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SENSITIVITY OF SPENT NUCLEAR FUEL GAMMA-RAY MEASUREMENTS

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ABSTRACT

While gamma measurements are a well understood technique for estimating burn up and cooling time in spent nuclear fuel, only a few isotopes are currently used and calculations are dependent on operator declarations. Utilizing a larger set of nuclide measurements as well as including low energy measurements (around 100 keV) could provide a significant increase in the information gained from such measurements. Nuclides investigated in this work include Cs-134, Cs-137, Eu-154, Eu-155, Ce-144, Sb-125, Rh-106, Am-241, and x-ray peaks from uranium and plutonium. Low energy, broad range, and high energy gamma measurements have been performed on a variety of spent fuel from PWR and MOX reactors. Selected measured fuel locations have been simulated with Origen, TransLAT, SCALE, and Monteburns to benchmark each of these programs with destructive analysis results. The program which best predicted the measurements was then used to perform a sensitivity analysis of the effects of fuel parameters (burnup, cooling time, and initial enrichment) on each nuclide measurement.

INTRODUCTION

Current safeguards approaches for spent fuel (in solid form) rely upon item accounting in which bulk plutonium and/or ^{235}U are not directly measured or accounted for. This is due to the inherent difficulties in measuring these dilute products in the solid spent fuel. There has recently been increased interest in developing methods for performing bulk accountancy measurements on solid spent fuel and these methods would all benefit from additional information about the fuel [1]. The additional characteristics of a spent fuel assembly that would possibly have safeguards relevance would be:

1. Burnup (BU)
2. Cooling time (CT) or the time since final discharge from the reactor
3. Initial enrichment (IE)
4. Quantification of strong neutron absorbers (e.g., ^{155}Gd and ^{241}Am)
5. Assembly power history which can be characterized by the following:
    a. Total days of irradiation
    b. Total days of shutdown between initial charge to the reactor and final discharge after the last cycle
    c. Power level of assembly during each cycle

Passive gamma measurements have been widely used for determination of burnup of spent nuclear fuel, but this use depends on knowledge of cooling time and initial enrichment or calibrated equipment for a narrow range of spent nuclear fuel [2]. Most current spent nuclear fuel gamma-ray measurements rely on only three nuclides: ^{137}Cs, ^{134}Cs, and ^{154}Eu. The main goal of this research was to determine the additional information about the fuel that can be obtained by using more nuclides visible in both broad-energy and low-energy photon (gamma-ray and x-ray) measurements.

MEASUREMENTS

Passive gamma-ray measurements of spent fuel have been extensively studied and are routinely used, especially for measurements in the 400-1500 keV range. Measurements of photons (gamma-rays and x-rays) in the 60-150 keV range in spent fuel have been much less explored. There are a number of interesting photon lines in this region. This includes x-rays from uranium and plutonium, gamma-rays from the decay of ^{241}Am, and several fission product decay gammas. Measurements of the uranium x-rays have been published extensively. Original work on the measurement of the plutonium $K_{\alpha 1}$ x-ray at 103.7 keV showed the feasibility of measuring this line in fast reactor fuel [3-4] and some recent work has demonstrated the feasibility of measuring this line in light water reactor fuel [5-6]. One of the main reasons low energy photon measurements (~100 keV) are not typically used on spent nuclear fuel is the that these photons only have a mean free path of approximately ? cm in uranium dioxide. Thus, in assembly measurements, the only detectable, low-energy photons are from the outer edge of the outer pin closest to the detector. This research focuses on pin measurements to investigate the extent of information available from combined low and broad energy photon measurements. However, it is expected that this information will also be useful in some cases for assembly measurements.

Between 2008 and 2010, nondestructive analysis measurements were performed at Oak Ridge National Laboratory (ORNL) on single fuel pins in air from a wide range of nuclear reactors. The fuels were irradiated in the North Anna (NA), Three Mile Island (TMI), Calvert Cliffs (CC), Limerick (LM), Surrey (SR), and Cattawba (MOX) reactors. These measurements included fuel from PWRs and BWRs, uranium dioxide and mixed oxide fuel, a range of cooling times from 9 months to 30 years, a range of burnups from 18 GWd/MTU to 66 GWd/MTU, and a range of initial enrichments of 2.4 to 4.2 wt% ^{235}U. These measurements were performed with the pin in a hot cell at ORNL and using both low-energy HPGe and broad-energy HPGe detectors. Details of the measurement setup and results can be found in Ref. 5-6. A summary of the nuclides identified in four of these spectra sets is provided in Table I (additional detail can be found in Ref. 7).

For the purposes of this paper, we focused solely on the NDA measurements on the fuel pin from Calvert Cliffs (CC). In October 2009, a coaxial HPGe detector was used to obtain a broad-energy gamma spectrum at position 552 mm on the CC pin, and a planar HPGe detector was used to obtain low-energy gamma spectra at positions 552 mm and 538 mm on the same pin.

DA was performed by ORNL in Spring 2011 on two fuel segments from the CC pin taken from the 552 mm and 538 mm positions. This DA included high-resolution gamma spectroscopy on the dissolved samples as well as inductively coupled plasma mass spectrometry (ICP-MS). Isotopes measured via gamma spectroscopy of the DA samples were Am-241, Cs-137, Eu-154. Isotopes measured by ICP-MS included the isotopes of Ce, Nd, Sm, Eu, Gd, U, and Pu. These two fuel samples were close in position to each other and appeared to have the same burnup based on Nd-148 and Cs-137 concentrations. Thus, the isotopic results from the DA were very consistent between the two samples.

TABLE I

Gamma Measurement Summary from ORNL Measurements

	NA	TMI	CC	MOX
Burnup [GWd/MTU]	35-67	27-59	41	18-52
Cooling Time [years]	4.2	13.3	27.5	0.75
Initial Enrichment	4.19% U-235	4.0% U-235	3.038% U-235	4.4% Pu (WG)
Cs-137 ($T_{1/2}$=30 y)	All Coax	All Coax	All Coax	All Coax
Cs-134 ($T_{1/2}$=2.06 y)	All Coax	All Coax	All Coax	All
Eu-154 ($T_{1/2}$=8.59 y)	All	All	All Coax	All
Ce-144* ($T_{1/2}$=284 d)	High BU Coax	None	None	All
Sb-125 ($T_{1/2}$=2.76 y)	High BU Coax	All Coax	None	All
Ag-110M ($T_{1/2}$=249 d)	High BU Coax	None	None	All Coax
Rh-106 ($T_{1/2}$=373 d)	All Coax	None	None	All Coax
Co-60 ($T_{1/2}$=5.26 y)	All Coax	All Coax	All Coax	All Coax
Bi-214	All Coax	All Coax	All Coax	All Coax
U x-ray	All	All	All Coax	All
Pu x-ray	All Planar	All Planar	All Planar	All
Eu-155 ($T_{1/2}$=4.76 y)	All Planar	All Planar	All Planar	All
Am-241 ($T_{1/2}$=432 y)	None	All Planar	All	Low BU Planar

Coax measurements are broad energy, planar measurements are low energy.

*Ce-144 is usually identified by the 2185 keV peak, but none of these measurements extended to this high of an energy.

REACTOR PHYSICS SIMULATIONS

The CC fuel pin locations were simulated using SCALE 6.0, TransLAT, Monteburns, and Origen-Arp based on the fuel history data from Ref. 8. This is a PWR reactor with CE 14 x 14 fuel assemblies. The fuel pin measured was pin MKP109 from assembly D047. This fuel had an initial enrichment of 3.038 wt% ^{235}U, a burnup of 41.8 GWd/MTU, and a cooling time of 27.5 years.

For use in a sensitivity analysis, a representative PWR 17x17 fuel pin with 4% initial enrichment, 45 GWd/MTU burnup, and 10 years cooling time was used as a reference case. ORIGEN 2.2 was used to perturb this base case to look at effects of change in several fuel parameters on isotopes of interest. The main fuel parameters for both fuel pins analyzed are shown in Table II.

In each simulation, concentrations of nuclides visible in NDA measurements (from Table I) and key neutron absorbers were tracked for comparison both between codes and with NDA and DA measurements. Key neutron absorbers include ^{241}Am, ^{133}Cs, ^{155}Gd, ^{241}Pu, ^{149}Sm, ^{151}Sm, ^{152}Sm, ^{99}Tc, ^{131}Xe, and ^{91}Zr. While some of them will be negligible in the Calvert Cliffs fuel being measured due to the combination of short half-lives and the long cooling time, they were still tracked for comparison with future simulations of other fuels.

TABLE II

Fuel Parameters for Calvert Cliffs and Reference Fuel Pins

	Calvert Cliffs	Reference Fuel
Assembly Type	CE 14x14 PWR	17x17 PWR
Initial Enrichment [wt% 235]	3.038	4
Burnup [GWd/MTU]	41.8	45
Cooling Time [years]	27.5	10

SIMULATED TO EXPERIMENTAL COMPARISONS

In order to determine which reactor physics code best predicted the specific isotopes of interest for this research, simulation results for all four codes were compared to the DA results. The simulated and DA values were all normalized per gram of uranium. The following isotopes were considered for comparison: ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{137}Cs, ^{154}Eu, ^{241}Am, ^{155}Eu, ^{149}Sm, and ^{155}Gd. The percent error between the calculated and DA values for each of these isotopes relative to total uranium is shown in Figure 1.

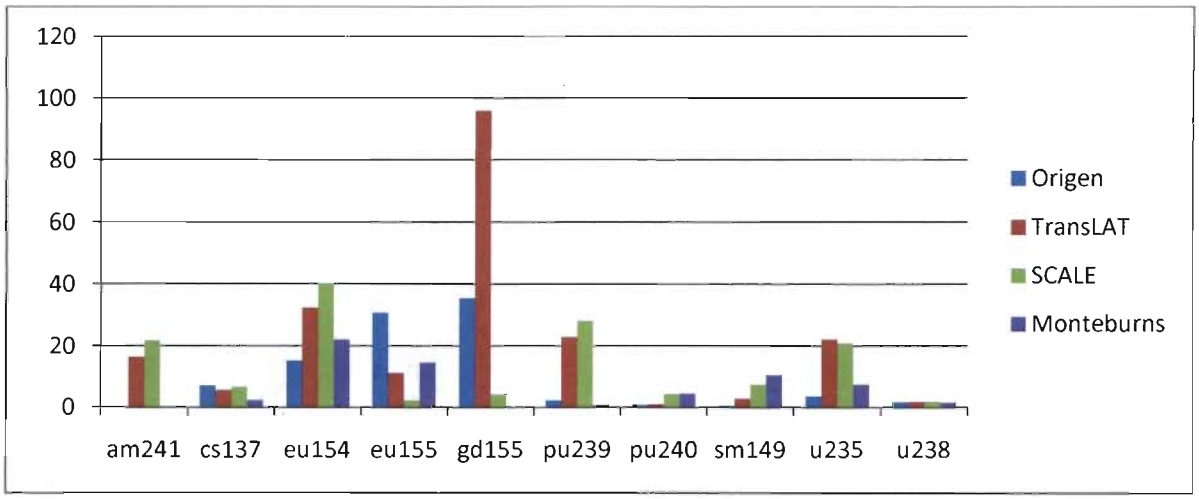

Figure 1. Percent error in calculated results compared to DA results for selected isotopes each code considered.

TABLE III

Cumulative Error Results for Each Reactor Physics Code

Simulation Code	CE
ORIGEN-ARP	0.985
TransLAT	2.140
SCALE	1.391
Monteburns	0.657

To provide a common figure of merit to judge each code, a cumulative error for each code for the 10 isotopes considered was used. The cumulative error was calculated using:

$$CE = \sum_{i=1}^{10} \left| \frac{DA_i - C_i}{DA_i} \right|$$

where CE is the cumulative error for the code, DA_i is the DA result (in grams per gram of fuel) for isotope i, and C_i is the calculated result (in grams per gram of fuel) for isotope i. Thus, a smaller CE value would suggest a more accurate code for calculating these isotopes. The CE values for each code are shown in Table III. Based on this figure of merit, Monteburns was judged to be the most accurate simulation code for this application. Calculate to experimental ratios for each of the isotopes considered for Monteburns are shown in Table IV.

TABLE IV

Comparison of Monteburns to DA Ratios for Ratios of Isotopes

C/E	Ratio
Pu/U	1.00
Am-241/U	1.02
Cs-137/U	1.05
Eu-154/U	1.24
Eu-155/U	1.17
Gd-155/U	1.02
Sm-149/U	0.91
U-235/U-238	0.94
Pu-240/Pu-239	0.96
Nd-148/U	0.99

NDA TO DA COMPARISONS

In the Calvert Cliffs fuel broad energy measurements, ^{154}Eu and ^{137}Cs are the only two easily visible fission product nuclides due to the long cooling time. From the NDA measurements, the ^{154}Eu/^{137}Cs ratio was calculated to be 0.00383, which has a 2.3% error from the DA value of 0.00393. This is generally considered to be excellent agreement.

The uranium and plutonium x-rays are not emitted spontaneously, but are created when higher energy photons interact with tightly bound uranium and plutonium electrons. In addition, the plutonium is not evenly distributed throughout the fuel; it is almost all on the very edge of the fuel pin. Figure 2 shows the radial distribution of plutonium in nine exponentially distributed regions for the Calvert Cliffs fuel as modeled in monteburns. This makes it very difficult to compare gamma measurements for these x-ray peaks (which are only measuring the signal from the outer edge of the fuel pin due to the low energy) to DA measurements of a solution of the entire fuel pin.

In this 30 year old fuel, most of the x-rays are stimulated by ^{137}Cs 661 keV photons. To verify the Pu/U ratio obtained in the NDA measurements, the fuel was simulated in MCNP. The fuel composition in each

of nine equal volume regions was obtained from Monteburns, and 661 keV photons were simulated in each region relative to the quantity of ^{137}Cs in that region. A surface tally was used outside the cladding to look at the ratio of plutonium to uranium x-rays. The ratio obtained from MCNP agreed very well with the measured value from gamma measurements with an error of 10%. This verifies that Monteburns is adequately calculating the plutonium distribution throughout the pin. If more burnups were obtained for this geometry, a calibration could be used to correlate between bulk plutonium to uranium ratio and NDA plutonium to uranium x-ray signal ratio.

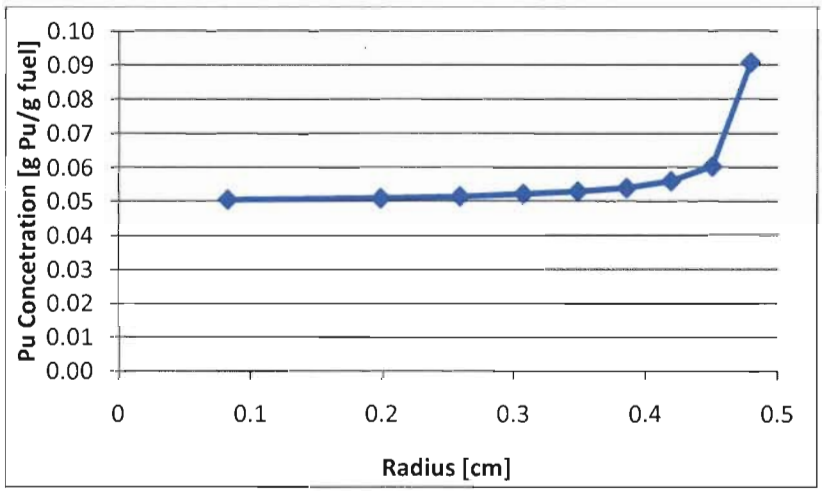

Figure 2. Calculated radial distribution of plutonium concentration in Calvert Cliffs fuel.

SENSITIVITY ANALYSIS

In order to determine how much knowledge about the history of spent nuclear fuel can be obtained from photon measurements, it is necessary to determine how each signal depends on fuel history parameters. There are many parameters which could change isotopic composition of the fuel. These parameters include burnup, cooling time, power level of the last burn cycle, total days of shutdown before the last burn cycle, initial enrichment, moderator temperature and density, cladding composition and thickness, fuel assembly pitch, and burnable poisons. For this work, the first four of these parameters have been investigated for a range of isotopes using ORIGEN 2.2 which is the burnup code used in Monteburns. A typical reference case of PWR fuel with initial enrichment of 4%, 45 GWd/MTU burnup from three cycles of 405.45 days at 37 MW/MTU with shutdowns of 60 days between each cycle and a cooling time of 10 years was used as the base case for these simulations. For each of the parameters investigated, a sensitivity coefficient was calculated for each isotope to estimate how dependent the final concentration of each isotope is on each fuel parameter. These sensitivity coefficients were calculated using:

$$S_{B,i} = \frac{\left(c_i^{54GWd} - c_i^{32GWd}\right)}{54-32} \tag{1}$$

$$S_{P,i} = \frac{\left(c_i^{45W/g} - c_i^{26W/g}\right)}{45-26} \tag{2}$$

$$S_{S,i} = \frac{\left(c_i^{240d} - c_i^{60d}\right)}{240 - 60} \tag{3}$$

$$S_{C,i} = \frac{\left(c_i^{15y} - c_i^{5y}\right)}{15 - 5} \tag{4}22$$

where $S_{B,i}$, $S_{P,i}$, $S_{S,i}$, and $S_{C,i}$ are the sensitivity coefficients for isotope i for burnup, power level of last cycle, shutdown days, and cooling time, respectively. c_i^x is the density of isotope i at with x being the value of the parameter being studied. A summary of these sensitivity value are shown in Table V. A larger value means that the concentration of that isotope changes more drastically in the range of the parameter specified. Comparisons between different parameters is not necessarily meaningful, since the ranges of each parameter are not comparable. The following general conclusions can be made from this sensitivity analysis:

1. All of the isotopes except Total U, Total Pu, and ^{241}Am are strong functions of burnup. Total U is almost completely insensitive to burnup. Total Pu and ^{241}Am are moderately sensitive to burnup.
2. Most isotopes are insensitive to the power level of the last cycle. ^{144}Ce, ^{106}Ru, ^{125}Sb, and ^{134}Cs are all sensitive to the power level of the last cycle (in decreasing order of sensitivity). This suggests that ^{144}Ce and ^{106}Ru would be good measures for determining the power level of the last cycle.
3. Most of the isotopes are insensitive to the shutdown time length. Only, ^{144}Ce, ^{106}Ru, ^{125}Sb, and ^{134}Cs are sensitive to shutdown time length. Also, these sensitivities correlate to the short half-lives of the isotopes.
4. As expected, the sensitivity coefficient for cooling time generally tracks with the inverse of the half-life of the isotope(s) analyzed.
5. Both ^{137}Cs and Total Pu are sensitive only to burnup and therefore both make excellent burnup monitors.
6. A strong correlation exists between sensitivities to power level of the last cycle and shutdown time length (i.e., those isotopes with strong sensitivities to one parameter also have strong sensitivities to the other). This will make unfolding the two parameters difficult.
7. Monitors for power of the last cycle, shutdown time length, and cooling time all require a burnup monitor be used first to eliminate the dependence on burnup.

TABLE X

Sensitivity Coefficients for Isotopes Visible in Spent Fuel Gamma-Ray Measurements Based on ORIGEN 2.2 Simulations.

Isotope	$S_{B,i}$	$S_{P,i}$	$S_{S,i}$	$S_{C,i}$
^{137}Cs	30.2088	0.8103	-0.0356	-29.6373
^{134}Cs	0.2755	0.0396	-0.0008	-3.2492
^{154}Eu	1.1637	0.0280	-0.0003	-2.3164
^{155}Eu	1.1751	0.0486	-0.0005	-4.2379

^{144}Ce	0.0010	0.0009	0.0000	-0.4440
^{125}Sb	0.0398	0.0088	-0.0003	-0.4405
^{106}Ru	0.0074	0.0027	-0.0001	-0.6312
^{241}Am	9.6225	-0.8654	0.0152	49.3957
Total U	-1236.3741	0.0000	0.0000	0.0000
Total Pu	83.1771	0.8877	0.0468	-51.4395

SUMMARY

Passive gamma measurements have been obtained for a range of spent nuclear fuel pins in order to determine the visible isotopes in either broad-range or low-energy gamma measurements. SCALE, Monteburns, TransLAT, and ORIGEN have been used to model these visible isotopes as well as a selection of neutron absorbers in one fuel measurement campaign for comparison to DA results. Monteburns was found to be the most accurate code for the isotopes considered to be important for this study. A sensitivity analysis was performed on a separate hypothetical reference fuel pin with ORIGEN in order to determine how dependent the concentration of each of the nuclides visible in gamma-ray measurements are on burnup, power level, shutdown time, and cooling time.

Future work will include investigating sensitivity of additional fuel parameters as well as looking into sensitivity of neutron absorbers not visible in NDA gamma spectroscopy measurements. These sensitivity results will be used to explore a method to calculate fuel parameters and possibly concentrations of neutron absorbers from NDA gamma spectroscopy measurements.

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