High Energy Delayed Gamma Spectroscopy for Plutonium Assay of Spent Fuel

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Abstract

The direct measurement of plutonium in spent reactor fuel is an unmet challenge in international safeguards. In this simulation study, we investigate the use of the delayed gamma rays from fission product nuclei to determine the amount of fissile isotopes (Pu-239, Pu-241, and U-235) in irradiated light water reactor fuel assemblies. Fission is stimulated with an interrogating neutron source, and the radiation from the short lived fission products is measured. This measured gamma spectrum is then fit to a linear combination of spectra from pure Pu-239, Pu-241, and U-235 to determine the proportion of fissile isotopes present. In this paper, we describe the modelling and analysis methods used to represent the background of radioemissions from long-lived isotopes originally present in the spent fuel and the short time scale delayed gamma signal. Results are presented for simulations using a nominal instrument design on a library of fuel assemblies with burnups ranging from 0 to 60 GWd/MTU.

1. Introduction

In order to safeguard nuclear fuel cycles, it is necessary to verify the quantity of fissile material in spent nuclear fuel. Current methods used for this task are confirmatory – given an operator-supplied history of irradiation, a prediction is made of the isotopic inventory. If passive measurements of certain high signature non-fissile isotopes (such as Cs-137 or Cm-244) are in agreement with the predictions, the fuel is assumed to contain the predicted quantity of fissile isotopes. This method is subject to high uncertainties [1], in addition to relying on operator supplied information. Ideally, an independent, direct, high accuracy measurement could be obtained to quantify the fissile mass present.

The intense radiation from spent fuel produces a high background which obscures the passive signal of U-235 and Pu-239. Consequently, we turn to active techniques. One potential method is to exploit the unique distribution of fission products produced by each isotope. Many of these fission products are unstable, and decay on a time scale of seconds to minutes. Their decay often involves the emission of highly energetic gamma rays. By detecting the intensity of the gamma emission lines, the amount of signature radioisotopes can be determined, and from this amount the original fissile composition of the fuel can be found.

This idea is not new. Marrs et al. have identified line intensity ratios that can differentiate between the fission of U-235, U-238, and Pu-239 as well as thermal and fast neutron fission when measured over a time scale of several
minutes to several hours after irradiation [2]. Beddingfield and Cecil identified several fission product gamma-ray lines near 1 MeV in energy whose ratios can be used to differentiate between U-235 and Pu-239[3]. Cole et al. [4] also explored the fission-product spectroscopy concept. Firestone et al. [5] determined the relative abundances of U-235, U-238, and Pu-239 in a mixed sample from ratios of counts due to fission products Rb-90, Rb-90m, and Y-95. However, these studies only investigated un-irradiated nuclear material. The ability to distinguish fissile isotopes in the presence of a high background was not assessed.

In this work, simulations are used to assess the viability of high energy delayed gamma spectroscopy (HEDGS) on spent fuel, under the premise that the huge parameter space of this assay technique can be more efficiently explored using modelling, and those modelling results can then help guide the planning and execution of time-consuming and expensive benchmarking measurements in the future. The work described in this paper builds on the previous study of fission-product spectroscopy for fissile isotope quantification and extends the concept to irradiated fuel assemblies [6].

2. Methods

2.1. Nuclear Data Sources

The Evaluated Nuclear Structure Data File (ENSDF) library [7] was used for nuclear decay data, including the gamma ray transition lines and intensities, lifetimes, and the daughter products. Where ENSDF data is not available, the on-line Chart of Nuclides at the National Nuclear Data Center (NNDC) [8] was used for decay rates and branching ratios of decay modes. Independent fission product yields were obtained from the Evaluated Nuclear Data File (ENDF) libraries [9].

2.2. Simulation Geometry

The fuel for our simulations is taken to be Westinghouse 17×17 PWR fuel assemblies [10]. A library of fuel assembly isotopic inventories was made using the ORIGEN-2 code [11] for burnups ranging from 0 to 60 GWd/MTU and a 10 year cooling time. The fuel composition and burnup was assumed uniform throughout each assembly.

The instrumentation geometry used is shown in figure 1. A cylindrical sleeve of high-density polyethylene (HDPE) moderator (45 cm diameter, 20 cm thick) is covered by a 17-cm thick lead shell for neutron reflection and gamma-ray shielding. A square hole is cut along the axis to admit the fuel assembly. A D-T neutron source (red circle) emitting at $10^{10}$ neutrons per second is placed in a cylindrical iron plug (19 cm diameter, 20 cm length) to initially slow the neutrons through inelastic collisions. A 14-cm diameter window is cut in the lead reflector, leaving 11 cm of lead in the window as a low energy gamma filter. A coaxial HPGe detector (7.95 cm in diameter, 8.05 cm long, approximately 114% relative efficiency, 3.2 keV resolution at 3 to 4 MeV energy) views a section of the assembly through this window.
2.3. Interrogation Protocol

We assume the following interrogation and measurement protocol: the neutron generator is turned on for ten seconds, during which time the HPGe detector is not recording. Then, the neutron generator is turned off for ten seconds while the HPGe detector records the delayed gamma signal without the interference of neutrons or prompt fission gamma rays. This is repeated 30 times, for a total assay time of 10 minutes.

2.4. Isotope Signature Templates

The detector response to each significant radioisotope – both from the long-lived isotopes originally present and short lived induced signal – is simulated independently. This allows us to do the lengthy computations only once, and rapidly sum from a library of responses when the source terms change. A wide range of burnups, interrogating neutron source intensities and dwell times can then be quickly analyzed.

The GEANT4 radiation transport toolkit [12] is used to propagate gamma rays from the spent fuel assembly through the interrogation chamber and into the detector. In each simulated decay, the decay mode (α, β-, β+, electron capture) is sampled from the relative intensities of available decay modes to determine the daughter nucleus. A nuclear transition is sampled from the distribution of transitions subsequent to radioactive decay and, if internal conversion does not occur, a gamma ray is emitted in the GEANT simulation. The excitation level of the daughter nucleus is decreased according to the transition, and a new transition is chosen from this level. In this manner, the decay cascade is followed until the daughter nucleus arrives at a metastable or ground state.

The method chosen to propagate the radiation from the source to the detector varied depending on whether we were investigating the passive signal, where the primary interest is the total count rate of the detector, or the active signal, where we areprimarily looking at the penetration of gamma rays between 3 MeV and 4 MeV. These methods are described in the next two sections. In all cases, full transport calculations including all relevant physics were performed once any radiation enters the HPGe detector.

Once the signature templates for each isotope have been determined, they can be sampled to emulate mixtures of radionuclides like those in the spent fuel. Given an expected number of decays (for example, a known activity times the measurement time), the total detector response is found by scaling the response of each isotope in the library by the ratio of the expected decays to the total number of decays simulated for that isotope, and then summing over all isotopes.
2.4.1. Passive background

The gamma-ray emissions predicted by ENDF for an isotope are uniformly distributed over the fuel pins within the interrogation chamber. Geometry biasing was employed by emitting gamma rays into a 24 degree cone centred on the detector. This approach was adopted after scoping calculations showed that emission into this cone was essentially indistinguishable from isotropic emission in terms of the number of peak counts recorded in the detector, for the emission energies of interest. Gamma-ray transport through the fuel assembly and surrounding HDPE moderator is ray traced — attenuation is handled analytically while scattered and secondary particles are neglected. Once the photon enters the lead shell, however, full transport is enabled to accurately capture buildup effects in the lead. The variance reduction techniques of geometry splitting and Russian roulette are used to reduce the computational load [13]. Importance cells are chosen as concentric spherical shells around the detector, with a factor of 2 increase in importance as the detector is approached. Scoping calculations of this transport approach are indistinguishable from full transport simulations for small scale runs (see next section and Figure 3 for more discussion of variance reduction methods).

Calculated total count rates produced by some of the long-lived fission products are given in Table 1. An example of the background spectrum calculated for HEDGS assay of a PWR fuel assembly is given in Figure 2. We choose to focus on the region from 3200 keV to 3700 keV (Figure 2) for several reasons. First, the penetration of the actively induced signal gamma rays (see next section) is relatively high in this region because the Pb cross-section is near a minimum. Second, the Pb attenuation is relatively uniform over this energy window (for 11 cm of Pb, attenuation ranges from a factor of 0.00522 for the Te-136 line at 3235 keV to 0.00533 for the 3616 keV Sr-95 line), which minimizes the need for self-attenuation corrections in the analysis of the signal emanating from different regions in the assembly. Perhaps most importantly, however, the passive background in this energy window is quite low. One of the major sources of background is spontaneous fission prompt gamma rays, which produces a smoothly varying background continuum. In previous HEDGS work [6], the spontaneous fission gamma rays were found to be several orders of magnitude below the in-detector Compton continuum associated with the HEDGS signal induced by the neutron interrogation (see next section). Another source of background in the high-energy region is neutron-capture gamma rays. This source term has not been considered in this work to date but is under investigation along with other potential sources of background in the high-energy region.

**Table 1.** Recorded count rates from significant long-lived passive background isotopes in PWR spent fuel, assuming the Figure 1 geometry.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>60 GWD/MTU</th>
<th>45 GWD/MTU</th>
<th>33 GWD/MTU</th>
<th>18 GWD/MTU</th>
<th>5 GWD/MTU</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs-137</td>
<td>26</td>
<td>19</td>
<td>14</td>
<td>8</td>
<td>2</td>
</tr>
<tr>
<td>Cs-134</td>
<td>13,097</td>
<td>7,977</td>
<td>4,785</td>
<td>1,459</td>
<td>129</td>
</tr>
<tr>
<td>Eu-154</td>
<td>86,180</td>
<td>57,598</td>
<td>34,002</td>
<td>9,890</td>
<td>657</td>
</tr>
<tr>
<td>Y-90</td>
<td>2,507</td>
<td>2,062</td>
<td>1,666</td>
<td>1,017</td>
<td>324</td>
</tr>
<tr>
<td>Rh-106</td>
<td>1,900</td>
<td>1,227</td>
<td>898</td>
<td>365</td>
<td>70</td>
</tr>
<tr>
<td>Pr-144</td>
<td>915</td>
<td>705</td>
<td>691</td>
<td>398</td>
<td>159</td>
</tr>
<tr>
<td>Tl-208</td>
<td>31</td>
<td>19</td>
<td>10</td>
<td>3</td>
<td>0</td>
</tr>
<tr>
<td>Total</td>
<td>104,655</td>
<td>69,607</td>
<td>42,064</td>
<td>13,131</td>
<td>1,341</td>
</tr>
</tbody>
</table>
In this work, we assume an HPGe detector that can operate at count rates up to 100 kHz without significant degradation in the energy resolution needed to resolve key peak regions. Since our signal is chosen over an energy range near the maximum mean free path for photons in lead, it is always desirable to reduce count rate by increasing the filter thickness rather than cutting down the field of view with collimators or increased distance, since the former method preferentially decreases the passive background compared to the signal. The thickness of the lead filter window in Figure 1 was chosen accordingly.

2.4.2. Active Signal

In order to simulate the intensity of the gamma ray signal, the fission rate must be known. To estimate this, GEANT4 is used to perform a neutron transport simulation of the 14.1 MeV neutrons from the D-T neutron source to the fuel in the notional HEDGS geometry of Figure 1. The location of every fission is recorded and written to a file. Ideally, the fission rates of each major contributing isotope (e.g. U-235, Pu-239 and Pu-241) would be recorded for each fuel pin in a specific assembly. However, because the high precision neutron data for GEANT4 includes no elements with an atomic number greater than uranium, some simplifying approximations were necessary to calculate the induced fission rate in this study. The fuel in the neutron transport simulations was modelled as pure uranium oxide at 5% enrichment. Out of $10^6$ neutrons emitted in the simulation, we obtained a total of $3.07 \times 10^5$ fission events, giving a mean of 0.307 fissions per neutron. The fission rate calculated with GEANT for this uranium-only fuel is predominantly induced by thermal neutrons (73%, up to 1 eV) but there are substantial epithermal (7%, 1 eV to 1 keV) and fast (20%, 1 keV up to 14.1 MeV) components as well. From this point forward in this study, it is assumed that all fissions are induced by thermal neutrons for two reasons. First, this approach allows a relatively simple scaling of the uranium-only fuel fission rate (i.e. thermal fission in U-235) to estimate the induced fission rates for the fuel that contains U and Pu isotopes. That is, the (known) relative mass concentration of each isotope is scaled by the thermal neutron fission cross section for that isotope to calculate the fraction of fissions due to U-235, Pu-239 and Pu-241, for each fuel region. The second reason for assuming only thermal fission is that fission-product yield data is readily available at thermal energies, but decidedly less so at other energies. The assumptions of uranium-only fuel and thermal-only fission are expected to be a significant source of systematic error in HEDGS assay, but are not addressed in this paper. Improving the fidelity of the induced fission modelling is a topic of future research and is discussed in the Conclusions section. It should be noted that the use of uranium-only fuel only affects the neutron transport...
simulations, which give the efficiency of fission for a given neutron emission rate and the distribution of fissions in space. For fuels other than fresh 5% enriched uranium, it is assumed that the fission efficiency scales with the nuclear fission cross section and concentration, and that the spatial distribution does not change.

The GEANT calculations of fission distribution for each fuel pin also supported the development of an approach to incorporate the axial variation of the induced fission rate in HEDGS assay. The fission distribution data of the middle 40 cm of each fuel pin was fit to a sum of non-negative “basis shapes” to obtain an approximate axial variation of fission density using the downhill simplex method [14]. As with the isotopic signature templates described earlier, the goal of the basis-shape methodology was flexibility and extensibility so that other HEDGS instrument designs can be easily incorporated and evaluated in the future. Examples of the basis shapes and how their combination is used to approximate the axial variation of the induced fission signal is described in more detail in [15]. To simulate the fission of assemblies with finite burnup and differing initial composition, we assume that the pin-to-pin and axial fission profile does not change from what was calculated using the uranium-only fuel described previously.

Once the distribution of induced fission events is determined for each PWR assembly using the methods described above, the resulting population of short-lived fission products during the assay period can be determined. Given the decay rate and decay-mode branching ratios of each isotope and isomer in our system and the initial population of the isotopes, a system of differential rate equations can be used to solve for the population of isotopes at any given time. We set up a Kaps-Rentrop method stiff differential equation solver [14] that determines the populations of the isotopes with time while keeping track of the total number of expected decays of each isotope. The population builds up with time when the neutron source is on; each fission adds the ENDF-tabulated independent yield to the isotopic population. While the source is off, the decays of the fission products are integrated to find the total number of decays when the detector is recording. The output is normalized to one fission per second, and then scaled by the neutron emission rate from the source ($10^{10}$ s$^{-1}$) multiplied by the fissions per neutron from the neutron transport simulation to return the total number of measurable decays for each isotope in our measurement protocol. Each basis shape for each pin is then apportioned decays based on its fraction of fissions in the neutron transport simulation. The contribution to the total signal is then found by summing the signal response of the detector to each isotope from each basis shape from each fuel pin, scaled by the number of decays for that isotope, basis shape, and pin.

As with the simulation of the passive background source terms, variance reduction techniques are also used in the calculation of the induced signal. Gamma photons are emitted only into the solid angle subtended by the detector and its shielding (Figure 3), and are ray traced from the point of emission until they enter the detector. Ray tracing takes full account of their attenuation, reducing the importance weighting of the tracked particle in accordance with the usual exponential fall-off of intensity, without performing time-consuming Monte Carlo steps. The effect is as if every interaction – photoelectric, Compton, or pair production – removes the photon from the beam without secondary radiation produced. Once the gamma photon enters the detector full transport is enabled, so that Compton down scatter and pair production effects within the detector and its shielding is taken into account.
3. Results

The left pane of Figure 4 shows the delayed gamma spectra for the fissile isotopes U-235, Pu-239, and Pu-241. The differences between the net count rates in key peak regions indicate that it should be possible to distinguish between the fissile isotopes on the basis of the delayed gamma spectrum. For example, a weak Te-136 intensity relative to Y-97 indicates a high Pu-239 content, while the presence of the Tc-106 line is indicative of the overall plutonium abundance.

The estimated statistical precision for fissile isotope assay can be calculated by assuming Poisson statistics for mean values of the bins in the spectra shown in Figure 4 (left pane) scaled by the relative proportion of the parent fissile isotope for the burnup under consideration. The simulated spectra for the unknown fuel assembly was then fit, in the least-squares sense, with the three fissile isotope “basis spectra” depicted in Figure 4 (left pane). By assuming a second detector viewing the assembly opposite the modelled detector, we can double the counts and reduce the statistical uncertainty in a given interrogation time. The results of such an analysis are given in Table 3, and shown graphically in the right pane of Figure 4.

Figure 4. Left: Example pulse-height spectra of active-interrogation signal for pure U-235, Pu-239, and Pu-241 for a HEDGS measurement protocol of 10-seconds on, 10-seconds off and 10 minutes of total interrogation time. Right: Simulated HEDGS assay results for a range of PWR fuel burnup. The true fissile isotope concentrations (solid lines) are compared to calculated values and associated statistical uncertainties (one-sigma error bars).
Table 2. Simulated assay of fuel elements of various burnups and comparison to true mass fractions. The statistical uncertainty (one sigma) of the calculated values is given.

<table>
<thead>
<tr>
<th>Burnup (GWd/MTU)</th>
<th>U-235 Calculated</th>
<th>U-235 True</th>
<th>Pu-239 Calculated</th>
<th>Pu-239 True</th>
<th>Pu-241 Calculated</th>
<th>Pu-241 True</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.946 ± 0.023</td>
<td>0.908 ± 0.059</td>
<td>0.084 ± 0.029</td>
<td>0.001</td>
<td>-0.039 ± 0.027</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>0.913 ± 0.024</td>
<td>0.915 ± 0.063</td>
<td>0.084 ± 0.029</td>
<td>0.001</td>
<td>-0.039 ± 0.027</td>
<td>0</td>
</tr>
<tr>
<td>18</td>
<td>0.806 ± 0.026</td>
<td>0.750 ± 0.066</td>
<td>0.230 ± 0.055</td>
<td>0.020</td>
<td>-0.036 ± 0.028</td>
<td>0.061</td>
</tr>
<tr>
<td>33</td>
<td>0.521 ± 0.026</td>
<td>0.546 ± 0.063</td>
<td>0.393 ± 0.039</td>
<td>0.104</td>
<td>-0.036 ± 0.028</td>
<td>0.061</td>
</tr>
<tr>
<td>45</td>
<td>0.389 ± 0.028</td>
<td>0.372 ± 0.068</td>
<td>0.525 ± 0.166</td>
<td>0.104</td>
<td>-0.036 ± 0.028</td>
<td>0.061</td>
</tr>
<tr>
<td>60</td>
<td>0.217 ± 0.027</td>
<td>0.191 ± 0.065</td>
<td>0.662 ± 0.142</td>
<td>0.147</td>
<td>-0.036 ± 0.028</td>
<td>0.061</td>
</tr>
</tbody>
</table>

4. Benchmarking Measurements

Benchmarking measurements were performed using the prompt gamma neutron activation analysis (PGNAA) facility at Oregon State University (OSU) with the goal of preliminary validation of the HEDGS modelling framework. The OSU PGNAA facility consists of an HPGe spectrometer (36.5% relative efficiency) with a 28.9-mm pinhole collimator 35.6-cm long. The HPGe spectrometer views a sample in the path of a collimated neutron beam from the OSU TRIGA® reactor. The collimator for the beam contains both a sapphire filter to remove fast neutrons and a bismuth filter to remove gamma rays emerging from the reactor core. The facility has a thermal neutron flux of $3 \times 10^7$ n/cm$^2$-s and a beam width of 2 cm. The sample used for the validation measurements was a pair of highly enriched uranium foils (93 wt% U-235) with a total mass of 340 mg.

Figure 5 shows the result of a measurement on these uranium foils using a protocol of 10 seconds of irradiation followed by 10 seconds of counting, repeated for 100 cycles. The simulated spectrum is scaled and offset vertically in Figure 5 to remove differences in absolute magnitude and allow a comparison of the key figure of merit in HEDGS: relative net peak count rates. A comparison of the simulated and measured net peak count rates is shown in Table 3. The agreement between simulated and measured relative intensity is generally good except for Te-136, which is significantly over-predicted by theory. Measurements of this type, using both U and Pu foils, will continue in collaboration with Oregon State University. These measurements will support efforts to identify deficiencies in the tabulated nuclear data and modelling methods needed to faithfully simulate HEDGS assay of spent fuel.
Figure 5. Measured spectrum of 93% enriched uranium, alternating 10 seconds of irradiation with 10 seconds of measurement for 100 cycles, compared to theoretical predictions. The simulation curve is scaled and offset vertically.

Table 3. Fitted strengths of gamma lines in the measured spectrum. The intensity relative to the 3600 keV Rb-91 peak are shown for both experiment and simulation.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (KeV)</th>
<th>Counts</th>
<th>Relative Intensity (measured)</th>
<th>Relative Intensity (simulated)</th>
<th>% difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Te-136</td>
<td>3235</td>
<td>40 ± 13</td>
<td>0.20 ± 0.10</td>
<td>0.41 ± 0.13</td>
<td>+105%</td>
</tr>
<tr>
<td>Tc-106</td>
<td>3260</td>
<td>10 ± 9</td>
<td>0.05 ± 0.09</td>
<td>0.12 ± 0.17</td>
<td>-</td>
</tr>
<tr>
<td>Y-97</td>
<td>3288</td>
<td>157 ± 16</td>
<td>0.77 ± 0.11</td>
<td>0.86 ± 0.14</td>
<td>+12%</td>
</tr>
<tr>
<td>Rb-90m</td>
<td>3317</td>
<td>82 ± 14</td>
<td>0.40 ± 0.11</td>
<td>0.38 ± 0.13</td>
<td>-5%</td>
</tr>
<tr>
<td>Rb-90</td>
<td>3383</td>
<td>82 ± 11</td>
<td>0.40 ± 0.10</td>
<td>0.57 ± 0.13</td>
<td>+42%</td>
</tr>
<tr>
<td>Y-97</td>
<td>3401</td>
<td>131 ± 15</td>
<td>0.65 ± 0.11</td>
<td>0.70 ± 0.14</td>
<td>+8%</td>
</tr>
<tr>
<td>Rb-93</td>
<td>3458</td>
<td>49 ± 8</td>
<td>0.24 ± 0.09</td>
<td>0.25 ± 0.12</td>
<td>+4%</td>
</tr>
<tr>
<td>Rb-90</td>
<td>3534</td>
<td>84 ± 13</td>
<td>0.42 ± 0.10</td>
<td>0.42 ± 0.13</td>
<td>0%</td>
</tr>
<tr>
<td>Y-95</td>
<td>3576</td>
<td>113 ± 13</td>
<td>0.56 ± 0.10</td>
<td>0.63 ± 0.14</td>
<td>+12%</td>
</tr>
<tr>
<td>Rb-91</td>
<td>3600</td>
<td>203 ± 16</td>
<td>1.00</td>
<td>1.00</td>
<td>-</td>
</tr>
</tbody>
</table>

5. Conclusions

A modelling framework has been developed to evaluate the viability of the High-Energy Delayed Gamma Spectroscopy (HEDGS) method for spent fuel assay. The modelling approach was developed with the goal of post-processing flexibility so that an isotope signature library, containing long-lived fission products that define the background and short-lived fission products that constitute the induced signal, can be utilized to emulate a wide range of instrument design choices and fuel compositions. To demonstrate the HEDGS modelling methods, the assay of PWR fuel assemblies using collimated HPGe spectrometers was simulated.

The preliminary viability results presented here indicate that statistical uncertainties of approximately 5% can be achieved for the relative abundance of U-235, Pu-239 and Pu-241, assuming two HPGe spectrometers, a commercially available D-T generator of approximately $10^{10}$ n/s, and a total assay time of less than 30 minutes. Lower statistical uncertainties could be achieved by increasing the assay time, D-T generator intensity or the number of HPGe spectrometers. The absence of obvious systematic bias in the calculated assay values for the simplified fuel assay scenario analyzed in this work, helps to verify that the modelling framework and analysis algorithms developed for HEDGS are functionally correct and are suitable for continuing study of HEDGS viability.

The estimates of statistical uncertainty presented here should not be taken as performance predictions for realistic HEDGS assay of actual spent fuel. This study did not address some of the effects expected to produce (potentially substantial) systematic errors in delayed-gamma assay. These effects include the relative rates of thermal, epithermal and fast fission in fissile and fertile isotopes, and the impact of neutron-absorbing fission products and actinides. Other effects that should be addressed include radial variation of burnup within fuel pins and burnup variation across the pins of an assembly. Future work will add more realism to the simulation framework so that the magnitude of these systematic uncertainties can be estimated and combined with the statistical uncertainties to predict total assay uncertainty. This additional inquiry, coupled to expanded empirical validation using pure and mixed samples, will support a more definitive assessment of fission-product spectroscopy for direct measurement of Pu in spent fuel.
6. Acknowledgements

This work was supported by the US National Nuclear Security Administration’s Next Generation Safeguards Initiative (NGSI). The authors would like to thank NA-24’s Steve Lamontagne and Kevin Veal for their programmatic and technical guidance on this project, and Steve Tobin and Jianwei Hu of Los Alamos National Laboratory for their assistance in the interpretation of the fuel assembly library that LANL developed under NGSI funding.

References