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Geant4 modifications for accurate fission simulations

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Abstract

Monte Carlo is one of the methods to simulate the generation and transport of radiation through matter. The most widely used radiation simulation codes are MCNP and Geant4. The simulation of fission production and transport by MCNP has been thoroughly benchmarked. There is an increasing number of users that prefer using Geant4 due to the flexibility of adding features. However, it has been found that Geant4 does not have the proper fission-production cross sections and does not produce the correct fission products. To achieve accurate results for studies in fissionable material applications, Geant4 was modified to correct these inaccuracies and to add new capabilities. The fission model developed by the Lawrence Livermore National Laboratory was integrated into the neutron-fission modeling package. The photofission simulation capability was enabled using the same neutron-fission library under the assumption that nuclei fission in the same way, independent of the excitation source. The modified fission code provides the correct multiplicity of prompt neutrons and gamma rays, and produces delayed gamma rays and neutrons with time and energy dependencies that are consistent with ENDF/B-VII. The delayed neutrons are now directly produced by a custom package that bypasses the fragment cascade model. The modifications were made for U-235, U-238 and Pu-239 isotopes; however, the new framework allows adding new isotopes easily. The SLAC nuclear data library is used for simulation of isotopes with an atomic number above 92 because it is not available in Geant4. Results of the modified Geant4.10.1 package of neutron-fission and photofission for prompt and delayed radiation are compared with ENDFB-VII and with results produced with the original package.

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Keywords: Neutron-induced fission; Photofission; Prompt and delayed radiation; Multiplicity; Fission fragment; Time and energy dependencies; Delayed neutrons; cross section; fission rate.

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1. Introduction

Radiation transport simulations are commonly used to aid in the design of systems and experiments. There are several simulations codes used for low-energy physics including MCNP (LANL), VIM (Argonne), and Geant4 (CERN, SLAC). The Geant4 Monte Carlo package was designed and developed by an international collaboration initially mainly for high-energy applications and has an increasing user base attracted by the easy adaptation for different sets of applications. Since then, new extensions have been added for low-energy simulation, including fission. However, some processes such as delayed neutron production have not been implemented.

In addition to the missing delayed neutrons, we also found that the prompt neutron and gamma-ray multiplicity following fission, the cross section and the decay time of the delayed gamma rays are incorrect. This means that Geant4 should not be used for fission simulations because the results would be inaccurate and incomplete. In this work, we modified the Geant4 code to correct the data and the models and to add the missing capability. The results were verified for the main fissionable isotopes with the ENDF\B-VII libraries.

2. Fission simulation with default models in Geant4

We performed Geant4 10.1 simulations with a small sphere of pure nuclear materials hit by a pencil beam of thermal neutrons or 12MeV photons to determine the characteristics of the fission products. Geant4 was run with the NeutronHP model (high precision neutron model for neutron-fission), Cascade model (photofission) and Radioactive Decay Physics to compare the results.

2.1. Multiplicity

Zucker and Holden (1986) measured the prompt neutron multiplicity following neutron-induced fission of several fissionable isotopes as a function of the energy of the incident neutrons. Their data shows an average multiplicity of 2.41 neutrons/per fission for U-235 induced by thermal neutrons. Lengyel et al. (2016) claimed an empirical calculation of average prompt neutrons for photofission, which shows average 3-3.4 neutrons/per fission of U-235 induced by 12MeV photons. Valentine (2001) used an approximation adopted by spontaneous and neutron induced fission modules. The prompt gamma-ray multiplicity ranges from 0-20 gamma rays/per fission with an average that varies with isotope and average prompt neutrons. We used Valentine's model to estimate the average multiplicity values of U-235 induced by thermal neutrons and 12 MeV photons to be ~6.7 and ~7.6, respectively.

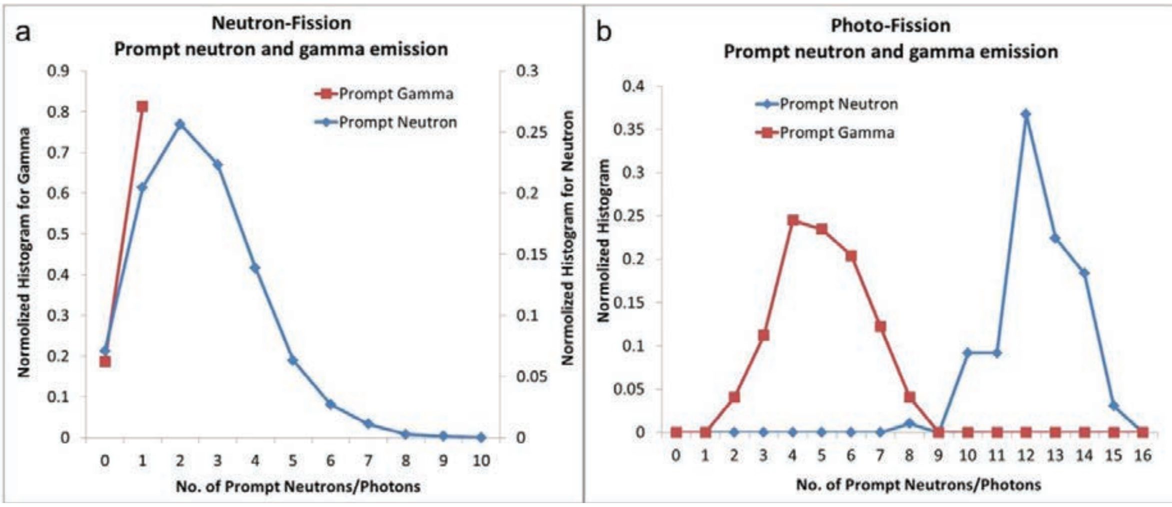

Fig. 1. Prompt multiplicity distributions for neutron induced fission (a) and photofission (b) simulated with Geant4 10.1.

The multiplicity distributions of prompt neutrons and gamma rays simulated with Geant4 for thermal-neutron and photon-induced fissions with U-235 are displayed in Fig. 1. The distributions do not agree with the accepted values. Fission induced by thermal neutrons only produced 0 or 1 gamma ray, instead 0 to 20 gamma rays with a mean value of 6.7, and fission induced by 12MeV photon produced almost three times higher number of neutrons and slightly fewer prompt gamma rays. We describe in section 3.1 how to correct these discrepancies.

2.2. Fission fragments yield distribution

During the fission process, in addition to prompt neutrons and gamma rays, the nucleus splits into two fragments. These neutron-rich fragments are highly unstable and decay by multiple β and neutron emissions resulting in delayed neutrons and gamma rays. The experimentally-measured atomic-mass fragment distributions are shown in Fig. 2a for various fissile isotopes induced by thermal neutrons. The distributions show two distinctive peaks at A~90-105 and A~135 to 145 depending on the isotope undergoing fission.

Geant4 supports two fission models, NeutronHP and Cascade that produce different fragment yield distributions as shown in Fig. 2b and 2c. It is evident that the fragment distribution resulting from the NeutronHP model agrees with experimental results, while the Cascade model shows a very different distribution. The fission fragment distribution plays an important role in fission simulation because it affects the properties of delayed radiation generated by the fission fragment decay.

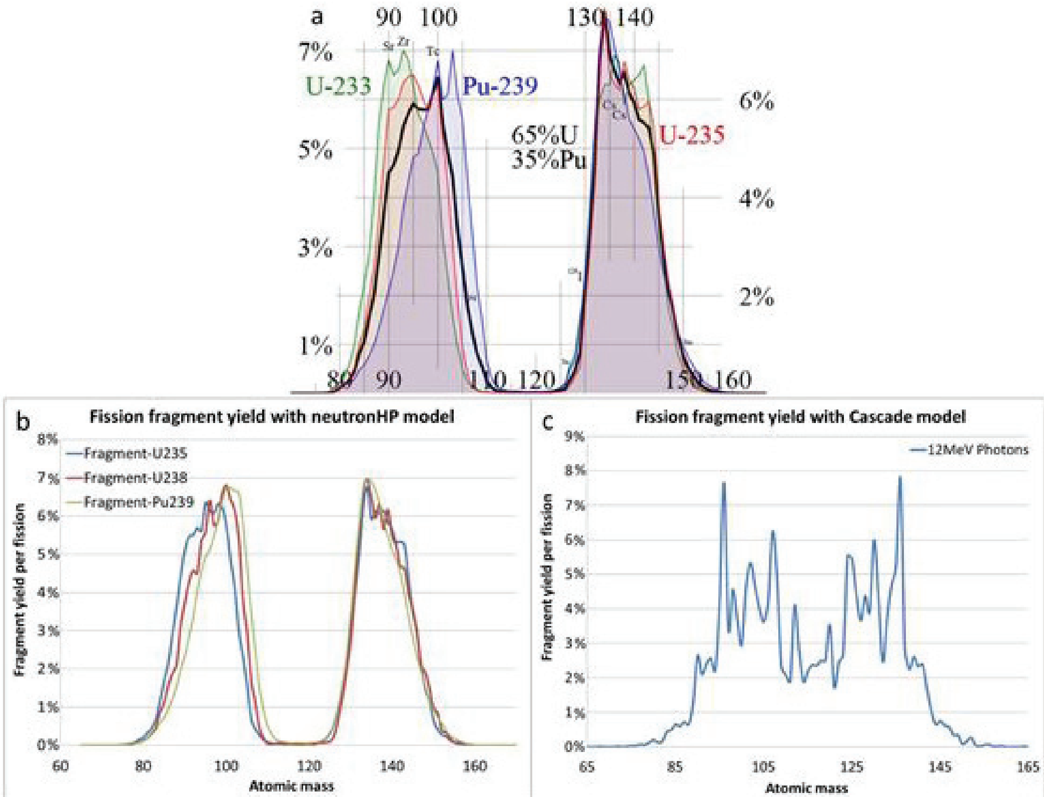

Fig. 2. Fission fragment distribution from Wikipedia in (a) from NeutronHP model in (b) and from Cascade model in (c).

2.3. Delayed gamma rays

The delayed gamma rays are modelled by the radiative decay physics of Geant4, where fission fragments and their daughter nuclei decay with gamma-ray emission. To verify the time properties of delayed gamma rays, we simulated the time spectrum with Geant4 and MCNP and compared with data from Gozani (1981). The time dependence is only sensitive to the target isotope for incident particle energies below 10 MeV. The reference data are from fits to measurements using five exponential decay constants as shown in Table 1 for U-235 and U-238 as Gozani (1981).

Table 1: Delayed gamma ray yield for neutron induced fission and photofission for energies up to ~10 MeV.

		Abundances(yield/fission)	
Group #	Half time (s)	U-235	U-238
1	0.29	0.067	0.54
2	1.7	1.05	1.76
3	13	1.92	2.81
4	100	1.73	2.02
5	940	1.55	1.55

The simulated time dependence of the delayed gamma rays generated by the NeutronHP and Cascade models are displayed in Fig. 3. The NeutronHP model generated the correct fragment yield (but wrong multiplicity as section 2.1), and the corresponding time curve agrees with the reference. However, the time spectrum of the Cascade model shows a significant discrepancy, which is explained by the wrong fragment distribution (Fig. 2b) that results in the wrong distribution of the decaying nuclei.

In addition, the energy dependence of the delayed gamma rays is sensitive to the characteristic energies of gamma-ray emission in fragment decay and the distribution of the fission fragments. The spectra of delayed gamma rays are complex since all generated fission fragments contribute to them.

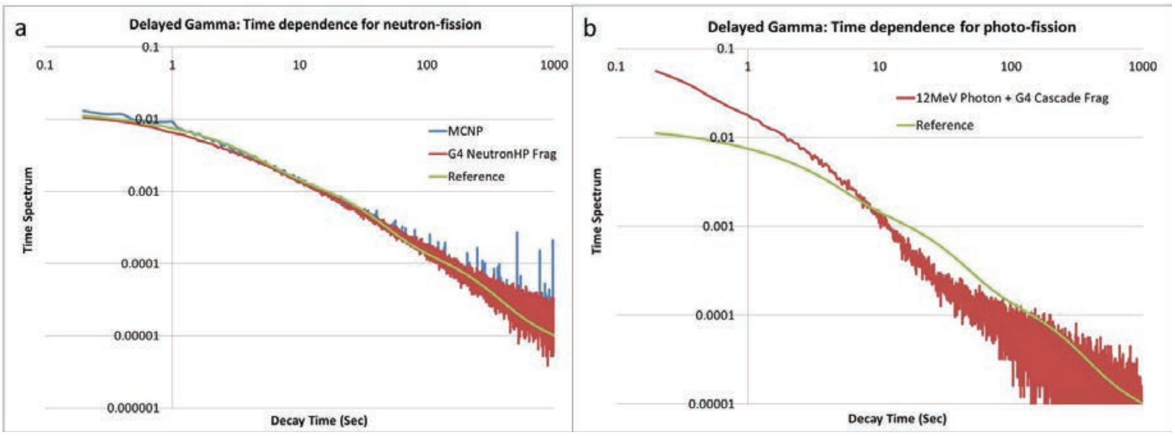

Fig. 3. Time dependence of delayed gamma rays produced in fission induced by thermal neutrons in (a) and 12MeV photon in (b).

2.4. Delayed neutrons

Geant4 (versions.10.1 and below) does not support delayed neutron generation because the data for the neutron decay channel of fragments are not available in the existing Geant4 code or libraries. Our new implementation of neutron emission, described in section 3.4, was added to the radioactive decay physics package to complete the delayed radiation.

2.5. Cross section of fission models

The cross sections used by Geant4 for the fission rates induced by neutrons and photons were investigated. It was found that the neutron-induced fission cross-section library used by the NeutronHP model is taken mainly from the ENDF/B-VII library, but with different energy bins.

Unlike the fission process modeled in NeutronHP, the photofission does not use a cross-section file. The photofission process, which uses the Cascade model, has hard-coded values that seem to originate from the model by Geant4 physics reference manual Chapter 46 and Wellisch (2003). The photofission was implemented as one branch of the photonuclear reaction that starts with photon absorption and is followed by the competition among neutrons, light particle evaporation and fission of the excited nuclei. The simulated photofission rates as a function of photons incident energies show that the estimated cross sections derived from the fission rates were lower compared with the data from ENDF/B-VII (see Fig. 8).

3. Modification of fission models in Geant4

The Geant4 fission NeutronHP and Cascade models were modified to correct the inaccuracies described in the previous section. The demonstrated Lawrence Livermore National Laboratory (LLNL) fission model (Verbeke, 2014) was integrated to sample the prompt radiation multiplicity and energy distributions of the neutron- and photon-induced fissions. The other detailed characteristics of fission products were modified to match libraries, including fragment distribution, the time dependence of delayed gamma rays, implementation of delayed neutrons and fission cross sections. The modifications were verified with simulation performed with the U-235, U-238 and Pu-239 isotopes.

3.1. Multiplicity from LLNL fission model

The multiplicity distribution of LLNL fission model was compared with results from the original Geant4 models. The multiplicity of both prompt neutrons and gamma rays from fission induced by thermal neutrons in U-235 are shown in Fig. 4a and 4b. The LLNL model reproduced the distributions described in section 2.

The multiplicity resulting from photofission in the Cascade model was replaced by the LLNL neutron-induced fission model under the assumption that the nuclei will fission in the same way, independent of the type of excitation particle. Therefore, we created a “neutron-equivalent” isotope with its corresponding excitation energy before accessing sampling functions of prompt radiation in neutron-induced fission. When a target nucleus captures an incident neutron, the nucleus has an additional neutron compared when capturing a photon. Therefore, the same fission data for neutron-induced fission for an isotope that has one fewer neutron can be used. In addition, the neutron separation energy of the neutron in the nucleus is subtracted from the excitation energy to account for the extra energy that the incident neutron brings in as it is captured by the isotope that undergoes fission.

The average multiplicity of 12 MeV photofission simulated by the modified Cascade model in U-235 is shown in Fig. 4c and 4d. The results are in agreement with the accepted values presented in section 2.1.

3.2. Energy spectra of prompt radiation

The fission spectra of prompt neutrons in the LLNL model follow the Evaluated Nuclear Data Library (ENDL). The spectra were fitted to the Watt expression (1) by Lestone (2007), which contains weakly-varying parameters a and b associated with the fission isotopes and their incident energy. As Fig. 5a and 5c show, the simulated spectra are indeed consistent with the Watt spectra.

$$f(E) = C \cdot e^{-E/a} \cdot \sinh(\sqrt{b \cdot E}) \tag{1}$$

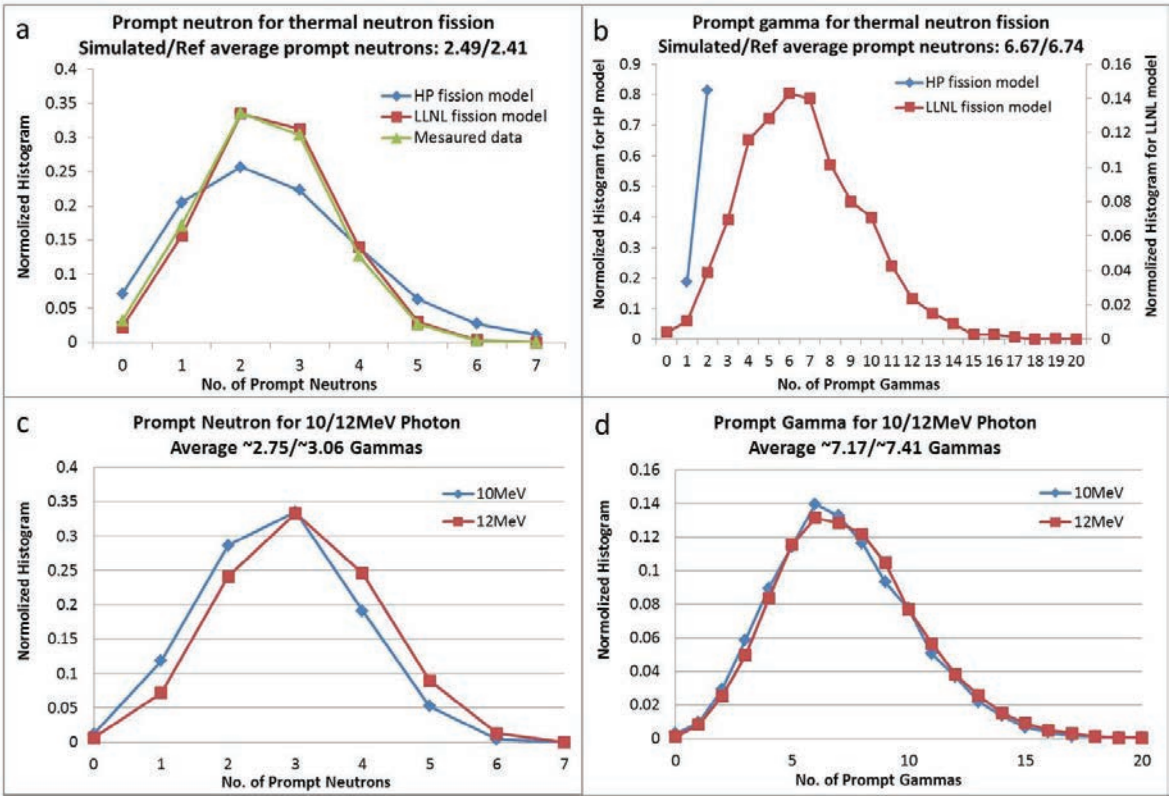

Fig. 4. Multiplicity of prompt radiation for neutron/photon induced fission by modified fission models.

Valentine (2001) fit the prompt gamma-ray spectrum from U-235 fission induced by thermal neutrons to the analytic expression shown in equation (2), which was implemented in the LLNL fission model. The prompt gamma-ray of the original Geant4 simulations, LLNL model is compared in Fig 5c and 5d, matching expression (2). Valentine stated that the spectra of prompt gamma rays from different isotopes are similar, so it has been implemented for all isotopes.

$$f(E)=\begin{cases} 38.13\cdot(E-0.085)\cdot e^{1.648E} & E<0.3MeV \\ 26.8\cdot e^{-2.30E} & 0.3\leq E<1.0MeV \\ 8.0\cdot e^{-1.1E} & 1.0\leq E<8.0MeV \end{cases} \tag{2}$$

The energy of both prompt neutrons and gamma rays are sampled independently with no energy conservation per event, energy conservation is satisfied in average.

3.3. Delayed gamma rays

As mentioned earlier, the time dependence of the delayed gamma rays following fission depends on the atomic number distribution of the fragments. As shown in section 2.2, the NeutronHP used in the neutron-induced fission generates the correct fragment distribution, while the Cascade model used in photofission does not. Therefore, the NeutronHP fragment generation was implemented in the photofission model. In this way, the modified fission process induced by neutrons and photons share the same sampling methods to produce prompt radiation and fission fragments.

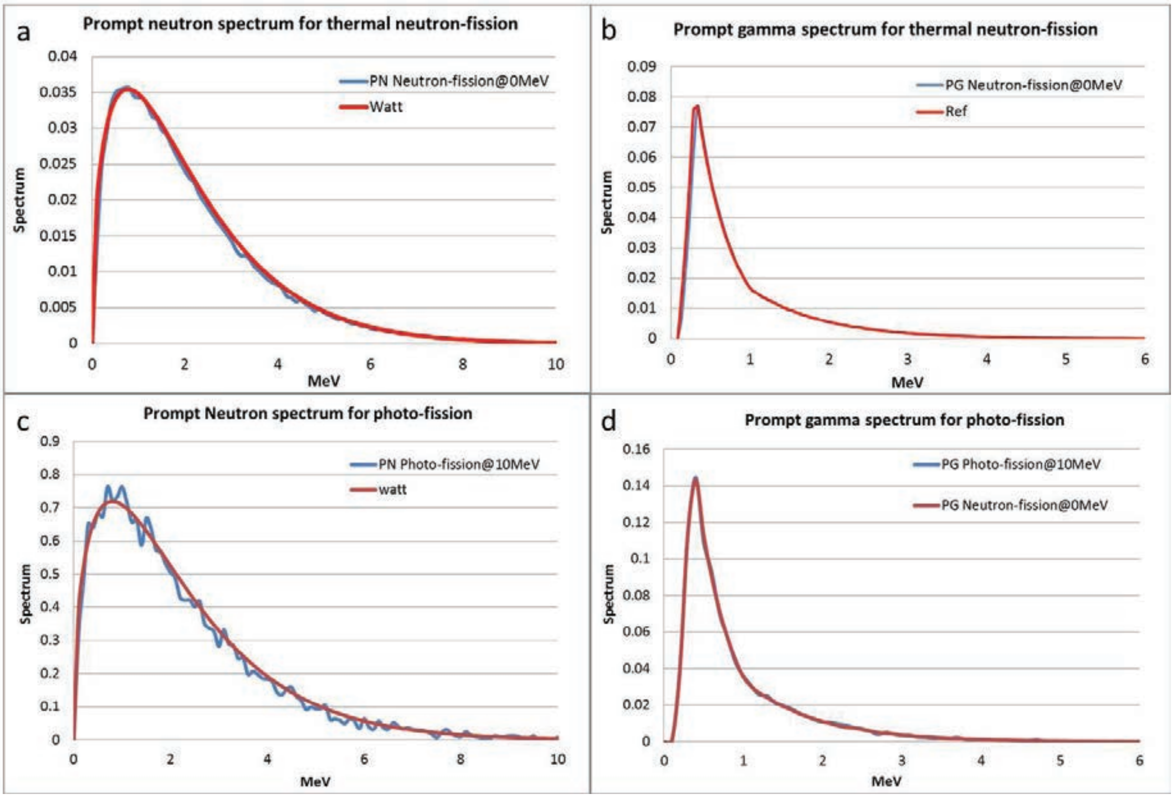

Fig. 5. Simulated energy spectra of prompt radiation by modified fission models.

As shown in Fig. 6a and 6b, the time dependences of delayed gamma rays in newly implemented neutron induced fission and photofission with correct multiplicity and fragment distribution were modified to reach agreement with the literature (Table 1).

3.4. Delayed neutrons

The default radioactive decay physics in Geant4 is able to produce delayed gamma rays but no delayed neutrons due to lack of Geant4 compatible libraries for neutron decay by fragments and daughter nuclei. Proper implementation of delayed neutrons would require adding neutron decay information to all involved nuclei, which is a daunting task. To complete the delayed radiation in fission models, we implemented delayed neutrons using a simplified method. A “virtual” fragment was created in the secondary fission list, which shares the prompt product generation time and only allows for the decay with one neutron emission. Since the virtual fragment and its daughter isotope are not natural isotopes, their presence does not affect any existing physics. In this approach, different virtual fragments are specified for delayed neutrons from different fissionable isotopes.

The implementation requires the isotope associated data to model the time and energy dependence of neutrons from all yield fission fragments. Similar with time dependence modelling of delayed gamma rays, the time and energy dependence of delayed neutrons can be expressed as a summation of independent exponential distributions as shown in equations (3) and (4) by the Cullen’s model (2004), where C_i, Tk_i and τ_i in equations are isotope associated parameters.

$$I(t) = \sum_i C_i \cdot e^{-t/\tau_i} \tag{3}$$

$$S(E)dE = \sum_i C_i \cdot \left(\frac{E}{Tk_i}\right)^{0.5} \cdot e^{-t/Tk_i} \cdot dE \tag{4}$$

The modified delayed neutron package is shared by neutron- and photon-induced fission. The time-decay constants for U-235, U-238 and Pu-239 and the energy-spectrum for U-235 were included in the modified package. The time and energy dependences of simulated delayed neutrons for U-235 photofission are displayed in Fig. 7 showing good agreement with data in Gozani (1981) and Cullen's model (2004).

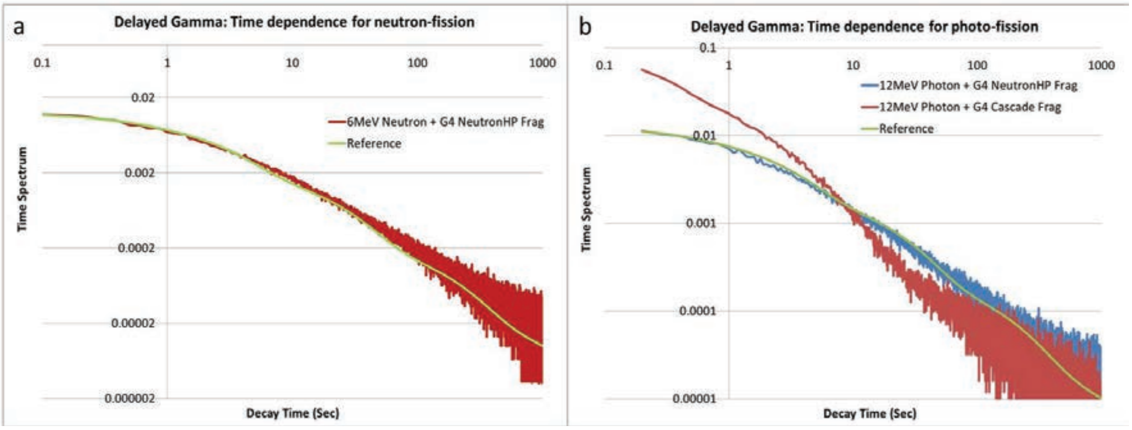

Fig. 6. Time dependence of delayed gamma rays with modified multiplicity and fission fragment yield for photofission.

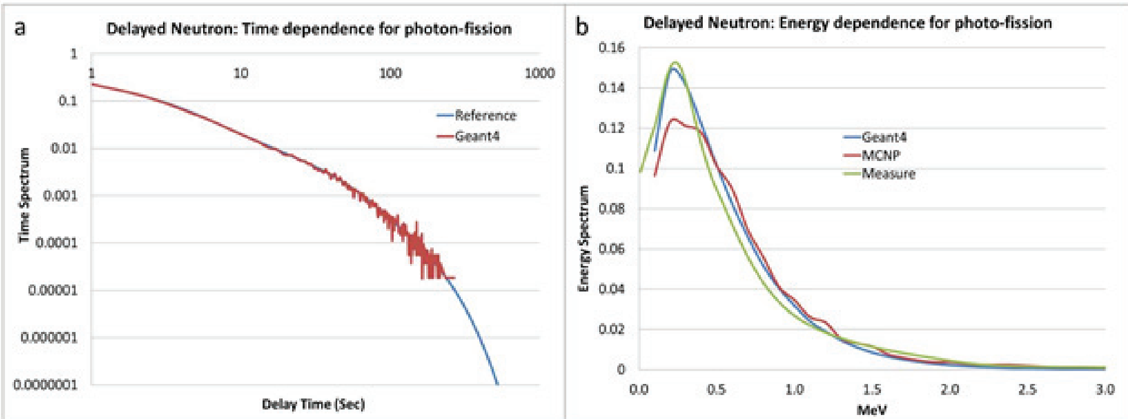

Fig. 7. Simulated delayed neutrons with time and energy dependence in photofission.

3.5. Cross section of fission models

The photofission rate in Geant4 10.1 was found to be lower compared with ENDF/B-VII library. To prevent significant changes to the Geant4 code, we did not modify the photofission model, which is implemented as one branch of the photonuclear reaction. Instead, an incident-energy-dependent factor was applied to increase the probability of the fission branch in photon nuclear reaction to match the fission rate in ENDF/B-VII. The modified fission cross section of U-235 photofission derived from the simulations as a function of the energy of incident photons are compared with ENDF/B-VII and the original Geant4 cross section in Fig. 8.

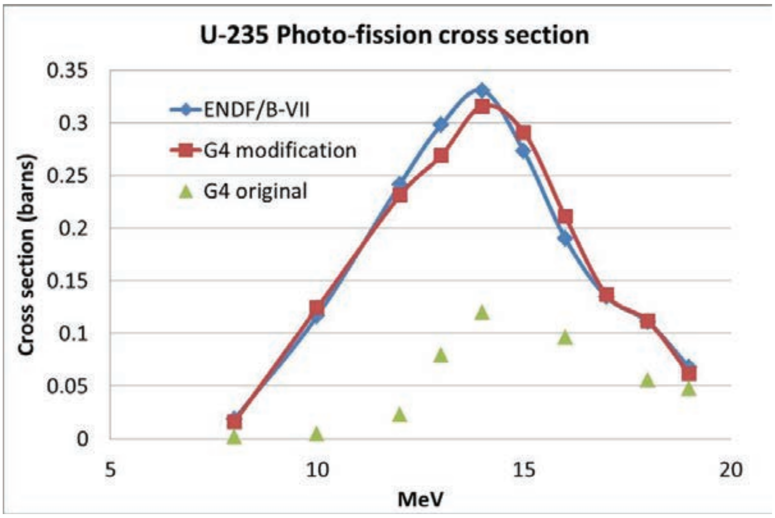

Fig 8. Cross section of photofission as function of energy of incident photons

4. Fission results for U-235,U-238 and Pu-239

The modified fission data and delayed-neutron implementation described in section 3 was extended to U-238 and Pu-239 and verified to agree consistently with the available references for fission products and their characteristics. The simulation for Z≥92 requires an extra data package that does not come with default Geant4 installation. We extended data for Pu by using SLAC nuclear data library provided with distribution restrictions.

5. Summary

In this work, we investigated the fission physics of the Geant4 simulation code for neutron- and photon-induced fission and demonstrated that the existing models and libraries do not produce the correct fission products (multiplicities, fission fragment and delayed radiation). To develop an accurate model, we modified and completed the characteristics of the products from the neutron- and photo-induced fission using the latest evaluated data. The prompt and delayed radiations with their characteristics were verified with corresponding references. Fission simulations of U-235, U-238 and Pu-239 were conducted for model verification, and we are planning to extend simulations to include other isotopes of interest. In this way, users that prefer Geant4, would be able to run accurate fission simulations.

6. Acknowledgments

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