

INVESTIGATION OF PASSIVE GAMMA SPECTROSCOPY TO VERIFY SPENT NUCLEAR FUEL CONTENT*

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ABSTRACT

Measurement of passive gamma rays emitted by the decay of radioactive isotopes is a widely used and fundamental technique to verify and determine characteristics of nuclear materials. Several initiatives are currently under way to investigate the use of advanced nondestructive assay methods for application to independently measure the fissile material and plutonium content in spent nuclear fuel. Although no single technique is capable of accurately measuring the actinide inventory, the integration of multiple measures has the potential to dramatically improve nondestructive analysis capability. These methods are likely to involve the use of high-resolution gamma detectors with the resolving power necessary for isotopic identification. Recent high-resolution gamma-ray spectroscopy measurements of spent nuclear fuel rod segments performed at Oak Ridge National Laboratory provided an important opportunity to analyze and identify the dominant and minor gamma-ray signatures that can be acquired using conventional HPGe detectors. The spent fuel source and detector spectra were simulated to identify potential low-intensity gamma signatures in the fuel that could be observed with modest improvements in detector capability. The measurements represent important experimental benchmarks for code and detector model simulations that will be needed to better determine advance detector design requirements for spent fuel measurements.

INTRODUCTION

The nuclear material content in spent fuel cannot be accurately verified at present without first destructively analyzing the fuel. Material accounting of spent fuel at nuclear facilities relies extensively on the utility's declaration of the fuel assembly content derived from reactor burnup calculations. There is currently no method that is independent of utility records to determine the fissile material or plutonium mass using nondestructive analysis (NDA) methods available for deployment in the field. Due to the high cost and long measurement times associated with destructive fuel measurements, there is considerable research interest in passive- and active-interrogation NDA methods. These methods use neutron and gamma emissions [1] as a means to strengthen capabilities of the International Atomic Energy Agency to verify and confirm inventories of spent nuclear fuel at nuclear power plants and to verify inventories in assemblies received at a reprocessing plant for material accountability.

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Gamma-ray spectrometry is a primary technique of importance to material control and accounting (MC&A). Gamma measurements are routinely used as a monitoring technique to provide gross and spectroscopic information on irradiated fuels [2,3]. Such systems already exist in many nuclear facilities, they are relatively small and noninvasive, and they provide real-time data. However, current applications are limited by the resolution and efficiencies that can be achieved with existing detector technology. The 2010 Nuclear Energy Research and Development Roadmap specifically addresses the need to develop advanced safeguards instrumentation, including active neutron and photon interrogation methods and advanced passive detectors such as ultra-high-resolution gamma spectrometers with sufficient resolving power for isotopic identification. Accurate modeling and simulation of the spent fuel decay and radiation emissions are critical to identifying the potential signatures and observables in irradiated materials and determining the design requirements (e.g., resolution and efficiency) for state-of-the-art detectors needed to quantify these signatures for nuclear process monitoring and MC&A measurements.

High-purity germanium (HPGe) detector gamma measurements of spent nuclear fuel rod segments from several commercial reactors performed at Oak Ridge National Laboratory (ORNL) recently provided an opportunity to analyze high-resolution spectra for potential signatures using modeling and simulation codes. This work specifically focused on analysis of gamma rays with energies > 400 keV emitted predominantly by fission products. Analysis of the lower-energy gamma rays and X-rays as a method to directly measure plutonium in spent fuel is the focus of other work reported at this conference [4].

The primary objectives of the present work include (1) experimentally benchmarking the capability of modeling and simulation tools to accurately calculate gamma-ray spectra measured by existing gamma detector systems and (2) identifying potential lower-intensity signatures that could be resolved with modest improvements in resolution and reductions in background from Compton scattering. Another application of this work relates to the use of inverse depletion methods that derive the properties of irradiated nuclear material by indirect means using signatures of radionuclides generated by fission and activation. Previous work has demonstrated that a limited number of nuclides can provide a unique fingerprint of its initial pre-irradiation content and its irradiation and decay history. The present study identifies what isotopes can be detected with existing HPGe detector technology and what could potentially be identified using high-energy gamma spectroscopy. Additional work on the application of inverse spectroscopy is ongoing and the subject of another paper [5].

IRRADIATED FUELS

The fuels studied in this work were irradiated in the Three Mile Island Unit 1 (TMI-1) Nuclear Generating Station and the North Anna Nuclear Generating Station. Fuel segments of fuel rods were examined at the Irradiated Fuel Examination Laboratory (IFEL) at ORNL. The Three Mile Island rod segments came from rods D5 and H6 of 15x15 Mark-B8V fuel assembly NJ05YU, with an initial enrichment of 4.013 wt % ²³⁵U and burnup of approximately 50 GWd/t. The assembly was irradiated two cycles and discharged after cycle 10 on September 8, 1995.

The North Anna fuel rod segments came from rod A8 of a 15x15 Mk-BW assembly design with an initial enrichment of 4.199 wt % ^{235}U . This assembly was irradiated for four consecutive cycles and discharged after cycle 16 on May 2, 2004. The measured rod achieved a very high burnup of approximately 70 GWd/t.

GAMMA MEASUREMENTS

Gamma measurements of the Three Mile Island and North Anna reactor fuel segments were made using an HPGe planar detector for detailed investigation of lower-energy gamma and X-ray emissions below 400 keV and a high-resolution HPGe coaxial detector to measure the gamma energy range from about 60 keV to 2.5 MeV. The present study focused on the high-energy gamma-ray spectra measured using the coaxial detector above 400 keV. Other investigations are currently analyzing the low-energy X-ray region measured by the planar detector.

The gamma measurements were performed at IFEL hot cell facilities in March 2008 and February 2010 using a highly collimated port and detector instrumentation installed outside the cell. At the time of measurement, the North Anna fuel had decayed for about 4 years, and the Three Mile Island fuel had decayed for 13 years. The counting times for the gamma measurements were relatively short—about 3 min each.

MODELING AND SIMULATION

High-resolution gamma spectra were calculated based on the detailed fuel and assembly design and operating data for the North Anna and TMI-1 fuel segments to provide direct comparisons with measurements for the purposes of spectral analysis and peak identification. Calculations were performed using the current version of ORIGEN that is distributed with the SCALE 6.0 code system [6]. ORIGEN explicitly tracks the time-dependent isotopic concentrations and activities for more than 2000 isotopes formed by fission, neutron transmutation, radioactive decay, and activation. ORIGEN has been extensively validated against the results of destructive radiochemical isotopic analysis [7] for more than 100 spent fuel samples and includes experimental data for about 20 actinides (isotopes of U, Pu, Np, Am, and Cm) and 40 fission products (isotopes of Ce, Nd, Pm, Sm, Eu, Gd, Sr, Cs, Mo, Tc, Ru, Rh, Ag, Sb, and I). The experimental fuels include properties that cover many modern commercial fuel designs and destructive measurements of samples from TMI-1 assembly NJ05YU [8].

The current version of ORIGEN includes a detailed gamma- and X-ray emission database of line energies for 1132 nuclides, and includes discrete gamma lines due to β^- , β^+ , α , IT, and ϵ decay and discrete X-rays due to β^+ annihilation radiation, fluorescence following K-capture (ϵ), or emission of conversion electrons. Continuous spectra from internal bremsstrahlung processes and theoretical nuclear decay models are represented. Spontaneous fission gamma rays and gamma rays from (α ,n) reactions are also included. The photon emission line data are binned into an arbitrary user-defined group structure using the calculated activities for 904 activation products, 174 actinides, and 1149 fission products. The calculated gamma-ray fission product spectra have been validated for decay times less than 2 s after fission [9]. ORIGEN also includes neutron source methods for spontaneous

fission, (alpha,n) reactions, and delayed neutrons based on methods and nuclear data adopted from the SOURCES-4C code developed at Los Alamos National Laboratory [10].

In this study spent-fuel gamma-ray spectra were calculated in approximately 2000 energy groups, resulting in an energy resolution comparable to the HPGe detector measurements. ORIGEN also lists the dominant gamma sources in each energy bin range, which can be used to identify gamma peaks in highly complex spectra. The ORIGEN code provides the data on the gamma emission source in the irradiated fuel.

The detector measurements were simulated using the emission spectra calculated by ORIGEN and the Gamma Detector Response Analysis Software (GADRAS) [11] to simulate what the HPGe detector actually measures. In this application the isotope gamma spectral database in the GADRA code was not used. Instead, a pseudo nuclide was defined in GADRAS with emission lines and intensities calculated by the ORIGEN code for all nuclides in the spent fuel. This enabled the aggregate gamma spectrum calculated by ORIGEN for the complete set of isotopes in spent fuel to be used directly in the detector calculations. Because GADRAS is currently limited to 256 lines per isotope, six pseudo nuclides were used to define the ORIGEN spectrum using 2000 energy channels that contained 1474 lines.

GAMMA SPECTROSCOPY ANALYSIS RESULTS

The simulated gamma spectrum for the TMI-1 fuel segment is compared to the measured HPGe spectra in Figure 1 over the full energy range. The measured and predicted spectra are offset in the vertical axial (counts) for purposes of qualitative comparison of the spectra. The predicted spectrum was based on the gamma emission rates in the fuel calculated by ORIGEN and the detector response from GADRAS. The calculations did not include self-attenuation of gamma rays in the fuel or absorption and scattering in the collimator. Consequently, the predicted spectrum was somewhat higher than measurement in the low-energy region (relative to the high-energy region).

All of the major features of the measured spectrum were simulated, including many of the low-intensity lines. The measurements included so-called sum peaks that were not in the calculated spectrum. The sum peaks are caused when multiple gamma rays are emitted nearly simultaneously, typically in a decay cascade, such that they cannot be resolved in time and appear as coincident gamma rays, resulting in a peak with an energy equal to the sum of the different gamma-ray energies. Activated ^{60}Co ($E_\gamma = 1173$ and 1332 keV) was also observed in the measured spectra but was not predicted because impurity ^{59}Co was not included in the model simulations. The ^{60}Co was primarily observed in the activated cladding. However, accurately modeling activation requires values of the initial impurity levels, which can be difficult to obtain. The background peak from ^{40}K ($E_\gamma = 1461$ keV) was also observed only in measurements.

The major peaks in the spectra were those typically observed in spent fuel measurements: predominantly ^{137}Cs , ^{134}Cs , and ^{154}Eu . These fission products have been widely used in previous studies for fuel characterization and burnup determination. However, many other minor peaks were observed in the spectra that might provide additional information on the

content of the fuel. A more detailed spectroscopic analysis was therefore performed using the TMI-1 spectrum to determine what, if any, additional information could be acquired from low-intensity signals using capabilities of existing high-resolution detectors and whether additional information could be obtained with modest improvements in detector resolution and background scatter suppression.

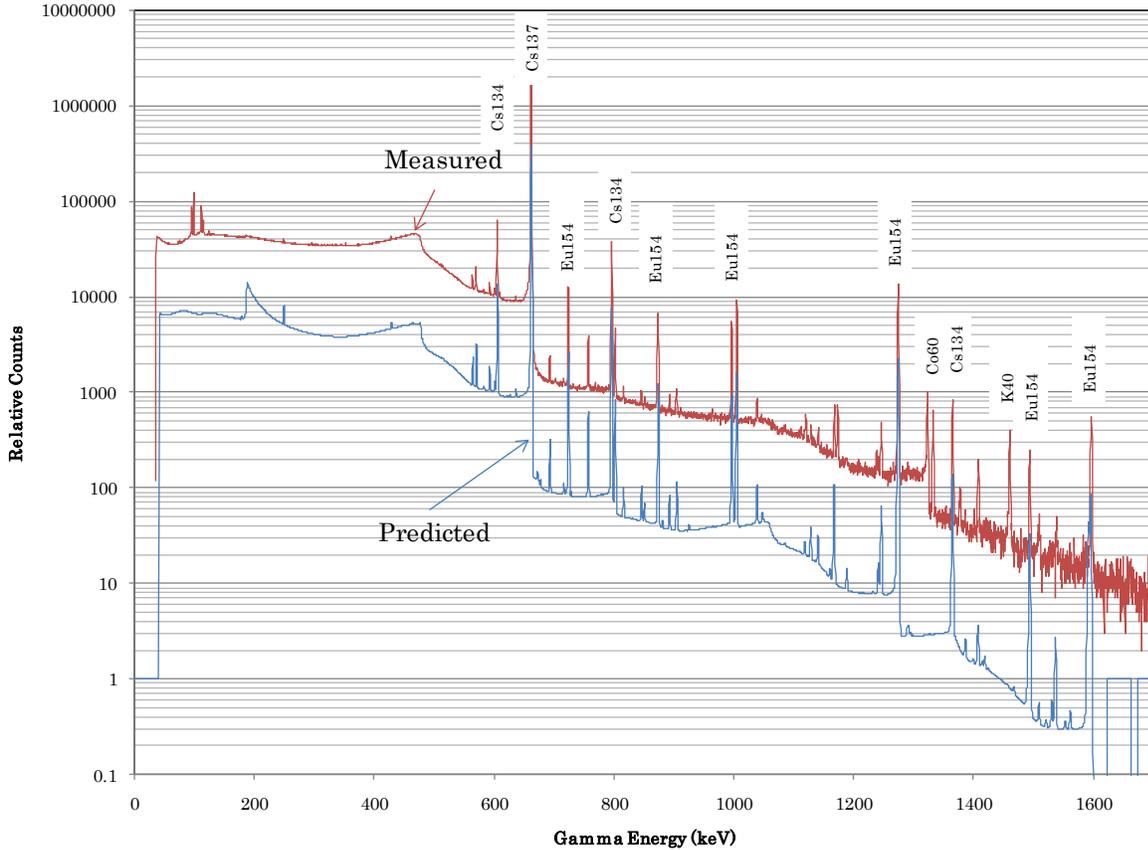


Figure 1. Measured Three Mile Island spent fuel segment gamma spectrum compared to predicted detector response simulated using the ORIGEN and GADRAS codes. (Spectra are arbitrarily offset in the vertical axis for purposes of comparing the results.)

The calculated gamma emission lines from the ORIGEN simulations of the TMI-1 fuel were compared to measurements in more detailed 400 keV intervals. The spectra between 400 keV and 1.2 MeV are compared in Figure 2. The major lines are identified, and minor lines are shown for signatures that approach the level of detection. More than 40 gamma lines were clearly identifiable. However, analysis of the lines indicated that they were attributable to only six fission products: ^{134}Cs , ^{137}Cs , ^{154}Eu , ^{125}Sb , ^{106}Rh , and ^{85}Kr . The isotope ^{154}Eu alone emits more than 150 discrete lines, of which 26 lines were observed in the TMI-1 spectra. The list of identified fission products and gamma energies from the spectra is provided in Table 1.

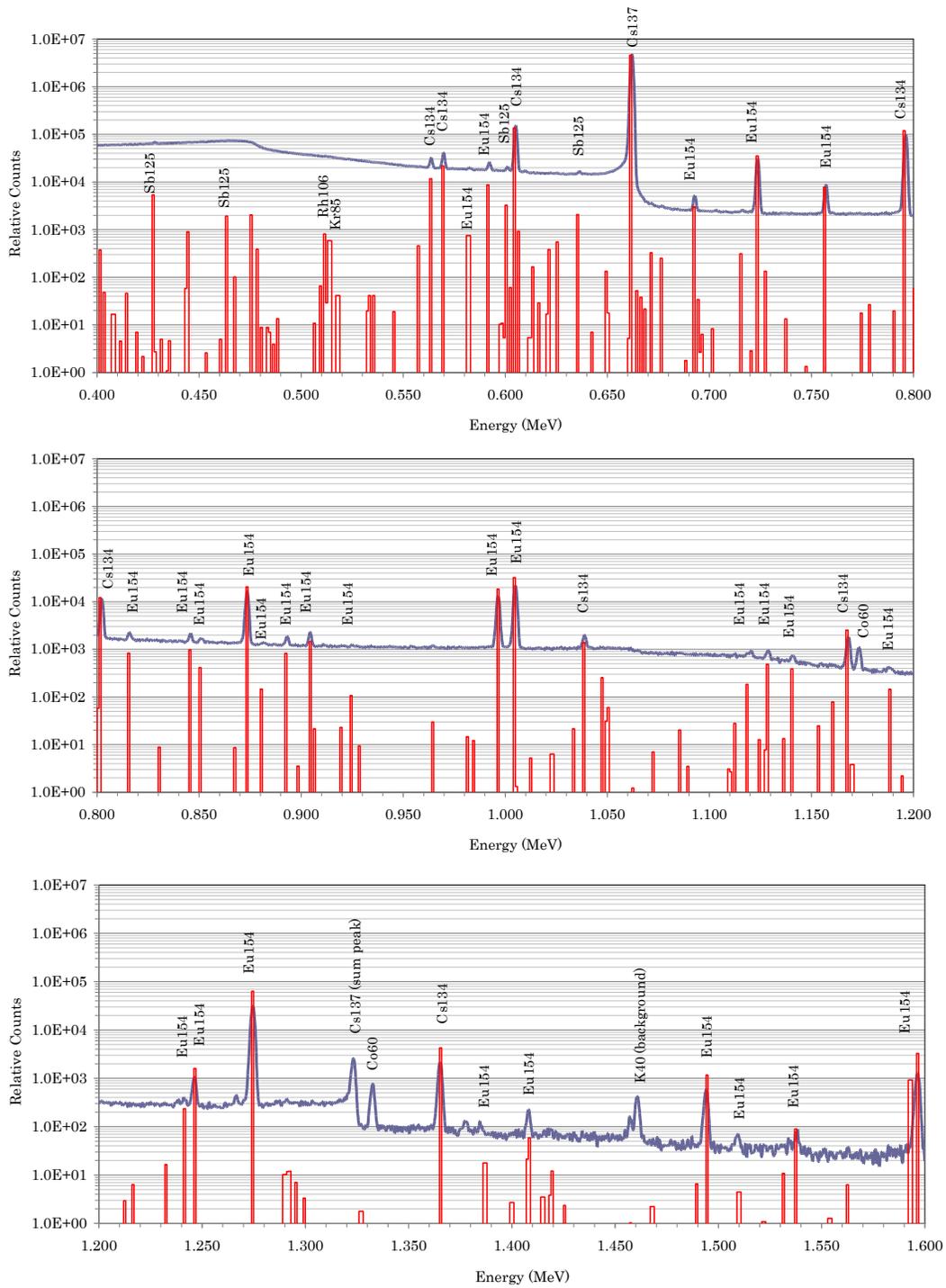


Figure 2. Three Mile Island fuel gamma spectrum measured (blue line) and calculated emission lines (red histogram) with 13-year cooling time.

Table 1. Nuclides and gamma peaks identified in spent fuel spectra

Isotope	Gamma Energy (keV)
Cs-137	661
Cs-134	475*, 563, 569, 604, 795, 801, 1038, 1167, 1365
Eu-154	247, 582, 591, 692, 723, 756, 815, 845, 850, 873, 880, 893, 904, 996, 1004, 1118, 1128, 1140, 1189, 1241, 1246, 1274, 1408, 1494, 1538, 1596
Sb-125	427, 463, 600, 606, 635
Rh-106	511, 621*, 1050*
Kr-85	513
Pr-144	696*, 1489*

* Gamma energies observed in only the North Anna spectra.

A similar analysis of the 3-year cooled North Anna fuel spectra identified one additional nuclide, ^{144}Pr , not observed in the TMI-1 spectra due to the 285 d half-life of the ^{144}Pr decay precursor, ^{144}Ce . For longer cooling times it will be increasingly difficult to detect the low activity of these isotopes.

The nuclides in Table 1 represent those that can be measured in spent nuclear fuel during interim storage or at receipt of the spent fuel reprocessing facility. The application of inverse depletion analysis tools to indirectly determine the concentrations of key spent fuel properties has been successfully demonstrated using absolute fission product concentrations and fission product isotopic ratios. Additional studies to perform the analysis using the measured fission products listed in Table 1 remain to be performed.

CONCLUSIONS

The dominant gamma emissions from ^{137}Cs , ^{134}Cs , and ^{154}Eu were observed in addition to minor peaks from ^{106}Ru - ^{106}Rh , ^{144}Ce - ^{144}Pr , and ^{125}Sb for fuel with short cooling times. Computational simulations of the gamma spectra were performed to identify many of the weakest lines that could potentially be resolved with modest improvements in the detector efficiency or configurations to reduce Compton scatter background.

This work found that gamma-ray signatures beyond the commonly seen high-intensity fission products in spent fuel are not likely to be observed even with improvements in HPGe detector configurations to significantly reduce background. Most of the weak emissions (both observable peaks and those below the background level) are associated with the decay emissions of the dominant fission products. This work was limited to the analysis of gamma rays with energies > 400 KeV. Investigation of gamma spectra for the low-energy X-ray region and active neutron interrogation to generate delayed gamma rays by induced fission are areas of gamma spectroscopy that appear to show the most promise for fissile material verification in spent fuel.

This work also validates the computational capability to accurately simulate high-precision spectra from irradiated fuel. These computational methods would also be applicable to the

analysis of passive gamma-ray signatures from active interrogation of spent fuel involving very short decay times (e.g., seconds) after fission.

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