Uranium and Plutonium Fission Product Gamma Intensity Measurements and MCNP6 Simulations

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INTRODUCTION

The fission product gamma ray spectra of irradiated thin HEU and plutonium disks (93.15 at% ^{235}U and 98.97 at% ^{239}Pu, respectively) has been previously measured by Beddingfield and Cecil [1]. They evaluated the intensities of prominent fission product peaks (FPPs) and examined the variation in their intensity ratios arising from ^{235}U vs. ^{239}Pu content. Those experiments were recently simulated by Durkee *et al*. when demonstrating the delayed gamma (DG) capability in MCNP6 [2]. This summary discusses additional comparisons in the same energy range (0.8 – 1.6 *MeV*) between MCNP6v1 and FPP measurements of solutions containing ^{233}U, ^{235}U and ^{239}Pu after a 60 *s* irradiation, 15 *s* decay and 180 *s* count.

DESCRIPTION OF ACTUAL WORK

A Description of the Experiment

Samples containing between 1.32 and 4.27 *μg* of fissile content were prepared from certified reference materials (Nat U. CRM421C, NIST, Gaithersburg, MD, CRM 111-A 99.4911 at% ^{233}U, New Brunswick Laboratory, and Isotope Products Laboratories Lot #1195.20, 97.937 at% ^{239}Pu, Eckert & Ziegler, Valencia, CA) in aqueous form. Solutions were doubly encapsulated in polyethylene vials before their 60 *s* irradiation in a predominately thermal SLOWPOKE-2 reactor. A counting arrangement consisting of a HPGe detector (GMX 18190, SH-GMX CFG:S/N 26-N1476A) and six ^{3}He detectors records both gamma and neutron energy depositions as a function of count time. Measurement procedures and the counting arrangement have been described in detail in Ref. 3. The HPGe detector was calibrated with a multi-nuclide standard source containing 13 isotopes with energies ranging from 47 *keV* to 1.836 *MeV* (Source 1423-99-21, Eckert & Ziegler, Valencia, CA).

MCNP6 Simulation

Irradiation conditions were simulated using an input deck provided by Atomic Energy of Canada Limited containing SLOWPOKE-2 geometry and material specifications [4]. This model was modified to include a vial containing an aqueous solution in an inner irradiation site. A flux (F4) tally was placed in the vial to determine the neutron energy group distributions. This flux profile was then recreated in a second deck with only a PE vial containing a fissile solution. The *DG=lines* option in MCNP6 simulated DG emissions; these energy and time of emissions were recorded by a surface current (F1) tally placed on the exterior of the vial. Time and energy binning parameters were chosen to correspond to experimental irradiation, decay and count times, and channel width. Gamma emissions up to 4 *MeV* were recorded.

DG detection was simulated by modeling the counting arrangement and recreating gamma energy distributions (obtained from the aforementioned F1 tally), from the vial position within the counting geometry. A pulse height (F8) tally was placed in the active zone of the detector, which recorded photon energy depositions. Energy resolution effects were recreated via use of the *Gaussian Energy Broadening (GEB)* card in MCNP6. Current simplifications in the simulations include the omission of ^{238}U content contributions in the simulations containing natural U.

MCNP6 DG production is explained in detail in Ref. 2. DG options available in MCNP consist of multigroup (MG) and line data, which are based on ENDF/B-VI evaluations. When *DG=lines* option is selected (as is the case in this work) the resulting DG emission will be comprised of discrete lines (currently available for 979 radionuclides in the gamma line data file *cindergl.dat* released with MCNP6v1) and continuous data evaluated at 10 *keV* intervals. Current efforts at Los Alamos National Laboratory include the development of a code ENDF2CINDER, which updates these files with ENDFVII data [5]. Gamma line emission files containing ENDFVII.1 library data were provided by T. Wilcox at LANL and were also used in this comparison.

RESULTS

Post Processing of MCNP Output and Measurements

Measurements and their associated MCNP pulse height tally outputs were imported into a Matlab™ script, which corrects for both dead time effects and background spectra contributions in the former. A Savitzky-Golay filter is applied to smooth the datasets before the algorithm identifies peaks in both spectra, and selects the most prominent experimental peaks for further comparison. An example of spectra analyzed in this work is shown in Fig. 1. Cumulative counts are normalized by total fissile mass in both measurements and simulations.

HPGe Detector Properties

MCNP6 simulations and measured waveform spectra after a 10 *min* count of the multi-nuclide standard provided a comparison of intrinsic efficiencies. MCNP6 simulations were found to over predict measured intrinsic efficiencies by 132 ± 10 *%*, with no observed energy dependence from 47 *keV* to 1.836 *MeV*. It is likely that dead layer growth in this older detector has contributed to a decrease in observed intrinsic efficiency [6]. Examination of the measured waveform also allowed for a determination of energy resolution broadening effects which were simulated in MCNP6 with a *GEB* card.

All MCNP6 outputs were normalized by the difference in intrinsic efficiencies and simulated fissile mass. Preliminary comparisons of FPPs from irradiated fissile material showed a considerable difference in observed spectra and MCNP6v1 output, Figure 1 (top). This is possibly due to the omission of > 4 *MeV* gamma reproduction in the counting MCNP6 deck. These energetic gamma rays would have a considerable Compton continuum contribution to energy deposition. A more direct comparison of fission product peak intensities was facilitated by the subtraction of local minima counts from both spectra, Figure 1 (bottom).

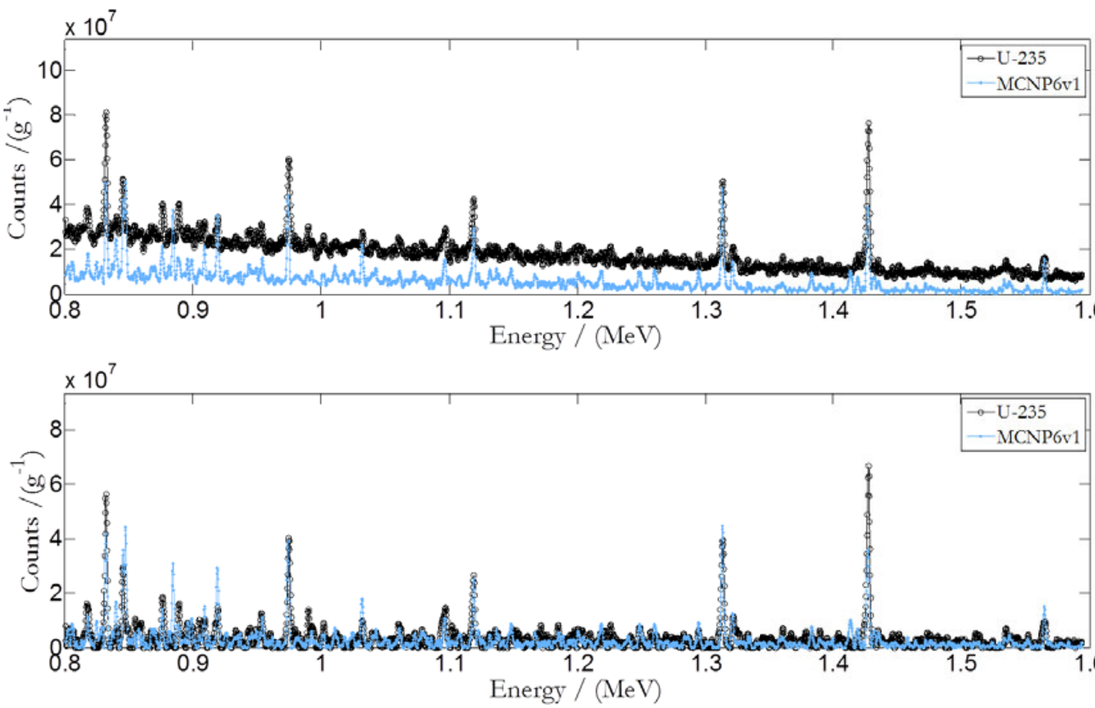

Figure 1: ^{235}U Measurements and MCNP6v1 without (top) and with (bottom) Compton continuum subtraction.

A Comparison of Fission Product Peak Intensities

Significant differences between MCNP6v1 and measurement FPP magnitudes, namely $^{90/90m}$Rb and $^{132/132m}$Sb, have been previously noted [3]. Several of these discrepancies could be attributed to the older ENDFVI data called upon in MCNP6v1 for delayed gamma emission simulations. Therefore updated gamma line files with ENDFVII.1 data were also used in simulations.

Figure 2 shows MCNP6 and measured cumulative counts for three samples, a ^{233}U solution (top), a mixture with the fissile content comprised of 40 wt *%* ^{239}Pu and 60 *%* ^{235}U (contained in Nat. U), and a natural uranium solution (bottom). Figure 2 also depicts the MCNP6 simulation results, which used updated gamma line (GL) files, however identical runs using default files provided with the current MCNP6v1 release were also examined.

Table 1 summarizes observed peak magnitudes and those predicted by MCNP6v1 (with and without updated gamma line data files) for the 10 most prominent peaks in each spectra. The peak intensities in each case were determined by summing the peak channel and the 4 adjacent channel counts, in both simulations and experiments. 68 % confidence intervals were calculated via the propagation of flux magnitude, counting statistics and intrinsic efficiency correction uncertainties. Results within the 95 % confidence interval have been shaded.

MCNP6 predicted the presence of all but one prominent peak for each spectra. This peak, at 1.145 *MeV,* was observed during several fissile irradiations (but not all). It is not believed to be a fission product peak. FPPs whose cumulative channel counts were less than 400 had high absolute errors, 38 ± 19 %. Updates to the cinder gamma line data files resulted in improved agreement for the previously noted discrepancies $^{90/90m}$Rb and $^{132/132m}$Sb.

A final comparison examined the intensity ratios of the three most prominent peaks in measured spectra: ^{94}Sr (1.428 *MeV*), $^{90/90m}$Rb (0.832 *MeV*) and $^{132/132m}$Sb (0.975 *MeV*). These ratios for each solution and the corresponding MCNP6 output are shown in Table 2. In each of the 9 cases the update to the gamma line files resulted in a lower absolute error between MCNP6 simulations and measurements. MCNP6v1 (with modifications to the gamma line data files) was able to predict these ratios within the 95 % confidence interval in 7 of the 9 comparisons, Table 2. There is no dependence of relative error on peak energy (in this 0.8 - 1.6 *MeV*) range, however a notable dependence on total cumulative counts is evident.

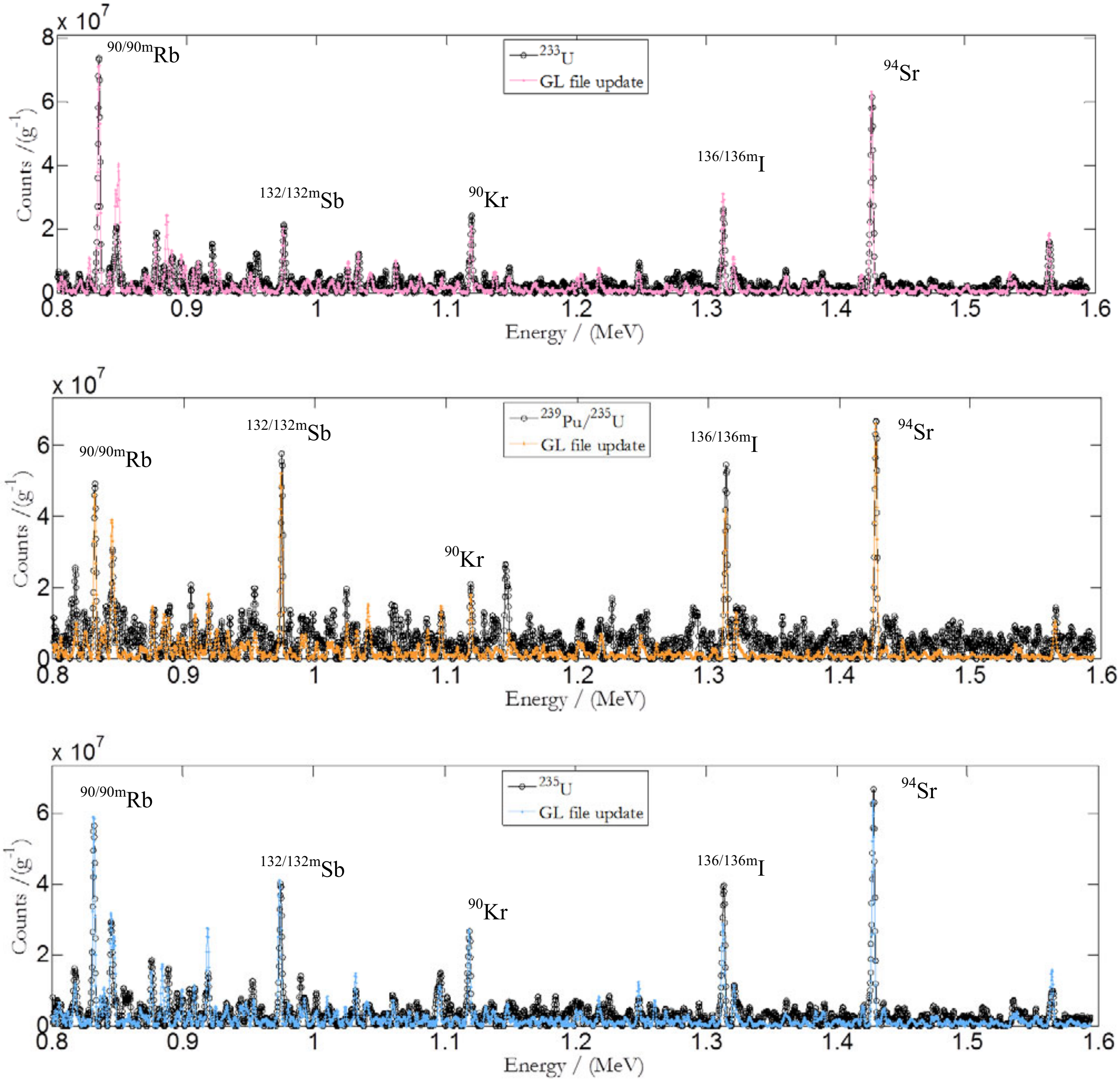

Figure 2: MCNP6 & Measured Spectra for ^{233}U (top), ^{239}Pu/^{235}U (middle) and ^{235}U (bottom) solutions.

Table 1: MCNP : Measured Ratios of Prominent Peaks (± 68 % Confidence Intervals). Results within the 95 % confidence interval have been shaded. FPPs with experimental cumulative counts exceeding 400 have been bolded.

^{235}U Sample			^{233}U Sample			$^{239}Pu/^{235}U$ Mixed Sample		
Energy / (MeV)	MCNP6v1	Updated *GL* files	Energy / (MeV)	MCNP6v1	Updated *GL* files	Energy / (MeV)	MCNP6v1	Updated *GL* files
1.428	**0.89 ± 0.06**	**0.95 ± 0.07**	0.832	**0.51 ± 0.04**	**0.95 ± 0.07**	1.428	**0.93 ± 0.07**	**0.99 ± 0.08**
0.832	**0.49 ± 0.04**	**1.03 ± 0.07**	1.428	**1.01 ± 0.07**	**1.03 ± 0.07**	0.975	**0.68 ± 0.05**	**0.93 ± 0.07**
0.975	**0.83 ± 0.06**	**1.00 ± 0.07**	1.314	**1.36 ± 0.10**	**1.20 ± 0.09**	1.314	**0.87 ± 0.07**	**0.79 ± 0.06**
1.314	**0.90 ± 0.07**	**0.70 ± 0.05**	1.119	**0.87 ± 0.07**	**0.87 ± 0.07**	0.832	0.51 ± 0.04	0.94 ± 0.07
0.846	**1.11 ± 0.08**	**1.05 ± 0.08**	0.975	**0.71 ± 0.05**	**0.92 ± 0.07**	0.846	1.16 ± 0.10	1.31 ± 0.11
1.119	**0.87 ± 0.07**	**1.01 ± 0.08**	0.846	**1.26 ± 0.10**	**1.52 ± 0.12**	1.145	-	-
0.876	**0.64 ± 0.05**	**0.82 ± 0.06**	0.876	**0.68 ± 0.05**	**0.87 ± 0.07**	0.817	0.35 ± 0.03	0.41 ± 0.03
0.817	0.62 ± 0.05	0.71 ± 0.06	1.565	**1.13 ± 0.09**	**1.14 ± 0.09**	1.119	0.66 ± 0.06	0.90 ± 0.07
0.889	0.84 ± 0.06	0.63 ± 0.05	0.919	0.65 ± 0.05	0.61 ± 0.05	0.954	0.40 ± 0.03	0.40 ± 0.03
1.097	0.77 ± 0.06	0.76 ± 0.06	0.953	0.65 ± 0.05	0.35 ± 0.03	0.905	0.33 ± 0.03	0.28 ± 0.02

Table 2: Ratios of ^{94}Sr, $^{90/90m}Rb$ and $^{132/132m}Sb$ Intensities: Measurements & MCNP6v1 (± 68 % CI).

	^{235}U Sample			^{233}U Sample			$^{239}Pu/^{235}U$ Mixed Sample		
Ratio	Exp.	MCNP6	Updated *GL* files	Exp.	MCNP6	Updated *GL* files	Exp.	MCNP6	Updated *GL* files
^{94}Sr:^{90}Rb	1.21 ± 0.09	1.66	1.12	0.85 ± 0.06	0.70	0.92	1.40 ± 0.12	1.83	1.56
^{94}Sr:^{132}Sb	1.65 ± 0.13	1.76	1.57	2.91 ± 0.23	1.54	2.05	1.20 ± 0.10	1.33	1.28
^{90}Rb:^{132}Sb	1.36 ± 0.10	1.06	1.40	3.41 ± 0.26	2.19	2.22	0.86 ± 0.08	0.73	0.82

CONCLUSIONS & FUTURE WORK

This summary discusses comparisons between MCNP6v1 and the measurement of fission product gamma intensities in the 0.8 – 1.6 *MeV* range. Several discrepancies in FPP measured intensities and MCNP6v1 predictions were resolved with the use of gamma line data files populated with ENDFVII.1 library data. For example, ^{235}U $^{90/90m}Rb$ and $^{132/132m}Sb$ MCNP to measured peak intensity ratios increased from 0.51 ± 0.04 to 1.03 ± 0.07, and 0.71 ± 0.05 to 0.92 ± 0.07, respectively.

Future work will see the continuation of these comparisons with varying irradiation, decay, and count times. Sample irradiations and counting will be performed in triplicate to reduce stochastic uncertainties in measured spectra. Also, FPP intensity comparisons with energies ranging from 10 *keV* to 2 *MeV* will be examined.

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