Fission Product γ-ray Measurements of ^{235}U and MCNP6 Predictions

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INTRODUCTION

The delayed neutron (DN) counting system at the Royal Military College of Canada (RMCC) has been recently upgraded to include the capability to additionally measure delayed gammas (DGs) emitted from special nuclear materials. This summary describes an initial comparison of DG measurements from irradiated natural uranium to MCNP6 [1] predictions.

DESCRIPTION OF ACTUAL WORK

A Description of the Experiment

The Delayed Neutron and Gamma Counting (DNGC) system utilizes the flux of a SLOWPOKE-2 reactor to induce fission in samples containing uranium and plutonium. Samples in the present experiments contained 0.3 *mg* of natural uranium and were prepared as acidified aqueous solutions from certified reference standards (CRM 4321C, NIST, Gaithersburg, MD) before being sealed in polyethylene vials (LA Packaging, Yorba Linda, CA). These samples were irradiated and then pneumatically transferred to the counting arrangement that simultaneously recorded the DGs and DNs emitted via fission product decay, as is depicted in Figure 1.

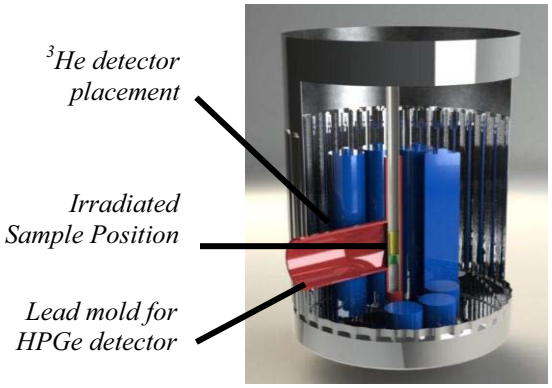

Fig. 1. A Schematic of the DNGC System

The counting system consists six ^{3}He detectors (RSP4-1613-202, GE Energy, Twinsburg, OH) imbedded in neutron moderating paraffin. Gamma rays are recorded by a High Purity Germanium (HPGe) detector (GMX-18190, SH-GMX CFG: S/N 26-N1476A). The detector is able to slide in and out of the lead mold (Figure 1) to modify geometric efficiency and detector dead-time. Lead shielding is extensively used in the system; to reduce environment background at the HPGe detector's crystal and to reduce γ background in the ^{3}He detectors. The fissile content was isolated from the detectors by the sample vial and polyethylene tubing.

A custom LabVIEW™ program has been written, which records the energy spectra of the delayed gammas and neutrons for predefined time intervals. For example, the measurements discussed here were performed as follows: 60 *s* irradiation of the sample, followed by 35 *s* decay before 565 *s* of counting. The cumulative counts for the entire energy spectra were recorded every ~2 *s* for both neutrons and gammas. Neutron measurements had been previously simulated in MCNP6 [2], their comparison is outside the scope of this summary.

The MCNP6 Model

MCNP6beta3 is the newest public release of the Monte Carlo code developed by Los Alamos National Laboratory. A description of the delayed particle features of MCNP6 can be found in Ref. 3. The DG=lines activation control option was used to sample DGs. Modeling the entire process (from irradiation, DG production, to the detection of photons) was computationally expensive. Thus two MCNP6 input decks were created to decrease relative errors in the output and speed up computations.

The first input deck modeled the irradiation of the uranium content and the resulting DG emissions. The fixed source option produced thermal neutrons, which irradiated a small amount of aqueous uranium content for 60 *s*. As this work is preliminary the magnitude of the flux within the SLOWPOKE-2 was not reproduced. Another modeled deviation from physical experiments was a ~1500x increase in fissile in order to improve the relative error of the DG emissions. Surface current (F1) tallies recorded the relative energy distributions of gammas emitted from the solution with chosen energy and time bins corresponding to the experiments conducted at the RMCC. The output corresponding to 565 *s* of counting after a 60 *s* irradiation and 35 *s* decay was used to define the source in the second MCNP input deck.

The second simulation deck reproduced the counting geometry and conditions. The geometry of the

DNGC system was modeled in MCNP using the dimensions measured during the system upgrade in 2012. The output from the previous simulation was used for source definition and placed inside the polyethylene vial. A Pulse Height (F8) tally was placed inside the germanium crystal, which recorded photon energy depositions in energy bins once again corresponding to measured bins. The measured energy resolution of the detector was reproduced with a Gaussian Energy Broadening (GEB) modifier, which defined the full width at half max (FWHM) values as a function of energy. The output of the F8 tally was then compared to measurements. At this time, the focus was on overall trends and the comparison of the individual DG peak intensities from measurements and simulations, no attempt to reproduce magnitudes has been made.

RESULTS

Both the measurement and simulations were read by a Matlab™ script, which corrected for dead time effects in the former. This script also contained an algorithm provided by Matlab Central that was modified to identify peaks in a user-defined energy range. A Savitzky-Golay [4] filter was applied to smooth the measured data in order to increase the effectiveness of peak identification. Uncertainties in the measurements were propagated by assuming an uncertainty associated (σ_c) with each count, C, as:

$$\sigma_C = \sqrt{C}$$

This error propagation did not account for uncertainties introduced via dead time corrections, spectra filtering, decay and count timings and sample concentrations, and is therefore an underestimation of the true uncertainty.

Once measurements had been corrected for dead time and experimental peaks were found, the analysis program identified peaks predicted by MCNP6. These MCNP peaks were then compared to measurements and if peak energies were within 0.8 *keV* of one another that pair was selected for comparison. The comparison was completed by calculating the area under each peak and subtracting background contributions to the spectra. For this preliminary comparison, the 25 most prominent measured peaks were selected for comparison to MCNP6 simulations. As there was no attempt to reproduce the absolute magnitude of gammas recorded, measurements and simulation peaks were all normalized to a total net area (of all 25 peaks) of 1, which allowed for the comparison of relative peak intensities.

Beddingfield and Cecil [5] identified > 800 *keV* fission product gammas as important energies when examining unidentified fissile material and their measurements have been previously compared to MCNP simulations [6]. These energies were therefore originally selected for comparison in this work. The signal to noise ratio in measurements for many peaks > 1.1 *MeV* was less than 2:1 so they were omitted from this analysis. The lower range of analysis was extended down to 685 *keV* to include multiple $^{132/132m}$Sb decay energies for reasons that will be discussed later in this section.

Figure 2 shows measurements, MCNP6, and relative intensity comparisons of fission product gamma-ray spectra produced from 685 – 1100 *keV*. Analysis of measurements identified 56 peaks that are marked in Figure 2, of which 49 were predicted by MCNP6 simulations. The 25 most intense measured peaks were all predicted by MCNP6 and are also shown on both the measured and simulated spectra. The relative intensities of these peaks are displayed at the bottom of Figure 2.

Overall, the relative intensities of the peaks were comparable in simulations and experiments with several notable exceptions. When the original range of analysis from 800 – 1100 *keV* was used a $^{132/132m}$Sb peak energy emerged as a prominent outlier. The range of analysis was therefore extended down to 685 *keV* to include another prominent $^{132/132m}$Sb decay at 697 *keV*. MCNP6 over predicts the intensities of both observed $^{132/132m}$Sb peaks by a factor of 1.5 and 1.4 at the energies of 697 and 974 *keV*, respectively. The observed over-prediction of $^{132/132m}$Sb by MCNP appears to concur with comparisons of highly enriched uranium metal measurements and MCNPX performed by Durkee *et al* [6].

Energy intensities corresponding to the decay of $^{90/90m}$Rb were significantly higher in measurements than simulations. The possibility of non-^{235}U fission contributions to measured $^{90/90m}$Rb counts was ruled out via the comparison of several experimental gamma counts, Figure 3. In each case the vial was irradiated for 60 *s*, allowed to decay for 10 *s* and counted for 170 *s*. In these comparisons the relative contributions of the irradiated empty vials, HNO_3 solutions and ^{238}U content were ascertained and none were found to produce a noticeable 832 or 1061 *keV* peak.

CONCLUSIONS

Preliminary comparisons of the measured gamma ray spectra of ^{235}U fission products at RMCC and MCNP6 predictions have been completed. In the range of 0.685 – 1.1 *MeV* MCNP6 predicted the presence of the majority of prominent measured peaks. The relative intensities of the 25 most prominent peaks were in general agreement with several notable discrepancies, notably $^{90/90m}$Rb and $^{132/132m}$Sb.

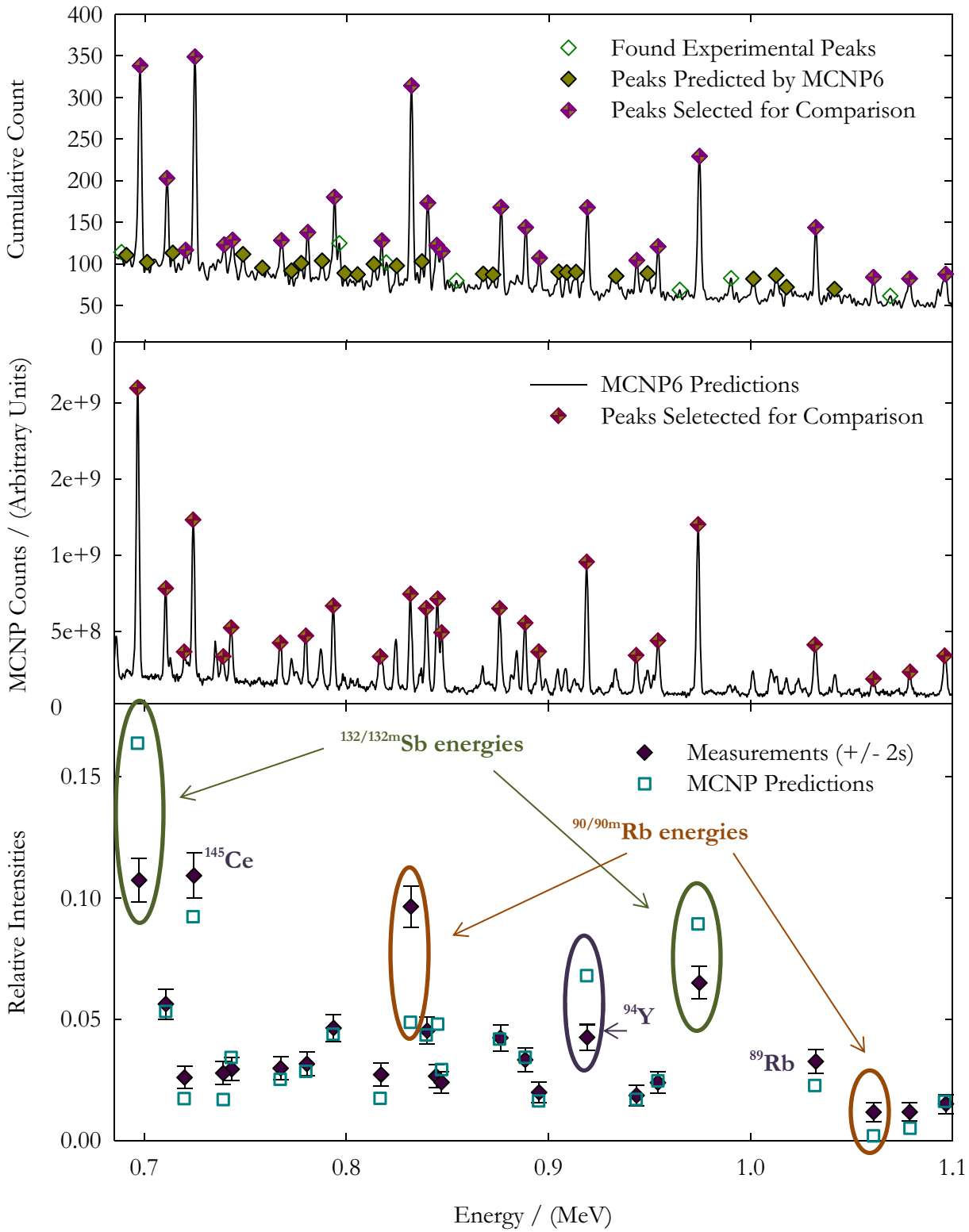

Figure 2: Measured and MCNP6 Simulated Delayed Gamma Count Comparisons

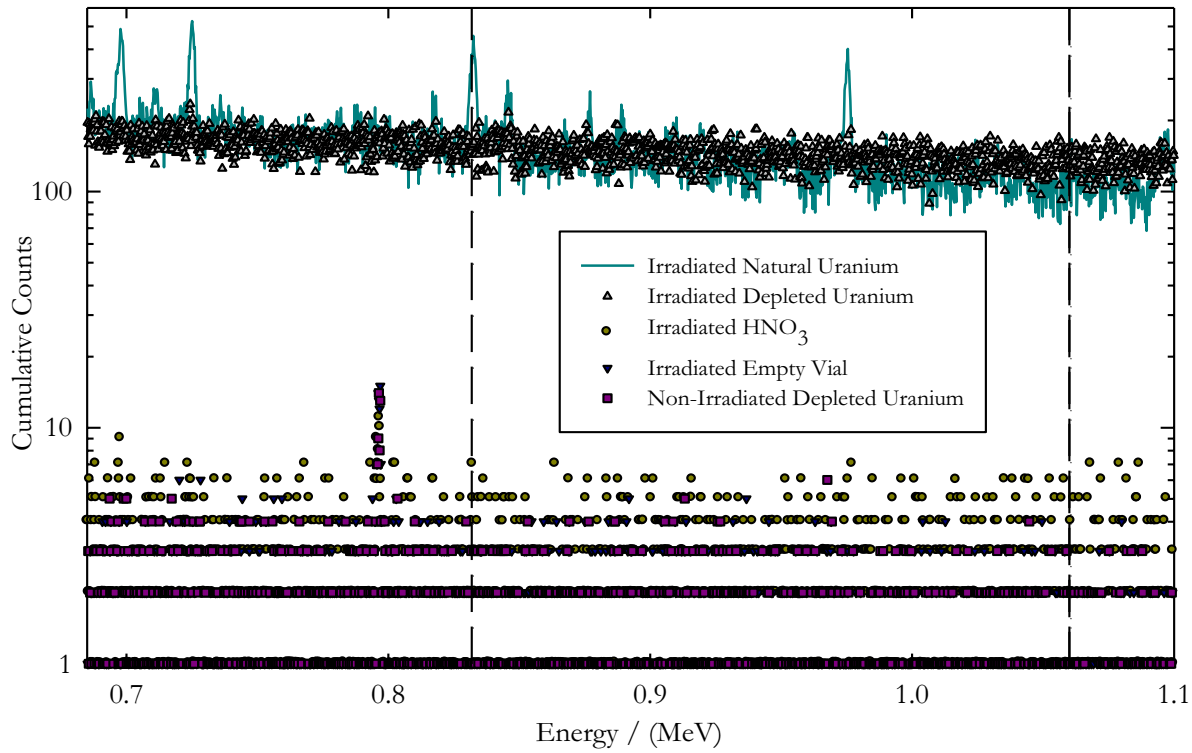

Figure 3: Relative count contributions from depleted & natural uranium, empty vials and the HNO_3 solution.

FUTURE WORK

Future work will include ^{233}U and ^{239}Pu measurements, varied irradiation, decay and count times and their simulation in MCNP6. A comparison of peak growth with respect to count time will also be included in future efforts.

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