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LLNL-TR-662329

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October 9, 2014

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This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

DELAYED GAMMA-RAY SPECTROSCOPY FOR NON-DESTRUCTIVE ASSAY OF NUCLEAR MATERIALS

LL13-Delayed Gamma-PD2La

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1. INTRODUCTION

This project addresses the need for improved non-destructive assay techniques for quantifying the actinide composition of spent nuclear fuel and for the independent verification of declared quantities of special nuclear materials at key stages of the fuel cycle. High-energy delayed gamma-ray spectroscopy following neutron irradiation is a potential technique for directly assaying spent fuel assemblies and achieving the safeguards goal of quantifying nuclear material inventories for spent fuel handling, interim storage, reprocessing facilities, repository sites, and final disposal. Other potential applications include determination of MOX fuel composition, characterization of nuclear waste packages, and challenges in homeland security and arms control verification.

2. DELAYED GAMMA-RAY SPECTROSCOPY METHOD

The delayed gamma-ray spectroscopy method, schematically shown in figure 1 for a measurement on used nuclear fuel, is an active interrogation method that employs a neutron source to irradiate materials and inducing fission for periods ranging from seconds to minutes. The irradiation is followed by a spectroscopic measurement of the beta-delayed gamma rays emitted from short-lived fission products. The energies of the delayed gamma rays (blue) extend far beyond the intense passive background from long-lived fission products in used fuel (green), which is negligible above ~3.2 MeV. In other than used fuel applications background levels are much lower so that more-intense lower-energy delayed gamma-ray lines can be utilized for more efficient measurements. Because the relative peak intensities and structures in the delayed gamma-ray spectra are characteristic for each fissionable isotope, the delayed gamma-ray spectrum can be analyzed as a superposition of contributions from ^{235}U, ^{239}Pu, ^{241}Pu, and ^{238}U and the isotopic fractions can be determined if the delayed gamma-ray spectra of the individual isotopes are accurately known.

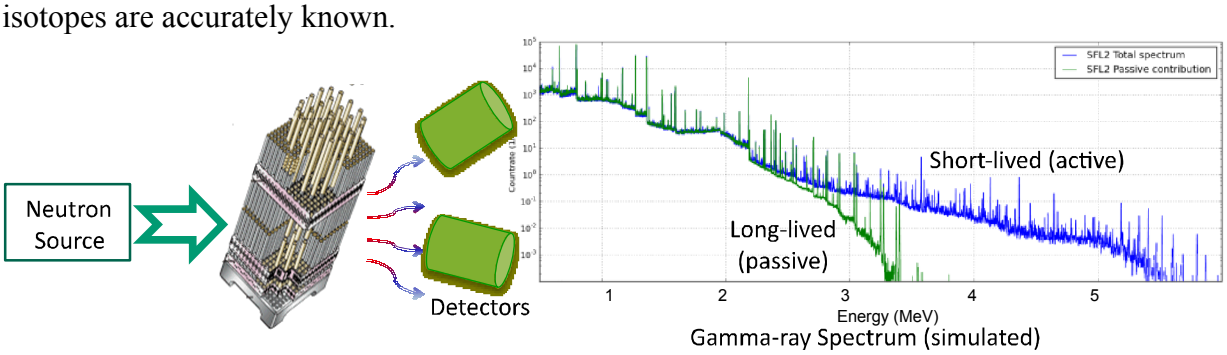

Figure 1. Delayed gamma-ray measurement on a spent fuel assembly.

3. RESEARCH PROGRAM

The overall goal of this project is to assess the potential and merits of the delayed gamma-ray spectroscopy method. The research has two key components: a strong experimental campaign to characterize the delayed gamma-ray signatures for specific isotopes and combined targets, and a closely linked modeling effort to assess system designs and applications. The experiments provide data to determine relative fission yields, to evaluate nuclear data libraries for gaps and accuracy, and to validate modeling predictions. Modeling capabilities are added to the existing framework and codes have been adapted as needed for analyzing experiments and assessing application-specific assay concepts. Detailed signature knowledge is essential for analyzing the delayed gamma technique and its capabilities, optimizing measurement parameters, and specifying neutron source and gamma-ray detection system. As an example of the latter, in applications that involve highly radioactive materials such as spent nuclear fuel, detector signal rate capabilities are critical. High-purity germanium (HPGe) detectors and fast $LaBr_3$(Ce) scintillation detectors have been investigated and their relative merits evaluated.

Several experimental campaigns were conducted at the Idaho Accelerator Center (IAC) using a photo-neutron source driven by a pulsed electron linac to irradiate ^{235}U, ^{238}U, ^{239}Pu, and combined ^{235}U/^{239}Pu targets. Delayed gamma-ray spectra were collected for different irradiation/detection time patterns ranging 15/30 minutes to 10/10 seconds that emphasized delayed gamma-ray lines from fission fragments with different half-lives. The experimental data are stored on a server at the IAC and made available for analysis to researchers participating in the project and others on request.

A second measurement campaign has been conducted using a thermal neutron beam at the Oregon State University (OSU) TRIGA reactor to perform delayed gamma-ray emission measurements on ^{235}U and ^{239}Pu targets for irradiation/detection times ranging 10/10 minutes to 10/10 seconds.

In addition to the measurements on targets of several grams, the experimental testing of the delayed gamma-ray method and validation of modeling was extended to the assembly level in measurements on a mock-up assembly at LANL. Using a neutron generator and HPGe and $LaBr_3$(Ce) scintillation detectors delayed gamma measurements were performed on several configurations of low-enriched and depleted uranium pins and a plutonium source.

The research has been carried out by a collaboration of researchers from four National Laboratories (LBNL, LLNL, LANL, and PNNL), Idaho State University's IAC, and OSU.

4. RESULTS, DISCUSSION AND CONCLUSIONS

Delayed gamma-ray spectra for ^{235}U and ^{239}Pu appear to be similar (see Fig. 2) but the relative peak intensities differ from peak to peak with 6 peaks in the 2650 keV to 4250 keV energy range differing by more than a factor of 3. Such differences are essential for the delayed gamma-ray technique to work.

Comparisons of measured and simulated spectra using nuclear data libraries ENDF/B-VII.1 and ENDF/B-VI.8 showed significant discrepancies due to large uncertainties in fission product yields. A procedure for extracting more accurate fission yields has been developed at PNNL that uses maximum likelihood estimators to fit the measured time-dependent delayed gamma emission. Initial results obtained from ^{235}U data measured at OSU are shown in figure 3 in comparison with nuclear data library values.

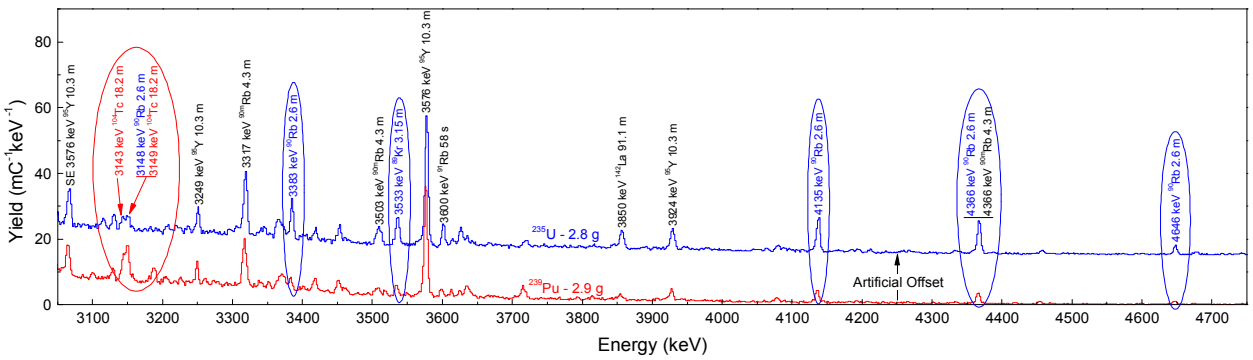

Fig. 2. Measured HPGe delayed gamma-ray spectra from 3050 to 4750 keV for targets containing ~2.8 g of ^{235}U (blue) or ~2.9 g of ^{239}Pu target (red) irradiated by moderated neutrons. Labels identify likely fission products. Circled lines differ by more than a factor of ~3.

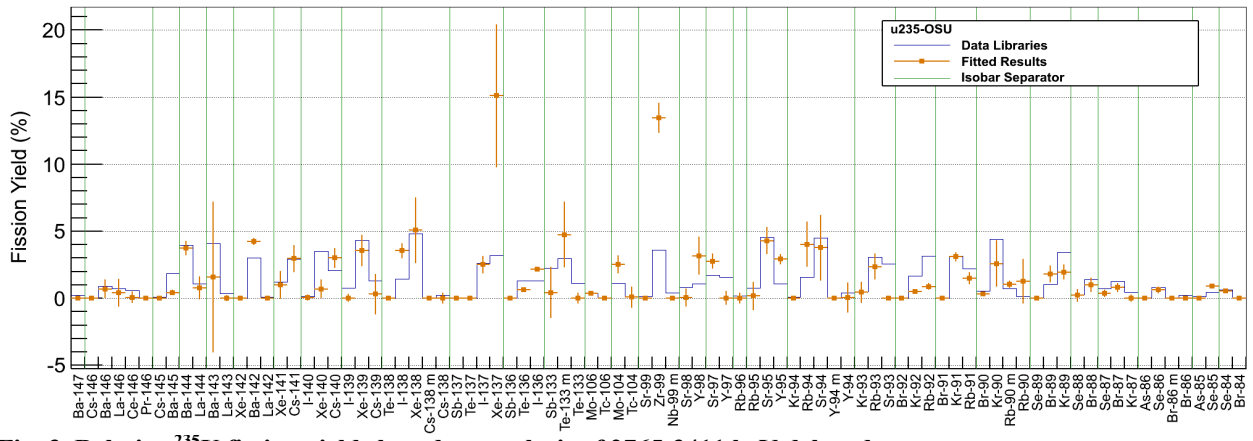

Fig. 3. Relative ^{235}U fission yields based on analysis of 2765-3411 keV delayed gamma energy range.

Several methods for quantifying isotopic ratios using basis spectra have been tested on the IAC ^{235}U/^{239}Pu data. So far using a selected number of peaks has given best results with accuracies of a few percent or better. The data suggests that cycles with irradiation/detection times of roughly 100 seconds may be optimal for delayed gamma-ray assays. As part of a system design optimization effort, a $LaBr_3$(Ce) scintillation detector with a digital data acquisition system was tested in delayed gamma-ray experiments at the IAC. The data is now being analyzed to find out if high the count-rate capability, exceeding 1 MHz, overcomes the effects of the lower energy resolution.

5. PUBLICATIONS

A. Favalli, M. Iliev, K. Ianakiev, J.L. Thron, E.T. Reedy, A.W. Hunt, V. Mozin, B. Ludewigt, "Delayed Gamma-ray Spectroscopy with Lanthanum Bromide Detector for Non-destructive Assay of Nuclear Material," *Proceedings of the INMM 55th Annual Meeting, Atlanta, Georgia USA, July 20-24, 2014.*

D.C. Rodriguez, S. Reese, R. Williford, E.T.E. Reedy, H.A. Seipel, M. Smith, V. Mozin, A. Favalli, M. Iliev, A.W. Hunt, L.W. Campbell, B.A. Ludewigt, "Utilizing Delayed Gamma-Rays to Establish Fission Yields of Short-Lived Isotopes," *Proceedings of the INMM 55th Annual Meeting, Atlanta, Georgia USA, July 20-24, 2014.*

Prepared by LLNL under Contract DE-AC52-07NA27344

LLNL-TR-662329