PHOTONUCLEAR PHYSICS MODELS, SIMULATIONS, AND EXPERIMENTS FOR NUCLEAR NONPROLIFERATION

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ABSTRACT

This work illustrates a methodology based on photon interrogation and coincidence counting for determining the characteristics of fissile material. The feasibility of the proposed methods was demonstrated using the Monte Carlo-based MCNPX/MCNP-PoliMi code system capable of simulating the full statistics of the neutron and photon field generated by the photon interrogation of fissile and non-fissile materials with high-energy photons. These simulations were compared to the prompt time-of-flight data taken at the Idaho Accelerator Center immediately following the photon interrogation of a depleted uranium target. The results agree very well with the measured data for interrogation with 15-MeV endpoint bremsstrahlung photons at two different detector separation distances.

Keywords: MCNP; MCNP-PoliMi; Correlation measurement; Nuclear safeguards

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1. INTRODUCTION

Recent efforts have focused on the development of new measurement systems for the identification of nuclear material enclosed in shielded containers having applications in the areas of nonproliferation and homeland security [1–4]. The detection of shielded highly enriched uranium is particularly challenging because uranium, in contrast to the even-numbered plutonium isotopes, has a very low spontaneous fission rate, making passive interrogation difficult. As a consequence it becomes important to investigate active interrogation methods based on neutron or gamma-ray sources. In this paper a measurement system based on coincidence-counting of neutrons and gamma rays from the photon interrogation of nuclear materials is proposed.

All photonuclear interactions will proceed analogous to compound nucleus formation for the photon energy range considered here (i.e., greater than 10 MeV). Once the compound nucleus has been formed, it may de-excite through fission or the emission of neutrons, photons, or charged particles. For neutron or charged-particle emission from the target nucleus, the incoming photon must possess kinetic energy greater than the separation energy of the particle to be emitted. These reactions are referred to as threshold reactions, with energy thresholds typically on the order of 5–6 MeV for heavy nuclei. To induce fission the incoming photon must have kinetic energy greater than the critical energy of the target nucleus, which is energy also around 5–6 MeV for heavy nuclei. Once the compound nucleus has been formed, de-excitation will proceed through one of the possible channels governed by the reaction cross section. The dominant photonuclear cross sections for U-238 are shown in Figure 1 [1]. The photonuclear cross section for U-238 is illustrative of the trend seen for the other isotopes of interest (U-235, Pu-239, etc.).

![Figure 1. Photonuclear reaction cross sections for U-238.](image-url)
To determine the feasibility and sensitivity of such a measurement system, a Monte Carlo code system consisting of modified versions of the codes MCNPX and MCNP-PoliMi has been developed. The codes simulate the neutron and photon fields generated during the interrogation of fissile (and non-fissile) material with a high-energy photon source (greater than or equal to 10 MeV). Photoatomic and photonuclear collisions are modeled and time-correlated detections and multiplicities computed. These simulation results are compared with experimental data taken at the Idaho Accelerator Center (IAC) linear accelerator (LINAC).

2. MONTE CARLO SIMULATIONS

Monte Carlo codes have been widely used to design and analyze measurement scenarios. However, when modeling the time-correlated events resulting from photon interrogation, the widely used Monte Carlo code MCNPX has some limitations. Specifically, when considering a single interaction, MCNPX deviates from physical reality, and the particles resulting from photonuclear interactions are not correctly modeled [5]. An enhanced version of MCNP4c called MCNP-PoliMi has been developed to simulate time-analysis quantities and include a correlation between individual neutron interactions and corresponding photon production [6].

2.1. Description of the MCNP-PoliMi Code System

MCNPX deviates from physical reality so that the particles resulting from photonuclear interactions are not accurately modeled on an event-by-event basis. A second, more complex deviation from reality relates to the emission of the gamma rays resulting from neutron interactions. In standard MCNP, secondary gamma rays are sampled from a distribution that is not dependent on the associated neutron interaction [5]. A recently developed, enhanced version of MCNP4c, called MCNP-PoliMi, preserves standard MCNP code structure, while correcting this deficiency [6–8]. The modified code simulates time-analysis quantities and includes a correlation between individual neutron interactions and the corresponding photon production. The primary modification inverts the order of two sampling routines: the secondary photon production and the neutron-collision-type determination.

MCNP-PoliMi is capable of running with all standard MCNP source types and includes several specific spontaneous-fission-source definitions (i.e., Cf-252, Pu-240, Pu-242, Cm-242, Cm-244), as well as Am-Li and Am-Be isotopic sources. In addition, a photonuclear source file may be generated using a modified version of MCNPX and read by MCNP-PoliMi. This source file is generated by simulating the interrogation of the target material by the photon beam and recording relevant information on all photonuclear events, including photofission, (\(\gamma\), n), and (\(\gamma\), 2n) reactions. The information recorded to this file includes the location and multiplicity of neutrons and gamma rays emitted by the photonuclear events. This source is read by MCNP-PoliMi, and the particles are transported through the system and into the detectors. The energy released during each collision in the detectors, the corresponding time, the incident particle type, and the target nucleus are saved in a dedicated output file.
A post-processing code is then used to load the required data from this file and compute the detector-specific response. In the case of a scintillation detector, the incoming radiation must deposit enough energy to overcome a specific threshold for light output. Different incoming particles interact in very different ways; photons interact primarily through Compton scattering on electrons, while neutrons interact through scattering on hydrogen. The event-by-event physics modeled in MCNP-PoliMi enables the simulation of detailed detection physics, which are typically disregarded in other simplified code systems. Specifically, the data shown in the MCNP-PoliMi output file enable the modeling of effects, such as the varying light outputs of different target nuclei, as well as the effect of multiple particle-scattering events.

2.2. Monte Carlo Model of Idaho Accelerator Center Experiments

Experimental data were acquired at IAC using a 15-MeV endpoint bremsstrahlung source and a depleted uranium target. The source diameter was assumed to be equal to that of the collimator. Time-of-flight measurements were made with two 2-in.-by-2-in. cylindrical plastic scintillation detectors. The detector directly in line with the beam was designated as the start detector; a lead-shielded stop detector was oriented 90° to the beam line. Figure 2 shows an illustration of the experimental setup.

![Figure 2. Schematic of the MCNP-PoliMi model of the existing Idaho Accelerator Center experimental setup (not to scale).](image-url)

Pulses of source photons interact with the target, producing secondary particles. Photons transmitted through the target serve as the trigger for the start detector. After the start signal, the stop detector is active to detect secondary particles arriving with some time distribution after the start detector is triggered (time = 0 ns). The resulting distribution,
shown in Figure 3, represents the time of flight of secondary particles arriving at the stop detector 111 cm from the depleted uranium target. Data were taken for two different stop-detector distances (1.11 and 2.01 m). The resulting distributions are characterized by two bulk features: a sharp initial peak followed by a smooth, longer distribution of counts.

The initial peak corresponds to the arrival of the prompt gamma rays at the stop detector; all of the photons arrive at essentially the same time because they all travel at the speed of light and start from approximately the same point. The subsequent distribution is the arrival of the photoneutrons at the stop detector. These secondary neutrons originate from three reactions: \((\gamma, n)\), \((\gamma, 2n)\), and \((\gamma, \text{fission})\). Neutrons from each of these reactions are sampled from appropriate energy distributions. MCNPX calculates the number of times each reaction occurs, as well as the average energy of the neutrons released; Table 1 summarizes this information.

Each reaction produces a different number of neutrons, each of which has a different energy; these energy distributions directly correspond to a distribution of velocities. Assuming that each neutron originates from the same point, they would arrive at the detector at a time equal to the velocity times the fixed detector separation. Therefore, the peak time in the distribution is proportional to the detector separation times the average neutron velocity (i.e., energy); Table 1 lists these values for a 1.11-m detector separation. This effect is illustrated in the results shown in Figure 3; the distribution at larger detector separation is shifted to later times due to the increased stop-detector separation.

Photons from the bremsstrahlung beam produce photoatomic reactions in addition to the photonuclear ones. By performing a dedicated MCNP-PoliMi simulation, it is possible to take into account these photoatomic contributions. However, these contributions carry no nuclear information about the sample and thus are totally neglected. This is a valid assumption in that the stop detectors are triggered on immediately after the initial, intense photon burst has passed and before neutrons and semi-prompt gamma rays arrive. This timing technique also dramatically reduces the rate of accidental coincidences, which is one limiting factor affecting the feasibility of correlation measurements such as those proposed here.

2.2. Monte Carlo Simulation Results

The simulations were performed using the most current version of the MCNPX/MCNP-PoliMi code system, which explicitly models the full statistics of the neutron/photon field generated from photonuclear events. In addition, a detector-specific post-processing code simulates the detector response to the incident particles. In this research plastic scintillation detectors were simulated with a detection threshold of 0.001 MeVee (equivalent to 10 keV incoming neutron energy). The threshold was treated as a free parameter in the final comparison. A threshold of 0.001 MeVee gave the best data fit after normalization to the neutron-peak area. The system geometry shown in Figure 2 was modeled, including a few assumptions that differ from the exact reality of the experimental setup.
Figure 3. Idaho Accelerator Center measurement results for bremsstrahlung interrogation of a depleted uranium target; data are shown for two different stop-detector distances.

Table 1. Percentage and average energy of the three possible photonuclear reactions occurring in U-238 as obtained from MCNPX. The average flight time to the 1.11-m stop detector is also shown.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Percentage of Total Reactions</th>
<th>Average Neutrons per Reaction</th>
<th>Average Neutron Energy</th>
<th>Average Neutron Flight Time</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(\gamma, n)$</td>
<td>64.27%</td>
<td>1.0</td>
<td>918 keV</td>
<td>83.64 ns</td>
</tr>
<tr>
<td>$(\gamma, 2n)$</td>
<td>11.58%</td>
<td>2.0</td>
<td>665 keV</td>
<td>98.27 ns</td>
</tr>
<tr>
<td>$(\gamma, \text{fission})$</td>
<td>24.15%</td>
<td>2.4</td>
<td>2.0 MeV</td>
<td>56.67 ns</td>
</tr>
</tbody>
</table>

First, the LINAC tungsten converter was modeled separately; the photon energy spectrum was determined, and a beam with a radius equal to that of the collimator was modeled. This model is equivalent to assuming perfect collimation, which is valid considering the effect of the lead collimator plugs and the thickness of the concrete wall. Second, in the post-processing, some detector-specific properties, such as threshold, dead-time, and pulse-generation-time, had to be assumed because they were not provided by IAC with the data. Finally, to make a direct comparison, the data from IAC, as well as the MCNP-PoliMi simulation results, were normalized to the total number of neutron counts. Figure 4 shows the results for both stop-detector separations.
Figure 4. Comparison of MCNP-PoliMi calculations to the Idaho Accelerator Center experimental data for (a) 1.11-m and (b) 2.01-m stop-detector separation from a depleted uranium target.

For the 1.11-m separation case shown in Figure 4a, the MCNP-PoliMi results agree very well over the full range of experimental data. There is some disagreement in the intensity.
of the photon peak; MCNP-PoliMi simulates only the particles from the photonuclear reactions. In the experiment there are some photons from the primary beam scattered into the stop detector. There is some error at later times when the response is low. These extra detections, not modeled by MCNP-PoliMi here, arise from background in the room or from other scattering sources not included in the model.

MCNP-PoliMi has the capability of grouping the detector response by the originating photonuclear reaction. Figure 5 illustrates this capability for the 1.11-m separation case. Neutrons from \((\gamma, 2n)\) have the smallest contribution to the total response, while neutrons from \((\gamma, n)\) and \((\gamma, \text{fission})\) reactions have a nearly equal contribution. These findings agree with the reaction percentage from MCNPX, as shown in Table 1, taking into account that each \((\gamma, \text{fission})\) reaction produces approximately two neutrons. The peaks visually appear at approximately the same time for each reaction despite their differing average energies. The neutron velocity is proportional to the square root of the energy, and the flight time is inversely proportional to the velocity. Therefore, the neutron flight time is inversely proportional to the square root of its energy; thus, the small difference in the average energy has a lessened effect on the resulting distribution.

![Figure 5. MCNP-PoliMi simulation results for the existing experimental setup at 1.11-m stop-detector separation; contributions to the total response from each photonuclear reaction are shown.](image)

3. CONCLUSIONS
Active interrogation of nuclear materials is at the forefront of technologies for the detection of concealed nuclear material. One central concept of most current systems is the detection of delayed neutrons from fission reactions induced by interrogating photons. However, valuable information exists in the prompt regime (sub-microsecond) that has yet to be fully utilized. The unique capabilities of the existing MCNP-PoliMi present the possibility of simulating such behavior.

The results presented here clearly show that the MCNPX/MCNP-PoliMi code system is capable of accurately predicting the prompt correlated-detector response to uranium interrogation with high-energy photons. A few assumptions were made to obtain these results, the most notable of which was the free adjustment of the detection threshold. The final value of 0.001 MeVee was taken to give the most accurate results after normalization to the neutron-peak area. We are currently investigating methods for comparing the data on a per nano-Coulomb basis; this would provide a more robust result depending only on the charge of the LINAC.

The MCNP-PoliMi code system calculates the full statistics of the neutron/photon field directly resulting from photonuclear interactions. This information is then analyzed by a detector-specific post-processor. In this work the physics of plastic scintillation detectors were simulated, and the time correlations were determined. These capabilities result in a code system that simulates the full interrogation process, from source particles to the final detector response, in an accurate way.

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