Simulation of delayed γ -ray emission following photofission reactions induced by pulsed bremsstrahlung x-rays using MCNPX and experimental validation

Xianfei Wen^{1, 2}, Haori Yang^{2, *}

¹*Nuclear Engineering Program, University of Florida, Gainesville, FL 32611, USA*

²*School of Nuclear Science and Engineering, Oregon State University, Corvallis, OR 97331, USA*

Abstract

There is a great demand to develop non-destructive techniques to identify and quantify Special Nuclear Materials (SNM) in homeland security and nuclear safeguards applications. Passive assay could be extremely challenging in some scenarios. Active interrogation technique based on photofission has been identified as one of the promising approaches. In radiation detection system design based on such technique, it is highly desired to have abilities to accurately and efficiently simulate delayed γ -rays emitted from photofission reactions. In this work, simulation results were compared with measurement outcomes to demonstrate the capabilities and limitations of the code MCNPX 2.7.0 in the simulation of delayed γ -rays from photofission of uranium and plutonium samples. First, high-energy delayed γ -rays ($E_\gamma \sim 2.7 - 4.5$ MeV) from photofission of ^{238}U were simulated and validated against the energy spectra measured in between linac pulses. Second, low-energy delayed γ -ray spectra ($E_\gamma \sim 0.6 - 2.7$ MeV) measured with a list-mode system after irradiation of ^{239}Pu were used in the validation.

Keywords: MCNPX; simulation; photofission; delayed γ -rays; nuclear safeguards;

*Corresponding author.

Address: School of Nuclear Science and Engineering, Oregon State University, Corvallis, OR 97331, USA.

E-mail: wenxianfei@ufl.edu (X. Wen); haori.yang@oregonstate.edu (H. Yang).

1. Introduction

Accurate identification and quantification of SNM is urgent in homeland security and nuclear safeguards applications as the peaceful use of nuclear energy around the world is increasingly demanded. Non-destructive evaluation techniques including passive and active assay are of great interest in nuclear communities [1-3]. Passive

nondestructive assay is very challenging in some scenarios. For example, it is not practicable to measure naturally emitted γ -rays or neutrons when nuclear materials are shielded in purpose with high atomic number / hydrogenous materials due to the low intensity and energy of such radiation. Measurement of delayed neutrons or high-energy delayed γ -rays from photofission reactions has been identified as an effective approach to address the challenges [4-11]. Radiation protection is challenging in real-world active interrogation based on photofission techniques due to the large flux of gammas produced by linac. Optimization of linac (e.g. energy, current, frequency) could be one way to minimize the unwanted dose to people. High-throughput detectors could also be effective.

Designing photofission technique based radiation detection system will largely rely on Monte Carlo simulation to provide valuable information on experimental parameters to improve sensitivity and accuracy, including linac beam (energy spectrum, current, repetition frequency, and pulse width), sample orientation with respect to beam and detector, distance between sample and detector, detector shielding, irradiation, cooling, and measurement time, etc. The simulation results can be used to optimize a particular system design or assay approach. So far, simulation of delayed γ -rays has been validated against measurement results for neutron-induced fission. Non-exhaustive examples are summarized here. Durkee et al. demonstrated the delayed γ -ray feature using a 0.01975-cm ^{60}Ni sphere bombarded by 15-MeV neutrons [12]. The (n,p) reaction caused ^{60}Co to be produced. The two prominent ^{60}Co lines (1.173 MeV, 1.332 MeV) were observed in the F1 tally binned in time from 10^5 to 10^9 s. The Fisher and Engle experiment involving ^{235}U irradiation by a Watt-fission neutron source was also used by Durkee et al. to illustrate the delayed γ -ray feature [13]. They showed that the simulated results were in good agreement with the low-resolution measured data. Durkee et al. also presented the comparison between the simulated results and the high-resolution energy spectra obtained by Beddingfield and Cecil [13]. Although the simulated spectra were in good agreement with the experimental data, large differences in peak amplitude were observed. Andrews et al. showed that the simulation successfully predicted the 25 most prominent peaks generated from fission products in the 0.1 - 1.6 MeV energy range [14]. For some peaks (e.g. 0.710, 0.724, 0.832 MeV), however, the ratios of simulated to measured counts were reported to reach ~ 0.6 .

Nevertheless, published literature on comparison between measured and simulated delayed γ -rays from photofission for code validation purpose is rare. The delayed γ -ray feature was illustrated by Durkee et al. in the

56 photofission of ^{235}U [12]. Monoenergetic photons (12 MeV) were used to induce the photofission reactions. Delayed
57 γ -ray spectrum with discrete lines was observed. However, it was compared with the measured delayed γ -rays
58 produced from neutron-induced fission of ^{235}U . It was the authors' intention to validate the simulation of delayed γ -
59 rays from photofission through comparing the simulation results with our high-fidelity measured data at Idaho
60 Accelerator Center (IAC). The structure of the papers is as follows. First, the high-energy delayed γ -rays from
61 photofission of ^{238}U measured in between linac pulses were used to validate the delayed γ -ray feature. Then the
62 measured low-energy delayed γ -ray spectra from photofission of ^{239}Pu using the list-mode system were used in the
63 validation process.

65 2. Simulation of high-energy delayed γ -rays from photofission of ^{238}U

66 The delayed γ -ray spectra to be used in the comparison were measured in our recent experiments at IAC. A
67 picture of the experimental setup is shown in Figure 1. The 25-MeV linac was used to generate the high-energy
68 pulsed bremsstrahlung x-ray beam. The endpoint energy of the bremsstrahlung beam was 22 MeV. The repetition
69 frequency of the linac was 10 Hz. The width of each linac pulse was approximately 4 μs . The amount of charge
70 carried by each pulse was around 86 nC. The target was an 18.9 g/cm^2 depleted uranium plate. The mass of the
71 uranium sample was approximately 2 kg. The detector used in the experiment was a p-type coaxial High-purity
72 Germanium (HPGe) detector (model number: GC4020, relative efficiency: 40%, crystal size: 2.4" diameter by 2.3"
73 height). High-resolution spectroscopy measurement of delayed γ -rays was performed with the Canberra Lynx MCA.
74 The measurement time was 45 minutes. The measured spectra were analyzed using the Genie2K software. More
75 detailed description of the experimental setup and data acquisition and analysis can be referred to our recent
76 publication [15].

Fig.1. The experimental setup for the measurement of delayed γ -rays from photofission [15].

2.1. Description of the MCNPX simulation code

In the simulation, the geometry model was a sphere with a diameter of 1 cm surrounded by vacuum. The sphere was filled by ^{238}U of 19.1 g/cm^3 . Importance of neutron and photon within the sphere was set at 1. Neutron and photon were not tracked once they exited the sphere. The 22-MeV bremsstrahlung was produced in a separate simulation. It was then used to create the pulsed linac source based on the embedded source feature. The neutron physics card was set as *phys: n 3j -101*. The fourth entry *dnb=-101* on this card was used to produce delayed neutron from fission based on the CINDER90 models. Therefore, the ACE libraries were deprecated in this case. All other entries on the neutron physics card were kept at default values. The photon physics card was set as *phys: p 3j 1 j -102*. Biased photonuclear particle production was enabled by setting the fourth entry (i.e. *ispn*) on this card to 1. To accurately obtain energies of delayed γ -rays, models based on line emission data were selected by setting the sixth entry on the photon physics card to -102. In addition to the neutron and photon physics cards, the Activation Control Card (ACT) was also used to control the production of delayed neutrons and photons from fission events. The act card was set as *act fission=all nonfiss=none dn=model dg=lines*. The first entry on this card enabled creation of delayed particles from fission events. Production of delayed neutrons and photons from non-fission events were disabled by setting the second entry to none. The third and fourth entries had the same functionality as the entries of *dnb=-101* and *dgb=-102* on the neutron or photon physics cards. The act card was followed by the lca card. This card was used to select the Bertini, ISABEL, CEM03, or INCL4 physics model and set parameters used in the

Bertini and ISABEL models. The eighth entry on the lca card was set at -2. All other entries on this card were kept at default values. The physics settings in the MCNPX code are summarized below.

phys:p 3j 1 j -102

phys:n 3j -101

act fission=all nonfiss=none dn=model dg=lines

lca 7j -2

The last section of the input file for the simulation included tally cards. Tally type 1 was used to count delayed γ -rays at the sphere surface. Tally energy card *e1 1 6000i 5* divided the energy range from 1 MeV to 5 MeV into 6000 bins. Tally time card *t1 100 1e37 NT* separated the tally into two time bins: $-\infty$ to 100 shakes and 100 to 1×10^{37} shakes (1 shake = 10 ns). The entry *NT* means no total bins will be printed in the output data file. Two additional tally cards (i.e. *ft* card and *fu* card) were used. Combination of the *ft1 tag 1* and *fu1 ZZAAA.99999* provided the ability to only count delayed γ -rays from fission in the isotope *ZZAAA*. This could avoid the interference by γ -rays created from non-fission events, such as neutron-capture reactions. To reduce statistical error to an acceptable level, the total number of particle histories was 1×10^8 . The detector response was not considered in the simulation. Although the simulated spectra resemble more the measured results when taking into account the detector response using *f8* tally, only the intensity of each peak was needed in the comparison and *f1* tally can provide such information.

2.2. Results and discussion

The comparison between the measured and simulated delayed γ -ray spectra in the energy range between 2.6 MeV to 5.0 MeV from photofission of ^{238}U is present in Figures 2-3. As shown in those Figures, most measured peaks were successfully predicted by the simulation. However, one notable peak (2971.0 keV) predicted by the simulation was not observed in the measured spectra. Measured and simulated relative intensities with efficiency correction of the identified peaks are shown in Table 1. The intensities were normalized to the 3287.6 keV peak because it had the strongest intensity. The efficiency calibration was performed using the depleted uranium sample. The calibration curve in the energy ranging from 200 keV to 2000 keV was extrapolated to obtain the calibration for high-energy region (2000 - 4500 keV). The relative intensities of multiple peaks, including 2717.3 keV (^{95}Sr), 2933.1 keV (^{95}Sr),

3401.3 keV (^{97}Y), 3458.2 keV (^{93}Rb), 3534.2 keV (^{90}Rb), 3549.5 keV (^{97}Y), 3599.7 keV (^{91}Rb), 3867.6 keV (^{93}Rb), 3932.4 keV (^{88}Br), 4078.3 keV (^{91}Rb), 4135.5 keV (^{90}Rb), 4166.3 keV (^{89}Br), 4180.5 keV (^{87}Br), and 4365.9 keV (^{90}Rb), obtained from the simulation agree well (difference between measurement and simulation less than 10%) with those from the measured results. However, large discrepancies exist in some peaks including those at 2789.3 keV (^{106}Tc), 2868.9 keV (^{136}I), 2941.3 keV (^{98}Y), 3310.0 keV (^{98}Y), 3317.0 keV ($^{90\text{m}}\text{Rb}$), and 4450.2 keV (^{98}Y). For example, for the 2789.3 keV (^{106}Tc) peak, the measured relative intensity was 0.585, which was much larger than the value predicted by the simulation (i.e. 0.585 vs 0.165). Differences in relative intensities could be attributed to the fact that the photofission library used by MCNPX 2.7.0 is primarily based on neutron-induced fission data due to the lack of validated experimental data or theoretical models for photofission reactions. For example, the cumulative yields for neutron-induced fission of ^{237}U are used as the photofission cumulative yields of ^{238}U .

Fig. 2. Comparison between the measured high-energy delayed γ -ray spectra and simulated results (2.6 - 3.5 MeV).

Fig. 3. Comparison between the measured high-energy delayed γ -ray spectra and simulated results (3.5 - 5.0 MeV).

Table 1. Efficiency corrected intensities relative to the 3287.6 keV peak (The uncertainty in the simulation was given in the MCNPX output file. The measurement uncertainty was due to the calculation of peak area. The measurement uncertainty could be reduced by increasing the irradiation time.)

Nuclide	Half-life (seconds)	Energy (keV)	Measurement	Uncertainty (%)	Simulation	Uncertainty (%)	abs(Measurement- Simulation)
⁹⁵ Sr	23.90	2717.3	0.367	19	0.320	0.6	0.047
¹⁰⁶ Tc	35.6	2789.3	0.585	11	0.165	1.0	0.420
¹³⁶ I	83.4	2868.9	0.491	17	0.322	0.7	0.169
⁹⁵ Sr	23.9	2933.1	0.375	18	0.282	0.7	0.093
⁹⁸ Y	0.548	2941.3	0.997	9	0.784	0.5	0.213
⁹⁷ Y	3.75	3287.6	1.0	6	1.0	0.4	-
⁹⁸ Y	0.548	3310.0	0.428	13	0.323	0.6	0.105
^{90m} Rb	258	3317.0	0.213	20	0.088	1.1	0.125
⁹⁷ Y	3.75	3401.3	0.868	10	0.783	0.5	0.085
⁹³ Rb	5.84	3458.2	0.170	21	0.113	1.0	0.057
⁹⁰ Rb	158	3534.2	0.137	27	0.161	1.0	0.024
⁹⁷ Y	3.75	3549.5	0.177	33	0.178	0.8	0.001
⁹¹ Rb	58.4	3599.7	0.590	10	0.529	0.5	0.061
⁹³ Rb	5.84	3867.6	0.140	18	0.085	1.1	0.055
⁸⁸ Br	16.34	3932.4	0.166	25	0.157	1.2	0.009
⁹¹ Rb	58.4	4078.3	0.235	17	0.206	0.8	0.029
⁹⁰ Rb	158	4135.5	0.215	13	0.169	0.8	0.046
⁸⁹ Br	4.35	4166.3	0.079	28	0.066	1.3	0.013
⁸⁷ Br	55.6	4180.5	0.093	25	0.081	1.2	0.012
⁹⁰ Rb	158	4365.9	0.266	13	0.198	0.8	0.068
⁹⁸ Y	0.548	4450.2	0.559	9	0.456	0.6	0.103

3. Simulation of low-energy delayed γ -rays from photofission of ²³⁹Pu

As shown in Table 1, the half-lives of the fission products emitting high-energy delayed γ -rays are very short, allowing the measurement to be performed only in between linac pulses. However, some fission products have relatively long half-lives (e.g. ^{131}Sb , 23.0 minutes), which makes measurement of delayed γ -rays long after the irradiation feasible. The low-energy delayed γ -ray spectrum used in the validation process was measured using the list-mode system after irradiation of the plutonium sample. The mass of the sample was approximately 3 g. The isotopic composition was 95 a/o ^{239}Pu and 5 a/o ^{240}Pu . The sample was irradiated for 69 minutes. The measurement was not started until cooling the sample down for 15 minutes. The total data acquisition time was 1.5 hours. The linac settings, detector, and MCA were the same as those used in the uranium measurement. For detailed experimental setup and spectra data analysis, the reader could refer to our recent publication [15].

The simulation input file was the same as those described in Section 2.1 except the tally and material cards. The tally energy card *e1 0.6 4400i 5* divided the energy range from 0.6 MeV to 5 MeV into 4400 bins. The tally time card was *t1 5.04e11 1.044e12*, allowing the delayed γ -rays produced in the time window from 15 to 105 minutes after irradiation to be separated from other delayed γ -rays. The material card was changed to account for the plutonium sample. Figures 4-6 show the comparison between the low-energy delayed γ -ray spectra measured using the list-mode system and simulated results. There was reasonable agreement between measured and simulated results. Most measured peak lines were successfully predicted by the simulation. However, the measured strong 1293.6 keV peak was not observed in the simulation. Some peaks were under-predicted by the simulation (e.g. 1097.3 keV). The ratios of peak amplitudes were not determined because of the large uncertainties in the peak area calculation, particularly for the peaks above 1.5 MeV. The validation demonstrated that the simulation package MCNPX 2.7.0 has capabilities and limitations in the simulation of delayed γ -rays produced from photofission of ^{238}U and ^{239}Pu . High-fidelity photofission data should be measured and included in MCNPX nuclear data library, allowing significant improvement of delayed γ -ray features.

Fig. 4. Comparison between the low-energy delayed γ -ray spectra measured using the list-mode system and simulation results (0.6 - 1.2 MeV).

Fig. 5. Comparison between the low-energy delayed γ -ray spectra measured using the list-mode system and simulation results (1.2 - 1.8 MeV).

Fig. 6. Comparison between the low-energy delayed γ -ray spectra measured using the list-mode system and simulation results
(1.8 - 2.7 MeV).

4. Conclusion

In this work, the measured high-resolution delayed γ -ray spectra from photofission of ^{238}U and ^{239}Pu were used to validate the delayed γ -ray features in MCNPX 2.7.0. Reasonable agreement was observed between measurement and simulation results. Most measured peaks were successfully predicted by the simulation. However, the measured strong 1293.6 keV peak from photofission of ^{239}Pu was not observed in the simulation. The 1097.3 keV peak from photofission of ^{239}Pu was significantly under-predicted by the simulation. A notable peak (2971.0 keV) from photofission of ^{238}U was not presented in the measured spectra, however, it was predicted by the simulation. For many peaks, the simulated ratios of peak amplitudes were also significantly different from measurement results. Photofission data (e.g. yields, cross section) should be measured and included in MCNPX nuclear data library. Delayed γ -ray features can be greatly improved when high-fidelity photofission data is available. The improvement could provide guidance for optimizing radiation system design based on photofission technique in homeland security and nuclear safeguards applications.

Acknowledgement

This research is being performed using funding received from the DOE Office of Nuclear Energy's Nuclear Energy University Programs.

References

1. T. Gozani, "Active nondestructive assay - principles and applications," NUREG/CR-0602, 1981.
2. W. Hage, et al., "Nondestructive fissile material assay by induced fission neutron correlation," *Nucl. Instr. Meth. Phys. Res. A*, vol. 551, pp. 396-419, 2005.
3. F. Carrel, et al., "Identification and differentiation of actinides inside nuclear waste packages by measurement of delayed gammas," *IEEE Trans. Nucl. Sci.*, vol. 57, p. 2862, 2010.
4. L. Campbell, et al., "High-energy delayed gamma spectroscopy for spent nuclear fuel assay," *IEEE Trans. Nucl. Sci.*, vol. 58, pp. 231 - 240, 2011.

- 204 5. J. Jones, et al., "Photonuclear-based, nuclear material detection system for cargo containers," *Nucl. Instr. Meth.*
205 *Phys. Res. B*, vol. 241, pp. 770-776, 2005.
- 206 6. J. Stevenson, et al., "Linac based photofission inspection system employing novel detection concepts," *Nucl.*
207 *Instr. Meth. Phys. Res. A*, vol. 652, pp. 124-128, 2011.
- 208 7. R. Runkle, et al., "Rattling nucleons: New developments in active interrogation of special nuclear material,"
209 *Nucl. Instr. Meth. Phys. Res. A*, vol. 663, pp. 75-95, 2012.
- 210 8. T. Gozani, "Fission signatures for nuclear material detection," *IEEE Trans. Nucl. Sci.*, vol. 56, pp. 736-741,
211 2006.
- 212 9. D. Beddingfield et al, "Identification of fissile materials from fission product gamma-ray spectra," *Nucl. Instr.*
213 *Meth. Phys. Res. A*, vol. 417, pp. 405-412, 1998.
- 214 10. F. Carrel, et al., "Identification and differentiation of actinides inside nuclear waste packages by measurement
215 of delayed gammas," *IEEE Trans. Nucl. Sci.*, vol. 57, pp. 2862, 2010.
- 216 11. X. Wen, et al., "Simulation and measurement of delayed γ -rays after photon-induced fission," *Nucl. Instr. Meth.*
217 *Phys. Res. A*, vol. 729, pp. 781-787, 2013.
- 218 12. J. Durkee, et al., "Delayed-gamma simulation using MCNPX," *Nucl. Tech.* vol. 168, pp. 761-764, 2009.
- 219 13. J. Durkee, et al., "Delayed-gamma signature calculation for neutron-induced fission and activation using
220 MCNPX. Part II: Simulations," *Progress in Nuclear Energy*, vol. 51, pp. 828-836, 2009.
- 221 14. M. Andrews, et al., "MCNP6 simulations of gamma line emissions from fission products and their comparisons
222 to plutonium and uranium measurements," *Progress in Nuclear Energy*, vol. 79, pp. 87-95, 2015.
- 223 15. X. Wen, et al., "Photofission product yields of ^{238}U and ^{239}Pu with 22-MeV bremsstrahlung," *Nucl. Instr. Meth.*
224 *Phys. Res. A*, vol. 821, pp. 34-39, 2016.