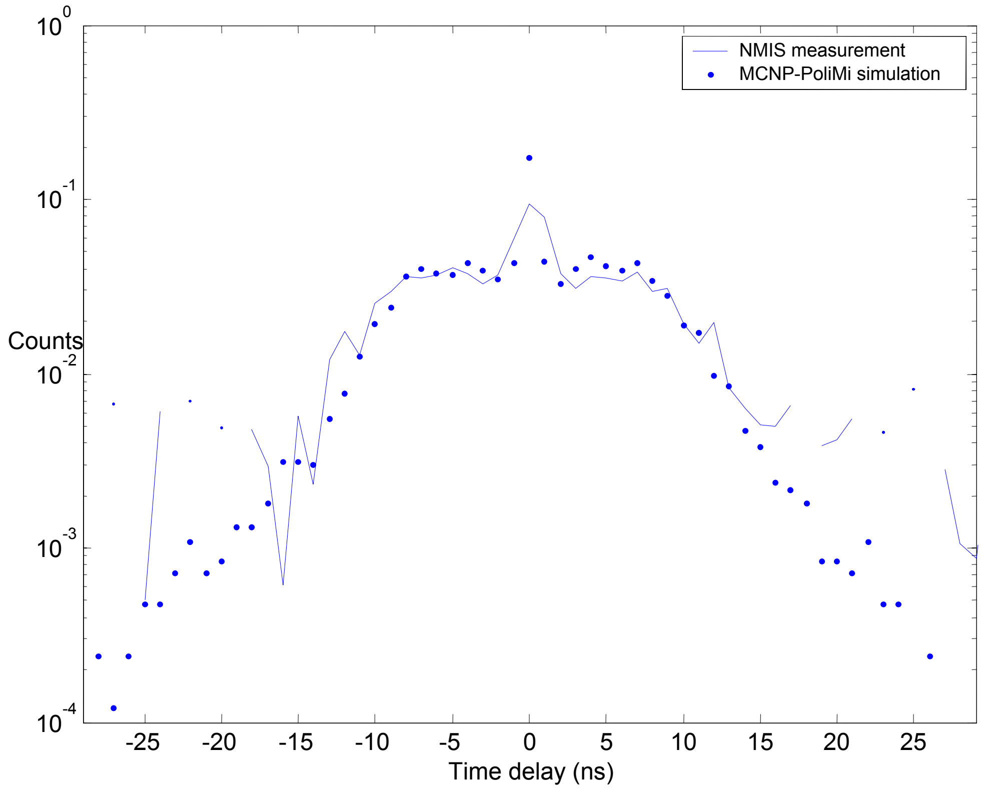

Figure 3. Detector-detector cross-correlation for plutonium shell *Pu7*.

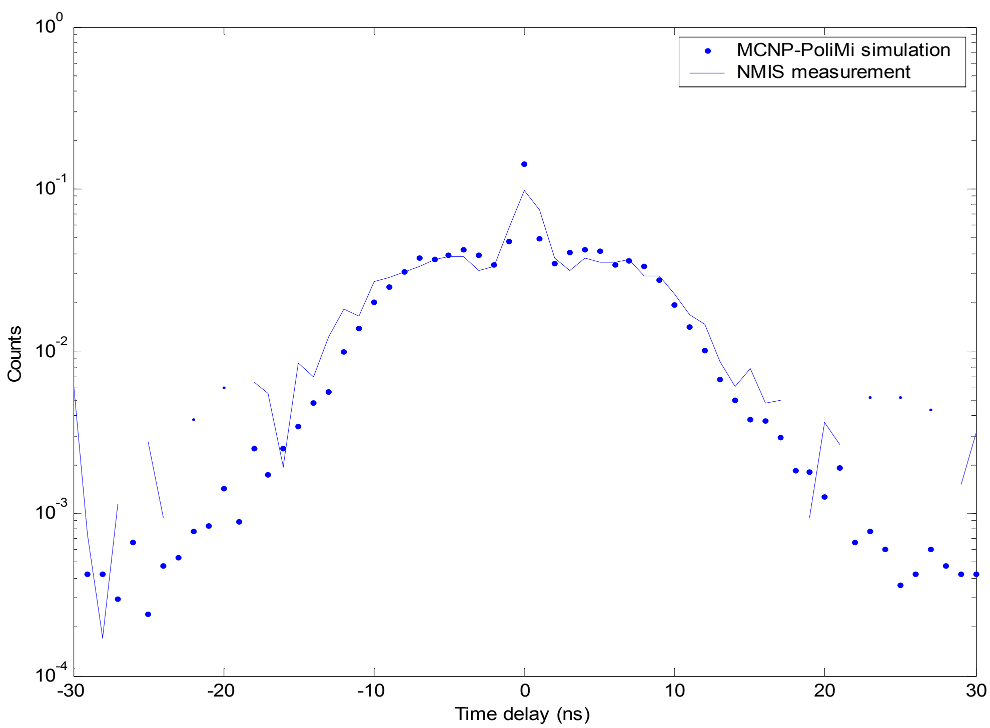

Figure 4. Detector-detector cross-correlation for plutonium shell *Pu4*.

The Monte Carlo simulation is generally in good agreement with the measured signature. As it can be seen, the width of the simulated gamma-pairs peak is smaller than the corresponding measured width by approximately 1 ns. This can be explained by considering time resolution of the electronics, which is not modeled in the post-processing code. In particular, the constant fraction discriminator timing is not taken into account in the post-processing code. The gamma-neutron peaks are in good agreement until time lags of approximately 15 ns. At greater time lags, the experimental data lack statistical significance to make a meaningful comparison.

The analysis of measurements performed with NMIS to obtain attributes such as mass and thickness requires the selection of features from the cross-correlation functions. In previous works, the features were shown to be related to the attributes of the fissile sample, for example its fissile mass [5, 6, 9]. Examples of such features are the area of the gamma peak and the area of the neutron peak. Table III shows the Monte Carlo error in the prediction of the area of the gamma and neutron peak for the cases shown in Figs. 3 and 4, together with the root-mean-square error.

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

Plutonium assembly	Gamma peak (% error)	Neutron peak (% error)
Pu4	+2.9	-0.74
Pu7	+6.1	-2.2
RMS	4.77	1.64

3.3. MCNP-PoliMi Simulations of Active Measurements on Plutonium

Further simulations were performed for three setups similar to the one shown in Fig. 5. In this 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 8.

Fig. 6 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 the parameters given in Section 2. The signature consists of two modes: the first mode 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 mode 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.

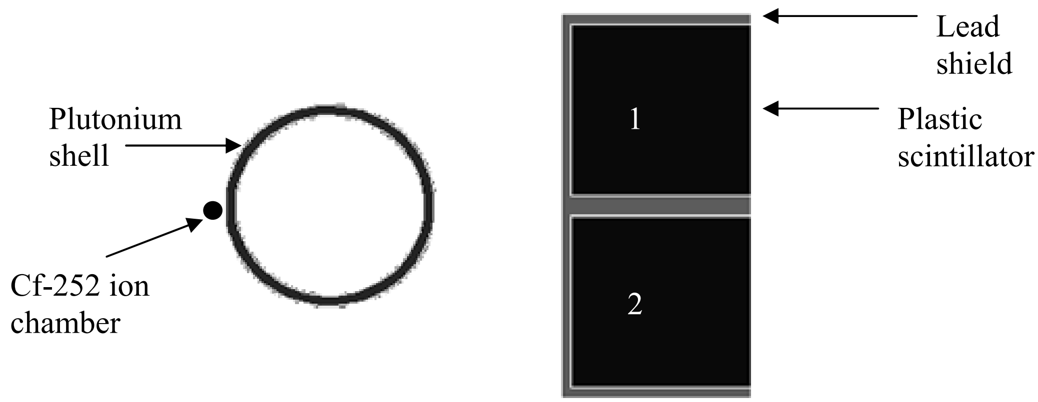

Figure 5. Geometry for one of the MCNP-PoliMi simulations for active measurements on plutonium shells. Cross section from top is shown, with plutonium spherical assembly between the Cf-252 instrumented source and two detectors (labeled 1-2).

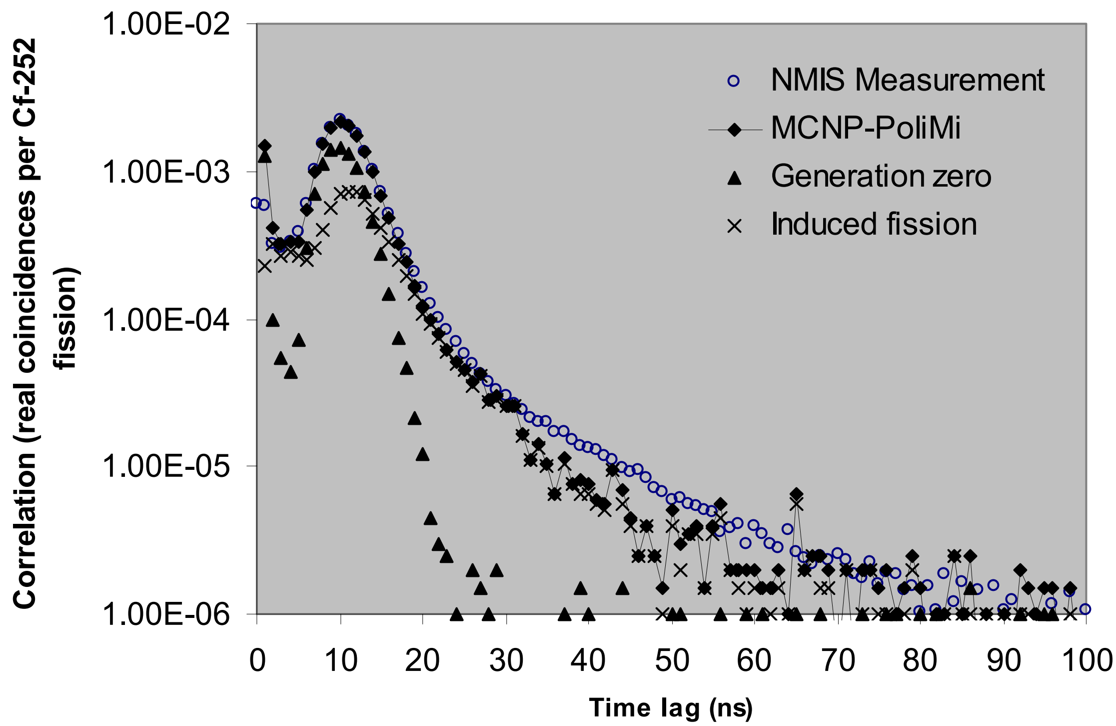

Figure 6. Source-detector cross-correlation for plutonium shell *Pu3*.

A possible explanation of the difference in the tail of the second mode 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 and discussed in Section 2.2. 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 code[2].

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). Fig. 6 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 13.

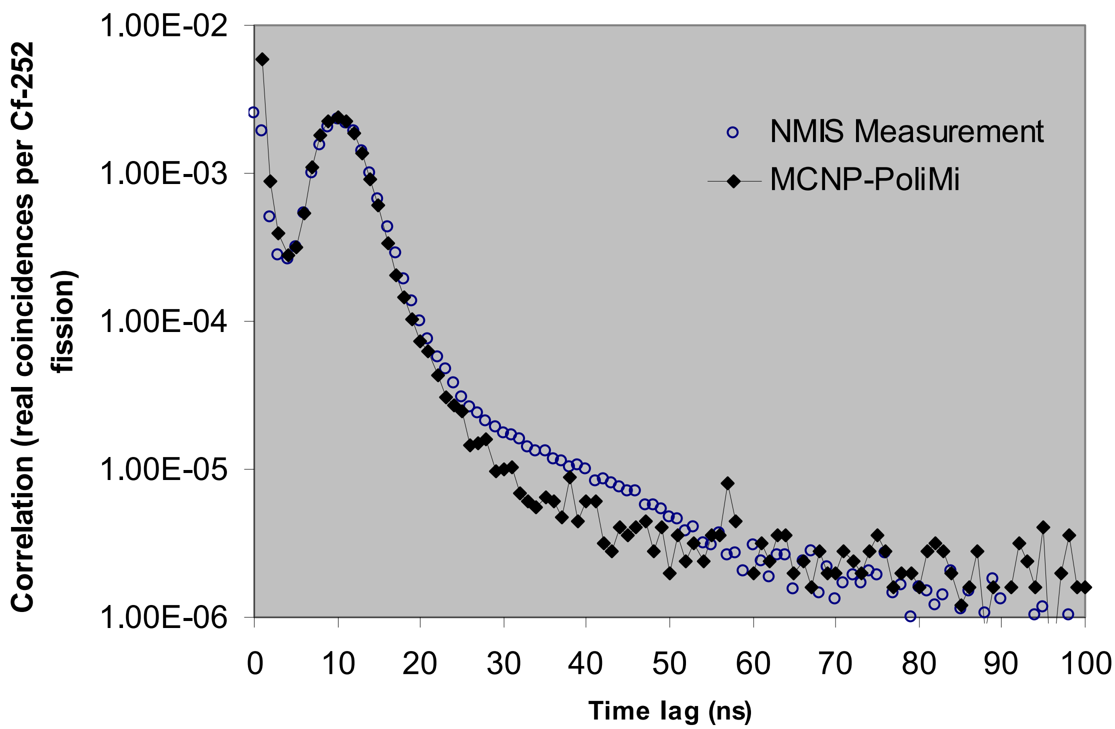

Figure 7. Source-detector cross-correlation for plutonium shell, *Pu4*.

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[2] 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.

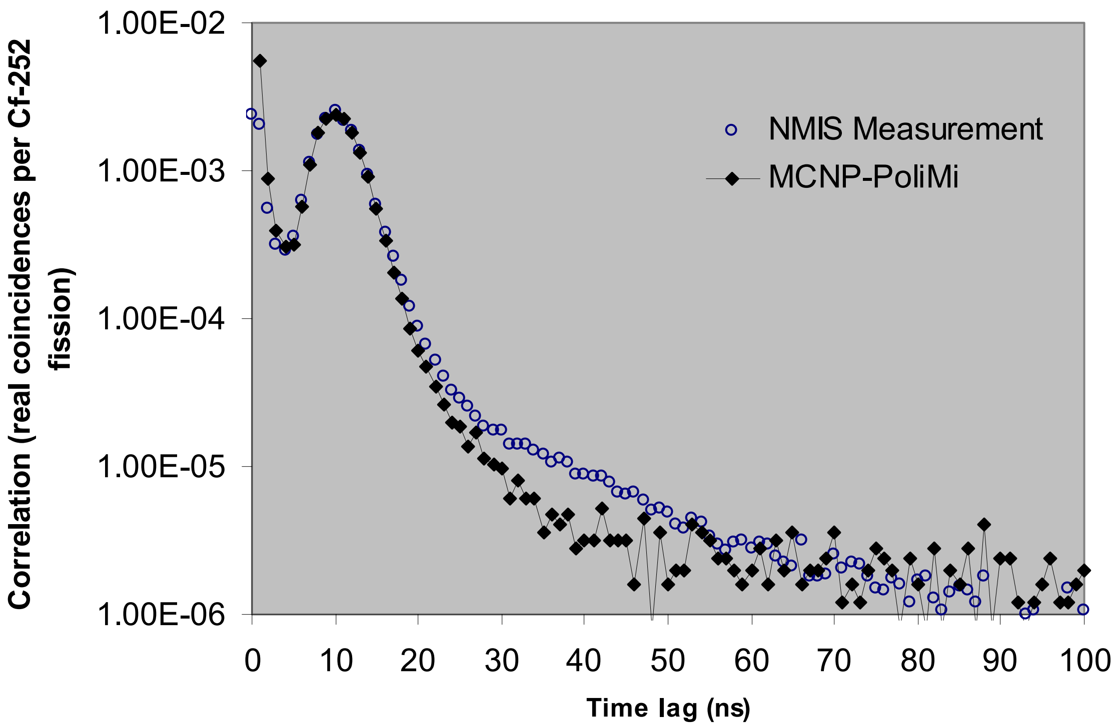

Figure 8. Source-detector cross-correlation for plutonium shell, *Pu7*.

Figs. 7 and 8 show the comparison between the experiment and the simulation result for the plutonium metal shells *Pu4* and *Pu*7, respectively. Similarly to the results shown in Figure 6 for assembly *Pu3*, the agreement is very good for delays up to 20 ns, approximately. Then, the simulated signature for the tail of the second mode is lower than the measurement.
Table IV shows the percentage error for the two modes 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 mode and 2.36% in the case of the second mode.

Table IV. Percent error in the MCNP-PoliMi simulation of active measurements.

System ID	First mode (% error)	Second mode (% error)
Pu3	+2.5	+0.9
Pu4	+9.2	+0.8
Pu7	+5.9	-3.9
RMS	6.47	2.36

4. 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 [14]. In these measurements, the Nuclear Materials Identification System (NMIS) was used in active mode, with a Cf-252 ionization chamber providing the trigger pulse.

4.1 Experimental Setup

The experiment was performed with the configuration shown in Fig. 9. In this 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. The table top was 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 14.

Many signatures were acquired in the measurements. Of these, the ones used for the code validation are the cross-correlation functions of the signals from the source and detector 1, and the cross-correlation of the signals between detectors 1 and 2.

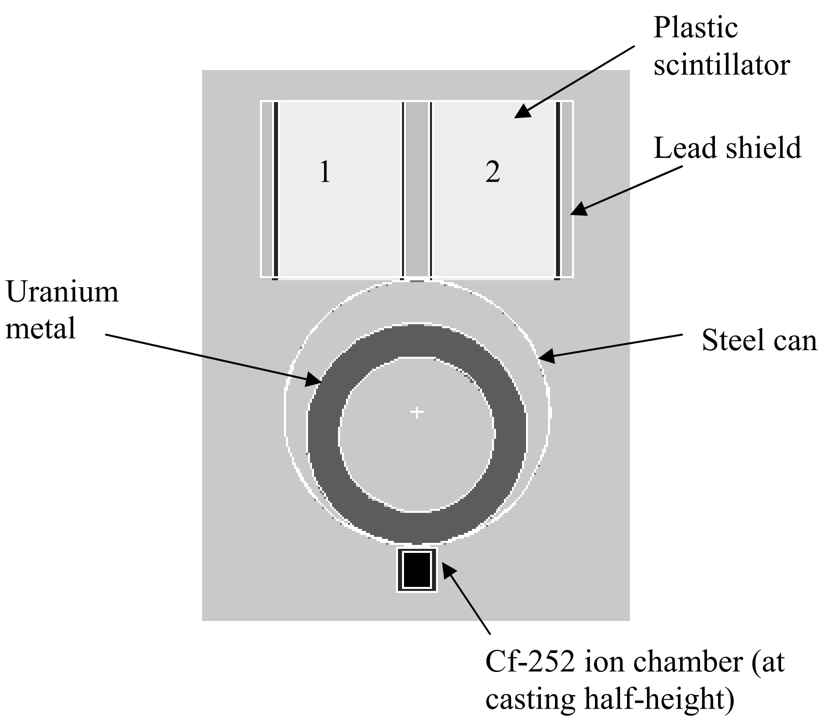

Figure 9. Geometry of the MCNP-PoliMi simulations. Cross section from top is shown, with uranium metal casting between Cf-252 source and four detectors placed in a 2x2 array (only detectors 1 and 2 are visible). Not shown, floor and table.

The properties of the uranium metal casting are reported in Table V.

Table V. 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

4.2 . MCNP-PoliMi Simulations of Active Measurements on Uranium

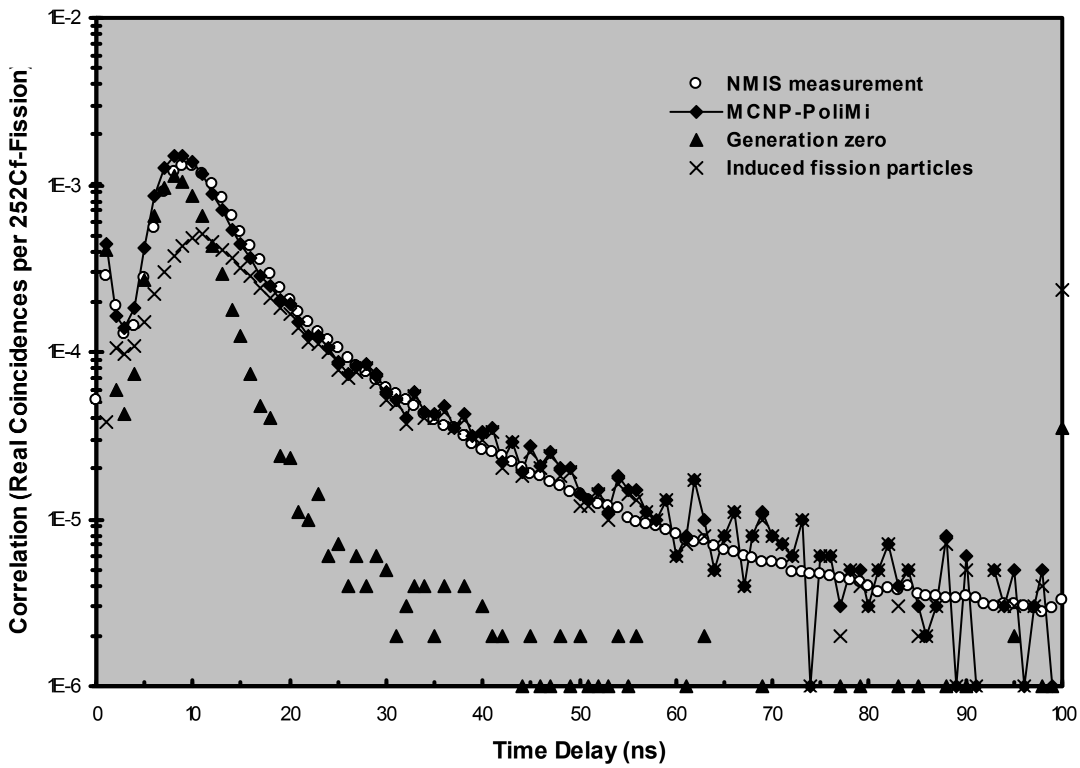

Figure 10. Source – detector 1 cross-correlation with uranium metal casting: MCNP-PoliMi simulation is subdivided into its generation zero component and induced fission particles component.

The measured and simulated source-detector cross-correlation functions are shown in Fig. 10. 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. 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. As it can be seen in Fig. 10, 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 VI reports the percentage error for the prediction of the area of the first and second mode in the case of cross-correlation between the source and detector 1.

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

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

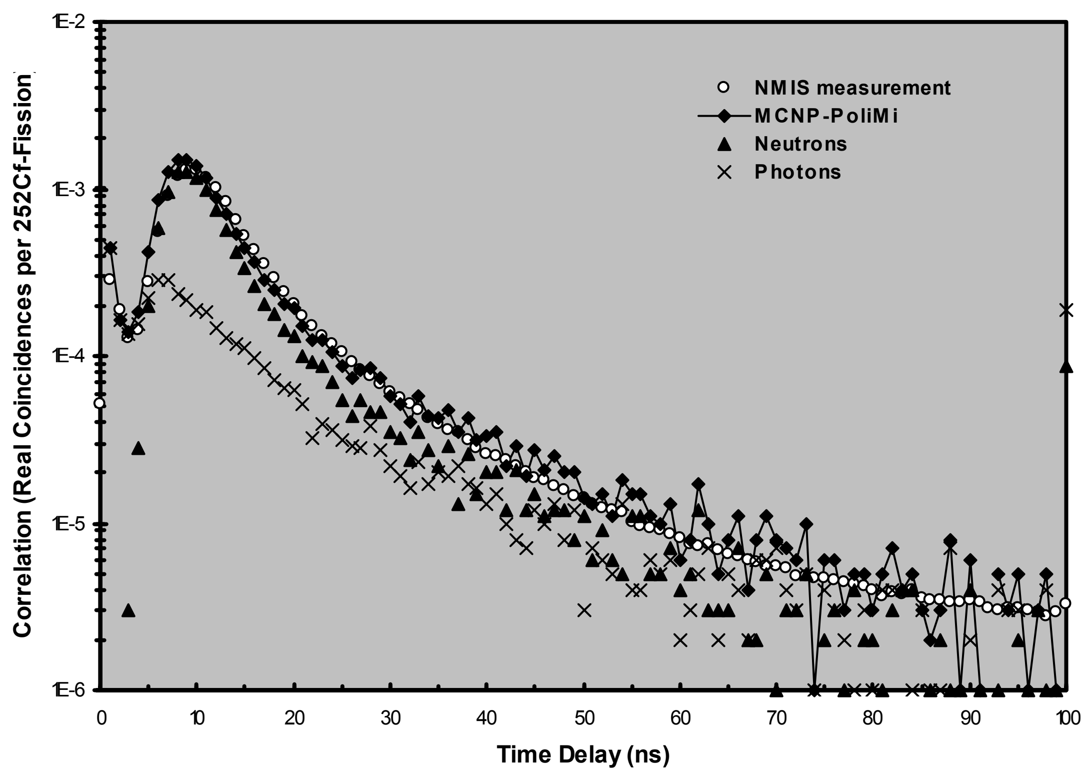

Figure 11. Source – detector 1 cross-correlation with uranium metal casting: MCNP-PoliMi simulation is subdivided into neutron and photon contributions.

A correlated pulse in the detector can be originated by a neutron or a photon. A split of the signature can be performed with the post-processing code according to the detected particle type. The result is shown in Fig. 11.

The cross-correlation between detector 1 and detector 2 was simulated as well. Figure 12 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 [15]. 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 VII, might be reduced accordingly.

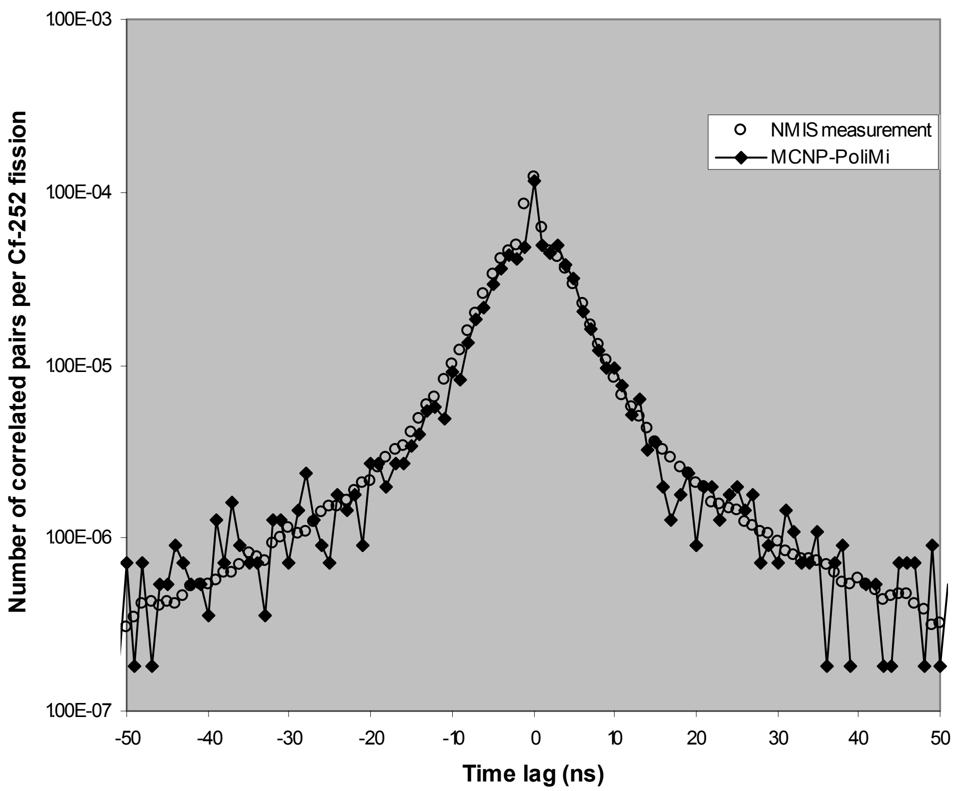

Figure 12. Detector 1 – detector 2 cross-correlation with uranium metal casting.

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 [16] 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 VII. Percent error in the MCNP-PoliMi simulation of measurements: total area.

Cross-correlation	Total area (% error)
Detector 1 – detector 2	-6.3

5. 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 the case of passive measurements on plutonium, the RMS percentage errors in predicting the areas of first and second modes of the cross-correlation were approximately 4.8 and 1.6%, respectively. In the case of active measurements on plutonium the area of the first and second modes of the cross-correlation had an RMS percentage error of approximately 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 region of partial disagreement of the simulated curve with the experiment.

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

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