USE OF A SLOWPOKE-2 REACTOR FOR NUCLEAR FORENSICS APPLICATIONS

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Abstract

A low enriched uranium SLOWPOKE-2 reactor is used as a neutron interrogation source in support of the identification and characterization of Special Nuclear Materials (SNM) at the Royal Military College of Canada (RMCC). Small amounts of fissile uranium and plutonium are sent into a SLOWPOKE-2 irradiation site before their transport to RMCC's delayed neutron and gamma counting (DNGC) system. The counting arrangement of the DNGC consists of an array of six ^{3}He and a high purity germanium detector. These detectors record the delayed neutron and photon emissions as a function of count time, to verify MCNP6 simulations of delayed particle emissions, and to detect and quantify trace amounts of fissile content. This paper discusses MCNP analyses done in preparation for an upcoming nuclear forensics exercise in the fall of 2014. MCNP6 simulations of the DNGC system focussed on the identification of characteristic gamma lines from prominent fission products. The relative intensities of these gamma lines are dependent on the SNM content in the sample. Gamma line pairs useful for SNM identification in RMCC's DNGC system are presented.

1. Introduction

The application of science to law is the primary goal of forensics studies; nuclear forensics applies forensics methodologies to nuclear material-related crimes [1]. A variety of tools are available for forensic scientists to identify and assess crimes involving special nuclear materials (SNMs). If SNMs are seized, determining their composition and identifying their source will aid in the prosecution and prevention of crimes [1]. In order to achieve continual improvements in best practice, the Nuclear Forensics International Technical Working Group conducts Collaborative Materials Exercises to evaluate and improve analytical techniques and instrumentation for nuclear material assay [2]. The Royal Military College of Canada (RMCC) will collaborate with Atomic Energy of Canada Limited and other Canadian institutions in the next Collaborative Materials Exercises, scheduled for the fall of 2014. In the Collaborative Materials Exercises, laboratories in participating countries determine the isotopic composition and source of SNM samples, and provide statistically-based statements to aid attribution.

When the Collaborative Materials Exercises sample is delivered to RMCC, the measurement capabilities of the delayed neutron and gamma counting (DNGC) system will be used to interrogate the sample after it has been irradiated in the SLOWPOKE-2 research reactor on site. The DNGC system consists of six ^{3}He detectors and a high purity germanium (HPGe) detector arranged around a central polyethylene sample holder [3]. Samples are encapsulated in polyethylene vials, sent via pneumatic tubes to a SLOWPOKE-2 irradiation site for a 60 *s* irradiation, before their transfer to the counting system containing neutron and gamma detectors.

The Collaborative Materials Exercises sample is known to contain enriched uranium with ^{235}U comprising < 10 % of the total U amount. Preparation for the evaluation of this sample also

includes the consideration of samples with may have been irradiated or mixed with other fuel types. Therefore the emissions of additional actinides including ^{233}U, ^{239}Pu, and ^{238}U are also considered in preparation studies. Differences in the fission product yields, displayed in Figure 1, can be used for nuclear forensics applications. For example, fission products emit delayed gamma rays and neutrons following β^- decay, the former with characteristic energies specific to the emitting fission product. The magnitude of neutron and gamma emissions, and the relative intensities of fission product's characteristic gamma rays can be used to identify and characterize SNM content [4].

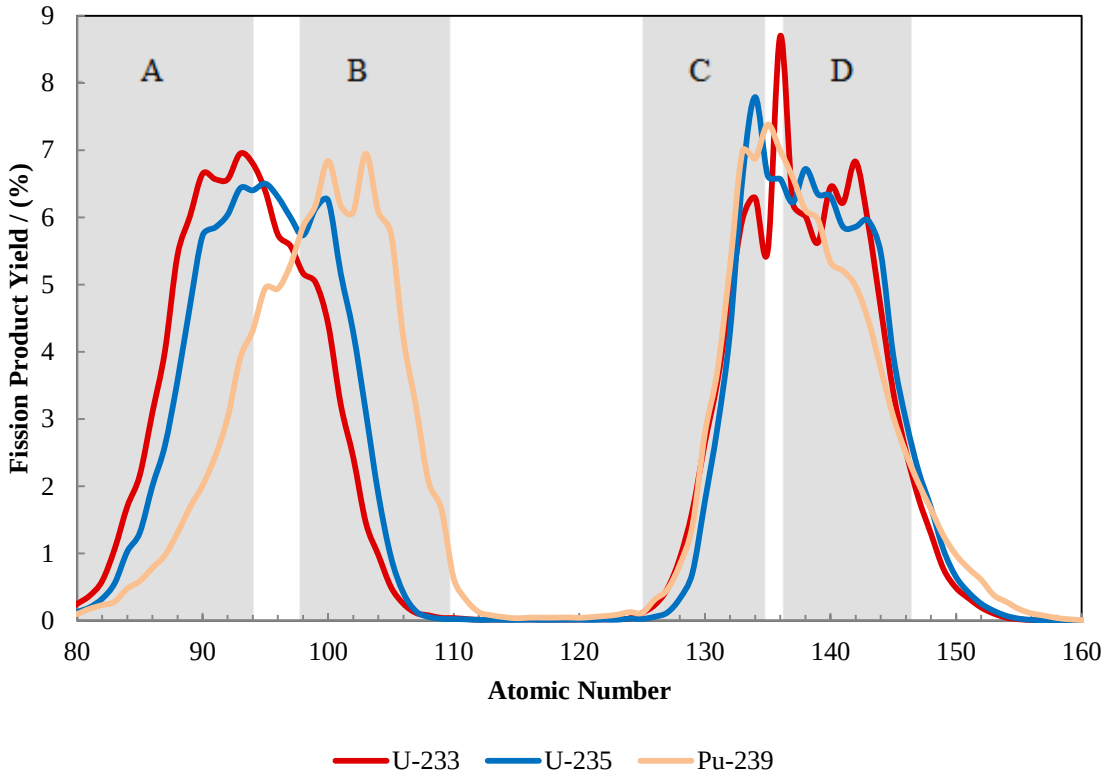

Figure 1: Fission product yield curves for ^{233}U, ^{235}U, ^{239}Pu, and ^{235}U-^{239}Pu Mixture

2. MCNP6 Simulations in support of the Collaborative Materials Exercise at RMCC

In preparation for the receipt of SNM samples, the Monte Carlo simulation software MCNP6 was used to simulate the delayed neutron and delayed gamma emissions from irradiated samples of isotopic composition and uranium enrichment ranges relevant to the Collaborative Materials Exercise. These MCNP simulations were performed in two steps. The first simulation reproduces the 60 *s* irradiation of an aqueous solution containing *mg* quantities of SNM in the SLOWPOKE-2, and the irradiated solution's subsequent delayed neutron and delayed gamma decay. Delayed neutrons and delayed gammas are counted for 10 *mins* after the end of the irradiation using surface current tallies at the edges of the vial. The reactor's flux energy distribution was obtained using a SLOWPOKE-2 reactor model provided by Atomic Energy of Canada Limited (AECL) [5,6]. The delayed gamma emissions during the counting period are

used as photons in source emissions for the second input deck, which models the propagation of a sample's emissions within the DNGC geometry. The gamma ray energy spectrum within the DNGC is modelled using pulse-height tallies, which allows the reproduction of HPGe energy-dependent resolution effects throughout the active volumes of the detectors.

The latest public release of MCNP, MCNP6.1 [7], was used with two modifications. The first modification is a delayed bin fix (DBF) in the MCNP executable, MCNP6DBF, and which contains time bin structure updates for delayed particles. The time bin structure update corrects an issue known to exist in MCNP6.1 [8], which has also been addressed in the next scheduled MCNP release, MCNP6.1.1 [8]. The second modification includes the update of the delayed gamma libraries sampled by MCNP6, which are currently based on ENDF/B VI evaluations. These libraries were updated to include the more recent ENDF/B VII.1 data in the modified versions. The simulations using MCNP6DBF will be verified before they are used to predict the gamma line emissions of Collaborative Materials Exercise samples.

Delayed neutron emissions can be treated using three options in MCNP6: DN=library, model, and both [9]. The first option, DN=library, uses ACE (A Compact ENDF) delayed neutron emission data stored in libraries to calculate the number of delayed neutrons and their energies. The data in the ACE libraries loses some time-dependent information because delayed neutron are sorted into six time groups. The second option, DN=model, uses precalculated delayed neutron emission probabilities to determine the energies of emitted neutrons. The time-dependent nature of all fission products are retained when using the DN=model option [10]. The final option, DN=both, uses the DN=library results and the DN=model results when entries are missing from the ACE libraries.

3. Results

3.1 Validation of MCNP6 as an appropriate tool to simulate delayed particle signatures

Although MCNP6 contains three options for delayed neutron emissions (DN=model, library, and both), delayed gammas can only be simulated using the model option. As discussed previously, the version of MCNP6 used to simulate delayed neutron and delayed gamma emissions from the Collaborative Materials Exercise samples uses the modified MCNP6.1 executable MCNP6DBF and CINDER data updated from ENDF/B VI to ENDF/B VII.1 data. Previous work [11] created several test suites, which compared MCNP6 simulations to delayed neutron measurements at RMCC. These test suites were used in this work to confirm that the DBF executable removes the anomalous delayed particle emissions at count times greater than 100 *s*, shown for ^{233}U delayed neutrons in Figure 2. Also, the test suite simulations confirmed that inclusion of updated data files does not affect delayed neutron simulations, Figure 3. The MCNP6DBF simulations were in best agreement with measurements at RMCC in all neutron cases examined. Complementary ongoing studies at RMCC also show that the use of MCNP6DBF with updated gamma line data files results in overall improved agreement between measurements at RMCC and MCNP simulations, Figure 4.

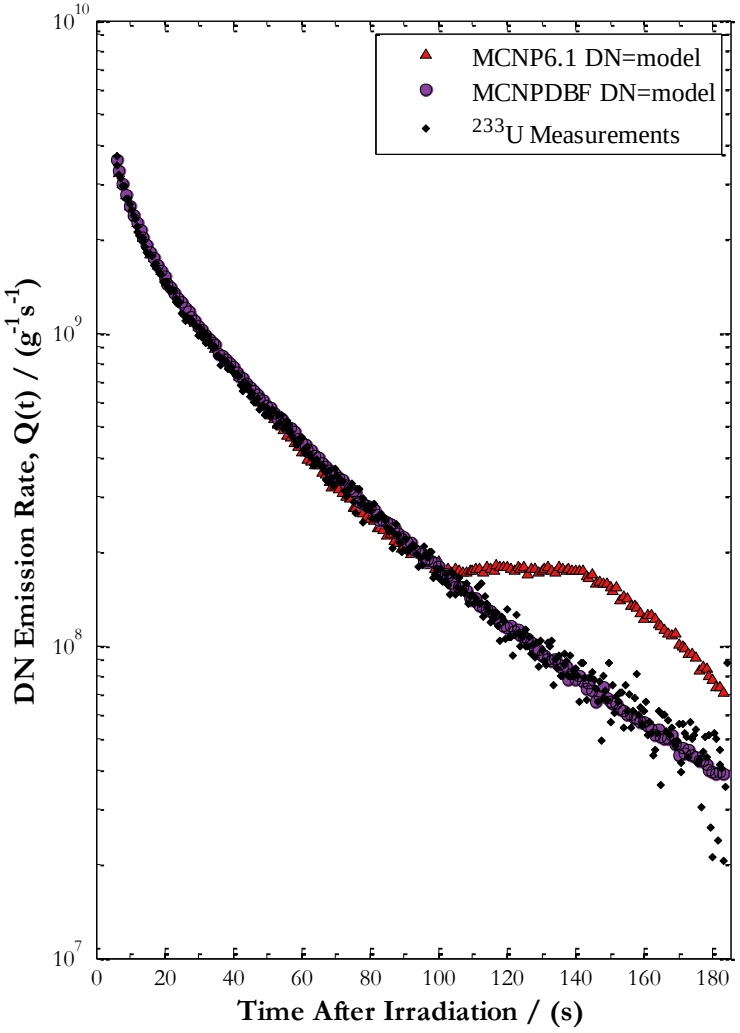

Figure 2: delayed neutron emission rates from ^{233}U measurements and simulations using MCNP6.1 and MCNP6 DBF [11]

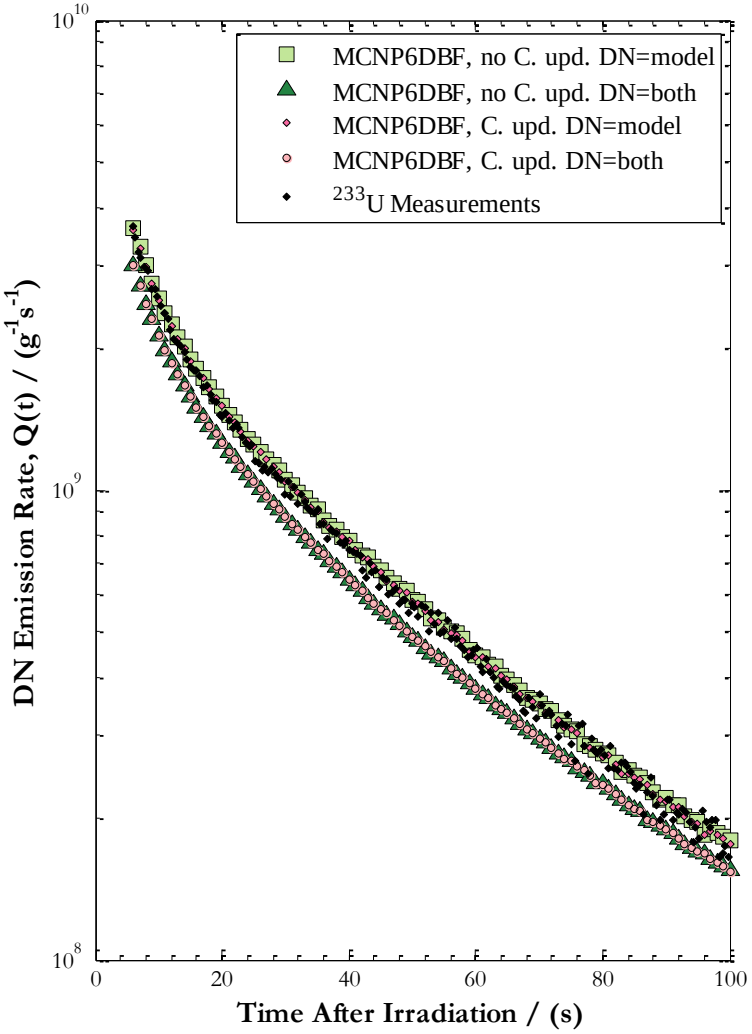

Figure 3: First 100 s of delayed neutron emission rates from ^{233}U measurements and simulations using MCNP6DBF, with updated CINDER files (C. upd.) and without (no C. upd.)

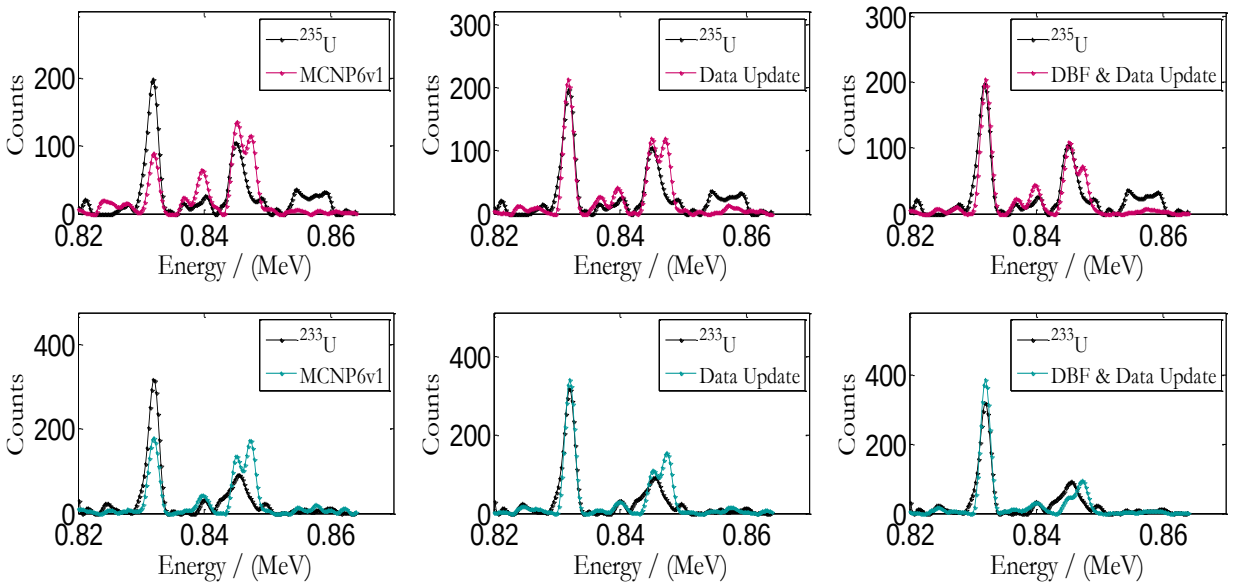

Figure 4: The effects of updating MCNP time bin structure and gamma line data on agreement with RMCC measurements of gamma emission from SNM [12].

Conclusions relevant to Collaborative Materials Exercise preparation can be drawn from the MCNP6 simulations discussed include:

i) Using the updated CINDER files is an improvement over the released CINDER files because their use improves gamma simulations, Figure 4, but does not affect delayed neutron simulations; and

ii) The most accurate model for the measured delayed neutron emission rates is obtained using MCNP6DBF and the DN=model option. The use of the two modifications to MCNP6 is justified because, as shown by the preceding comparisons, they improve upon the simulations using MCNP6.1.

3.2 Application to Nuclear Forensics Research

3.2.1 Delayed Neutron Signatures

Previous work has compared MCNP6 simulations with neutron measurements from the DNGC system at RMCC. It was shown that once a fissile nuclide was identified, its mass could be experimentally determined via the assay of cumulative delayed neutron counts with an average relative error and accuracy of -2.2 and 1.5 %, respectively. However, the characterization of mixtures of fissile content through delayed neutron assay alone at RMCC has proven difficult because the differences between nuclides' delayed neutron temporal behaviours are small and subject to experimental noise. Algorithms comparing the delayed neutron temporal behaviour of binary mixtures to behaviours predicted by analytic models found the fissile mass percentage of single nuclides in the binary mixtures with average absolute errors of ± 4%, but were unable to

conclusively discount the presence of a second nuclide in single-nuclide samples [13]. These analyses required using the average of measurements performed in triplicate, which significantly impacted the speed of nuclear assay. Additionally, due to the high thermal to fast flux ratio of the SLOWPOKE-2 in the outer sites (17.3 +/- 0.5 [14]), ^{238}U will rarely undergo fission, and cannot be detected via delayed neutron counting. Delayed gamma emissions will provide additional information regarding sample content as photon energies can be used to determine fission or activation products. Therefore, both the delayed gamma and delayed neutron signatures will be used to help identify SNM content in the Collaborative Materials Exercise exercise.

3.2.2 Delayed Gamma Signatures

The simulations included samples of pure ^{233}U, ^{235}U, ^{239}Pu, and a 40% ^{235}U- 60% ^{239}Pu mixture. Although gamma energies were recorded between 0 and 2 MeV, only energies between 0.6 and 1.8 MeV were investigated in greatest detail. Spectra in this energy range are less complex, leading to easier identification of fission product peaks. Additionally, more energetic photons are less susceptible to attenuation in the sample and matrix surroundings. Finally, photons with energies > 1.8 MeV have low detection efficiencies in the DNGC system, resulting in decreased signal to noise ratios. The fission product yield curve was categorized into four groups, labelled A, B, C, and D, based on the relative yields of the SNMs in Figure 1. These groups were chosen to identify fission product intensity ratios with the largest variance between individual SNM, for example ^{233}U favours fission products in group A, ^{239}Pu favours those in group B, and ^{235}U straddles both groups.

The gamma ray spectra for the four SNM samples, shown in Figure 5 to Figure 7, were simulated using MCNP6 with the DBF executable, updated gamma line files, and model option for delayed particle emission. Gamma lines were identified by searching nuclide data [15] and a table of gamma ray energies [16]. Three criteria were used to determine whether or not a nuclide was present.

1. Whether peaks in the simulated spectra occurred at that nuclide's prominent gamma ray energies;

2. Whether that nuclide has an atomic mass within the peaks of the fission product yield curve; and

3. Whether that nuclide has an appropriate half-life. so that it decays significantly within the 10 minute counting period.

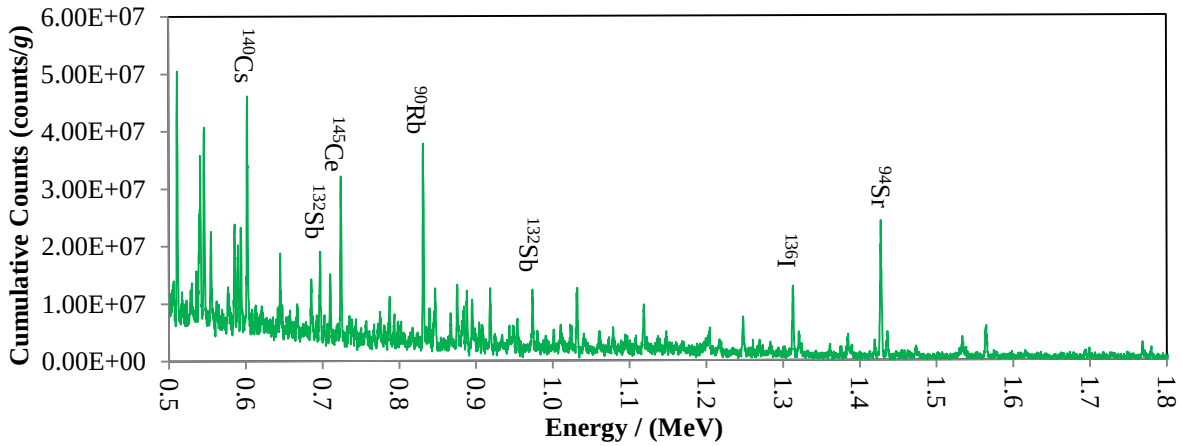

Figure 5: MCNP6 simulation of gamma-ray spectrum for ^{233}U

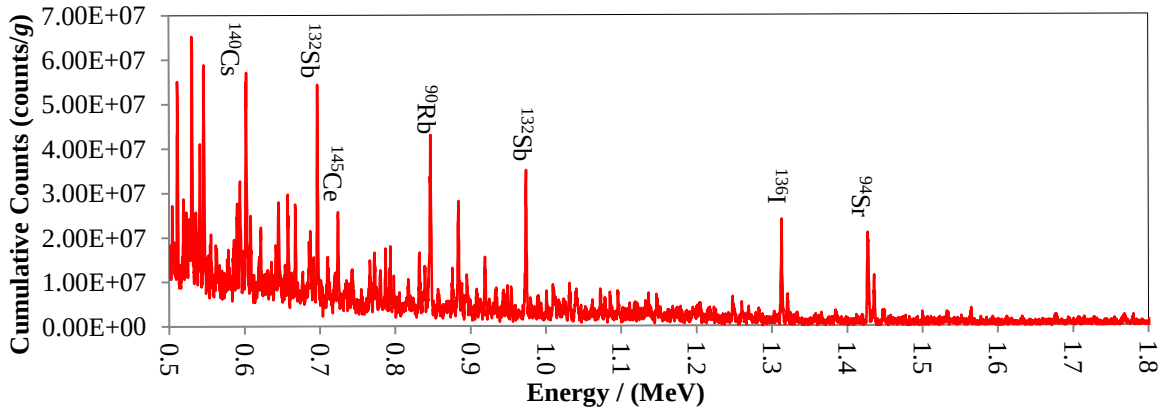

Figure 6: MCNP6 simulation of gamma-ray spectrum for ^{235}U

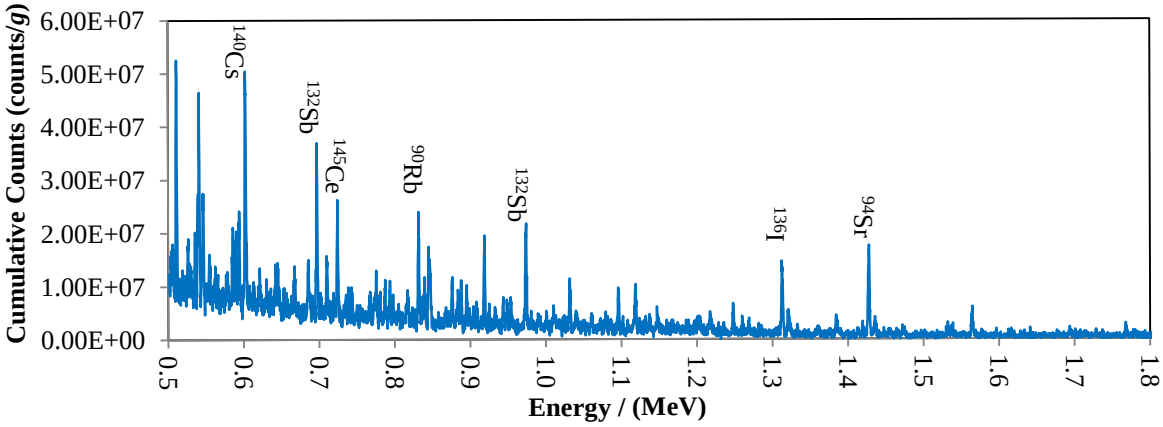

Figure 7: MCNP6 simulation of gamma-ray spectrum for ^{239}Pu

The absolute magnitudes of gamma ray spectra are dependent on many factors, not limited to: a nuclide's fission product yield, half-life, irradiation, decay, and counting times of an experiment, sample density and energy-dependent efficiency and resolution trends. The selection of gamma line pairs for identification, rather than individual lines, eliminates many of the systematic errors in magnitudes when measurements are compared to MCNP simulations. To limit the effects of energy-dependent efficiencies in the detector, line pairs were constrained to be within 400 keV of one another. Gamma pairs are chosen for intensity ratios such that an nuclide in group A or B, whose gamma intensity varies greatly in the four SNMs, is compared to an nuclide in group C or D, whose gamma intensity varies little in the four SNMs. Thus ratio differences are primarily dependent on the differences between SNMs observed on A and B. ^{233}U has the highest yield of the SNMs in group A and ^{239}Pu has the lowest, therefore A/C and A/D intensity ratios are expected to be highest for ^{233}U and lowest for ^{239}Pu. Conversely, the ^{233}U and ^{239}Pu yields are reversed in group B, resulting in a reverse in expected B/C and B/D intensity ratios. The line pair intensity ratios for the four SNMs are displayed in Table 1.

Table 1 Gamma line pair intensity ratios for ^{233}U, ^{235}U, ^{239}Pu, and ^{235}U-^{239}Pu mixture

fission product yield groups A/C					
Fission Products	Line Pair (keV)	^{233}U (rel. err.)	^{235}U (rel. err.)	^{239}Pu (rel. err.)	^{235}U-^{239}Pu Mixture (rel. err.)
^{93}Sr/$^{132/132m}$Sb	710/975	0.82 (0.05)	0.35 (0.02)	0.20 (0.06)	0.30 (0.06)
^{90}Rb/^{132}Sb	832/975	2.86 (0.04)	0.78 (0.02)	0.10 (0.04)	0.39 (0.07)
^{89}Rb/^{136}I	1032/1313	0.65 (0.04)	0.42 (0.02)	0.26 (0.03)	0.24 (0.07)
fission product yield groups B/D					
Fission Products	Line Pair (keV)	^{233}U	^{235}U	^{239}Pu	^{235}U-^{239}Pu Mixture
^{104}Tc/^{144}La	358/397	0.08 (0.04)	0.09 (0.03)	0.74 (0.02)	0.43 (0.04)
fission product yield groups C/D					
Fission Products	Line Pair (keV)	^{233}U	^{235}U	^{239}Pu	^{235}U-^{239}Pu Mixture
^{132}Sb/^{140}Cs	697/602	0.46 (0.05)	0.96 (0.02)	1.14 (0.02)	0.92 (0.05)
fission product yield groups A/D					
Fission Products	Line Pair (keV)	^{233}U	^{235}U	^{239}Pu	^{235}U-^{239}Pu Mixture
^{90}Rb/^{140}Cs	832/602	1.51 (0.03)	0.84 (0.02)	0.13 (0.04)	0.39 (0.06)
^{90}Rb/^{145}Ce	832/723	1.64 (0.04)	1.10 (0.02)	0.22 (0.04)	0.49 (0.07)

The A/C and A/D intensity ratios decrease overall from ^{233}U, ^{235}U, the mixture, to ^{239}Pu. The fission product yield decrease in group A matches the trend found in Figure 1, which favours the production of fission products from ^{233}U and ^{235}U. The fission product yield for group C remains relatively constant for ^{233}U, ^{235}U, and ^{239}Pu. Therefore, the observed decrease in intensity ratio largely reflects the fission product yield s in group A. Group D displays a decrease in fission product yield however it is not as pronounced as the decrease in yield of group A. The C/D intensity ratios decrease in order from ^{239}Pu, ^{235}U, to ^{233}U. The decreasing ratios reflect the fission product yield variations of SNMs in groups C and D. The gamma line pair intensity ratios can be used to distinguish SNMs.

The same analysis was performed using four different ^{235}U enrichments: 0.72% natural uranium, 3.3% low enriched uranium (LEU), 10% enriched uranium, and 93.3% highly enriched uranium (HEU). The gamma-ray energy spectra for the enrichments can be found in Figure 8. Each gamma line pair has intensity ratios within uncertainties for all enrichments, which confirm that ^{238}U fission is negligible in the SLOWPOKE-2 site used for the Collaborative Materials Exercises. The simulations also tracked changes in MCNP net multiplication with increasing fissile mass. The net multiplication value is simply unity plus the gain in neutrons from fission and nonfission events (*eg*. n,2n). These simulations confirmed that net multiplicity remained negligible up until 20 *mg* (SLOWPOKE-2 licensing limits for fissile content irradiated a SLOWPOKE-2 site). Therefore any sample self-multiplication effects will be negligible in the Collaborative Materials Exercise at RMCC.

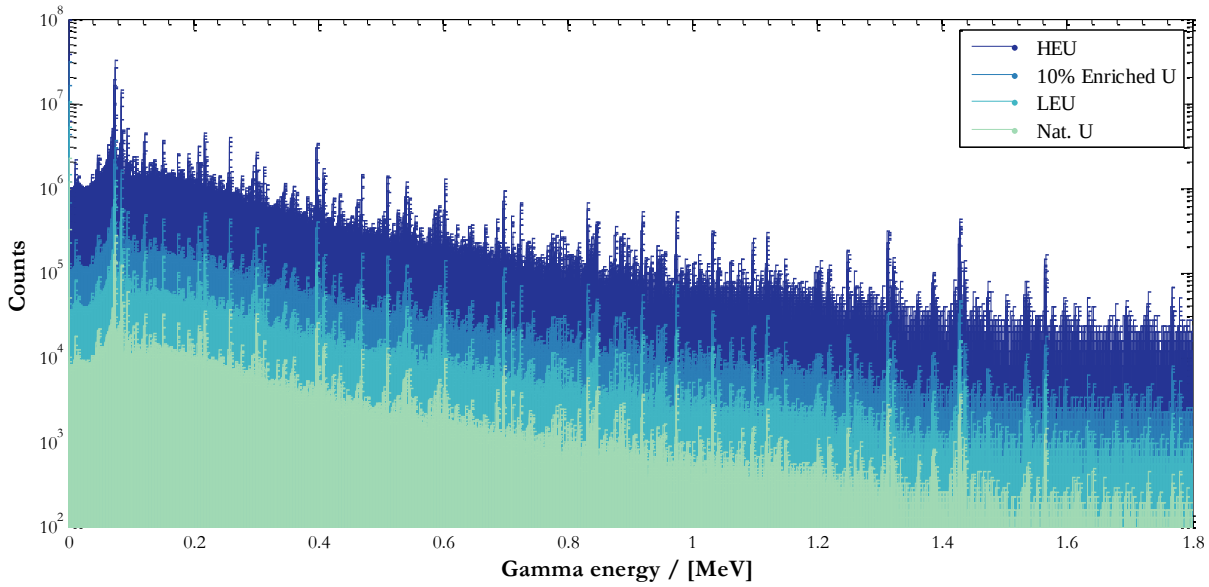

Figure 8: MCNP6 simulation of gamma-ray spectra for varying enrichments of uranium

A final set of MCNP6 comparisons included the simulation of gamma emissions from 93 % HEU and 3 % LEU samples in the DNGC system. The total spectra were normalized by total ^{235}U content, which resulted in fission product peaks of the same magnitude, Figure 9 (peaks around 78 and 79 keV). This simulation confirmed the prominence of the characteristic 75 keV gamma emission corresponding to ^{238}U activation and the decay of ^{239}U. Ratios of this 75 keV peak and prominent fission product peaks identified in Table 1 will therefore be examined in the Collaborative Materials Exercise to determine sample enrichement levels.

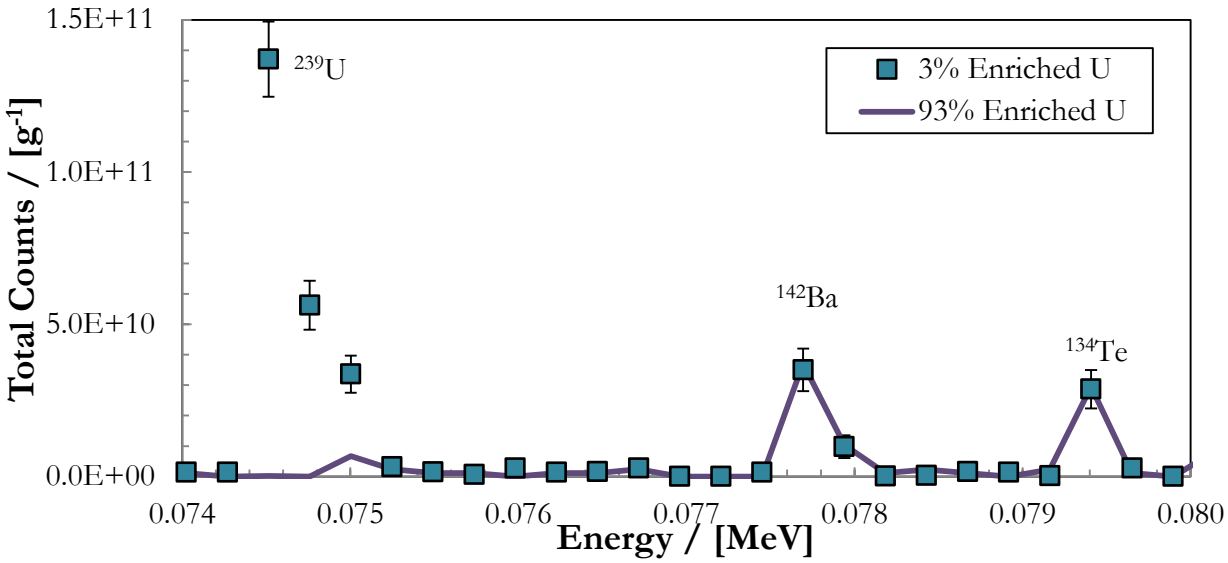

Figure 9: MCNP6 simulations of the characteristic 75 keV gamma emission from ^{239}U.

4. Summary and Conclusion

RMCC's SLOWPOKE-2 research reactor will be used in conjunction with the DNGC to investigate SNM samples to contribute to the International Technical Working Group's Collaborative Materials Exercise. A modified MCNP6 executable that fixes time binning in physics model-based delayed neutron simulations and a gamma-line emission data file containing updated data were found to produce more accurate delayed particle simulations than the unmodified version of MCNP6. This modified version of MCNP6 was used to model potential sample compositions and examine the possibility of using gamma line pair intensity ratios for nuclear forensics assay. The ratio between gammas emitted by fission products in groups A and C can be used to distinguish the presence of ^{233}U, ^{235}U, and ^{239}Pu. The 75 keV gamma peak from ^{238}U activation will be referenced to fission product peaks magnitudes to discern sample enrichment levels. The work demonstrates the contribution to experimental analysis that can be made by simulation models. Future work can include the analysis of more complex samples and other common nuclides found in fuels.

5. References

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