Compton Suppressed $LaBr_3$ Detection System for use in Nondestructive Spent Fuel Assay

S. Bender,[1,2] B. Heidrich,[1,3] K. Ünlü[1]

Service Provided: Penn State Breazeale Reactor, Neutron Beam Laboratory

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Introduction

Nuclear material accountancy is of continuous concern for the regulatory, safeguards, and verification communities. In particular, spent nuclear fuel reprocessing facilities pose one of the most difficult accountancy challenges: monitoring highly radioactive, fluid sample streams in near real-time. Current accountancy methods for nuclear fuel reprocessing facilities are resource intensive and time-consuming. The adaptation of passive gamma-ray detection coupled with multivariate analysis techniques could reduce the personnel requirements and sample processing times. In measured gamma-ray spectra from spent nuclear fuel, the Compton continuum from dominant fission product photopeaks obscure the lower energy lines from other isotopes.

The application of Compton suppression to gamma-ray measurements of spent fuel may reduce this effect and may allow other less intense, lower energy gamma-ray peaks to be detected, potentially improving the accuracy of analysis algorithms. Several investigations into the use of room temperature detectors for gamma-ray spectroscopy of spent nuclear fuel have shown that $LaBr_3$ is a suitable detector material for such applications because of its moderate resolution, room temperature operation, and fast scintillation time.

Compton suppressed spectroscopic measurements of spent nuclear fuel using $LaBr_3$ and HPGe primary detectors were performed in two configurations: as intact fuel elements through a collimator and as feed solutions to simulate the measurement of a dissolved process stream. These measurements directly assessed and quantified the differences in measured gamma-ray spectra due to the application of Compton suppression.

Experiment

The Radiation Science and Engineering Center (RSEC) at Penn State houses the Penn State Breazeale Reactor (PSBR), a TRIGA Mark-III research reactor, and an inventory of irradiated research reactor fuel spanning 48 years of operation. Several irradiated fuel elements of varying cooling time from the PSBR spent fuel inventory were measured using three Compton suppression systems that used three different primary detectors, HPGe, $LaBr_3$, and NaI(Tl), with the same 22.86 cm x 22.86 cm NaI(Tl) annular guard detector. The fuel elements evaluated varied in cooling time, or the length of time after removal from the reactor core. The fuel element characteristics are summarized in Table 1. All of the fuel elements had about the same initial enrichment (19.5-19.9%) and uranium loading (12 wt%). Fuel element 230, which had cooled for approximately one year, had a relatively short cooling time compared to the typical three-year cooling period before a power reactor fuel assembly is reprocessed. Fuel element 238, a current core rod at time of measurement, represented the case of low burnup and high activity. This fuel element was removed from PSBR core 54 loading and only allowed to cool for two days before measurement. Finally, fuel element 202 from the original PSBR core loading demonstrates the case of long cooling time and, consequently, lower activity. The fuel element burnup values were determined using the PSBR neutronics code TRIGSIMS (TRIGA Simulator-S) [10].

TABLE 1. Fuel elements examined using Compton suppression systems

Fuel Element ID	Cooling Time (y)	Average Burnup (MWd/MTU)	Dates Irradiated
202	18.8	26,150	7/13/1972 to 7/8/1994
230	0.9	19,349	10/20/1998 to 5/12/2012
238	0	5,043	6/14/2007 to 4/13/2013

One of the beam ports of the PSBR Neutron Beam Laboratory was used to measure the gamma-ray spectra from the fuel elements using the measurement geometry shown in Figure 1. A 15.24 cm long, 0.3175 cm diameter aperture lead collimator was positioned at

[1] Radiation Science and Engineering Center, The Pennsylvania State University, University Park, PA 16802
[2] Currently at the Remote Sensing Laboratory, Joint Base Andrews, MD
[3] Currently at the Idaho National Laboratory, Idaho Falls, ID

the pool-end of the beam port followed by a 73.66 cm long cement collimator with an aperture diameter of 1.27 cm. Additionally, adjustable lead shielding with a slit window was located in front of the detector to further collimate the beam. A fuel element holder, fabricated from ultra-high molecular weight polyethylene, was secured to the face of the aluminum flange to ensure the proper alignment of the fuel element during measurement. The placement of the polyethylene tube against the face of the beam port aluminum flange minimized the effect of possible water attenuation in the fuel element measurements.

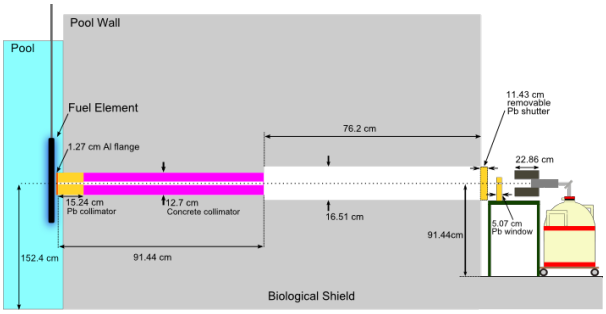

FIGURE 1: Diagram of the experimental design showing placement of the fuel rod and detectors with respect to the PSBR beam port. The HPGe detector is displayed in the diagram, but the $LaBr_3$ detectors were located in the same place when in use.

Three Compton suppressed primary detectors were used to record gamma-ray spectra emitted from the fuel elements: HPGe, 1.5"x1.5" $LaBr_3$, and 3"x3" NaI(Tl). The HPGe detector used was a standard electron (p-type) closed-end coaxial with 50% relative efficiency. Pulse pileup rejection/live time correction (PUR-LTC) circuitry in the spectroscopy amplifier (Canberra model 2026) was also used to manage the high count rate in each primary detector. Each primary detector was positioned inside a 9 in x 9 in (22.86 cm x 22.86 cm) NaI(Tl) annulus to provide Compton suppression capability. Genie 2000 software was used for data collection and analysis.

Dissolved fuel samples of Approved Test Material (ATM)-105 and ATM-109, irradiated fuel elements from commercial BWR reactors, were obtained from Pacific Northwest National Laboratory. The samples had been processed using the beginning stages of the PUREX method and represented the unseparated feed solution from a reprocessing facility. Compton suppressed measurements of the ATM fuel samples were recorded using the HPGe and $LaBr_3$ primary detectors, inside the NaI(Tl) annular guard detector, to simulate the siphoning of small quantities from the main process stream for long dwell measurement periods.

Results

A comparison between gamma-ray spectra from the same fuel element measured with each detector is shown in Figure 2, to emphasize the difference in energy resolution and the effect on the number of peaks available for analysis. No additional photopeaks were able to be resolved using NaI(Tl) as the primary detector in the measurement of any of the fuel elements.

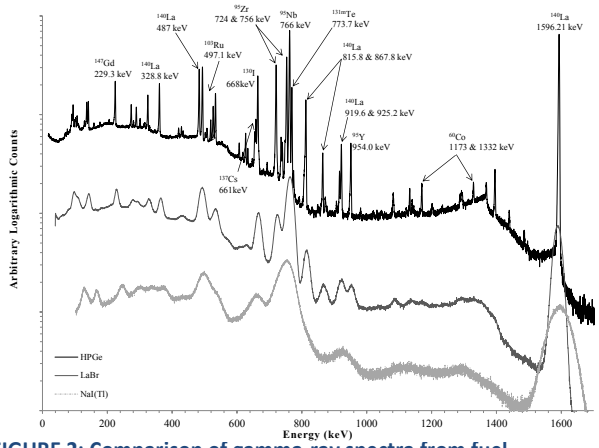

FIGURE 2: Comparison of gamma-ray spectra from fuel element 238 measured using each detector, highlighting the difference in energy resolution.

From fuel element 238, the HPGe-based Compton suppression system recorded eight additional features and the $LaBr_3$-based Compton suppression system recorded four additional features. The eight additional peaks resolved in the HPGe suppressed spectrum were 75.44 keV (^{153}Sm), 250.36 keV (^{177}Lu), 351.12 keV (^{182}Re), 557.54 keV (^{154}Eu), 1025.33 keV (^{238}Np), 1036.06 (^{132}I), 1296.27 keV (^{154}Eu), 1299.22 keV (^{152}Eu). The four additional features in the $LaBr_3$ suppressed spectrum are 1028 keV (^{238}Np), 1132 keV (^{154}Eu), 1215 keV (^{91}Sr), 1281 keV (^{170}Lu).

Four additional spectral features were observed in the spectra recorded from fuel element 230 using both the HPGe- and $LaBr_3$-based Compton suppression systems. The four additional peaks in the suppressed HPGe spectrum are present at 463 keV (^{125}Sb), 475 keV (^{134}Cs), 670 keV (^{154}Eu), and 675 keV (^{152}Eu). There are also several peaks that are detectable in the unsuppressed spectrum but for which Compton suppression can offer significantly improved resolution. The four additional peaks able to be resolved in the suppressed $LaBr_3$ spectrum are 48 keV, 427 keV (^{125}Sb), 835 keV (^{54}Mn), and 1040 keV (^{134}Cs). The peak identified at 48 keV is actually a spectral feature caused by the application of Compton suppression, which may also contribute to the multivariate analysis algorithm.

No additional photopeaks were resolved using Compton suppression with any detector system from the PSBR spent fuel element 202, cooled for approximately 19 years.

The addition of Compton suppression to the measurement of collimated spent fuel allows additional photopeaks to be resolved, even at elevated count rates. The average value of suppression factors

determined from the gamma-ray spectra measured from the intact fuel element was 7% higher for the $LaBr_3$ than for the HPGe detector. The suppression factors recorded were 2.44 and 2.27, respectively.

The measurement of the dissolved ATM fuel sample measurements showed several photopeaks from ^{137}Cs, ^{134}Cs, ^{154}Eu, ^{152}Eu, and ^{241}Am, which were appropriate based on the age of the samples. The results of the unsuppressed and suppressed gamma-ray spectra measured from the ATM fuel samples using the HPGe- and $LaBr_3$-based Compton suppression system, found in Figures 3 and 4, respectively, show that no additional photopeaks were able to be resolved using Compton suppression with the enclosed source geometry.

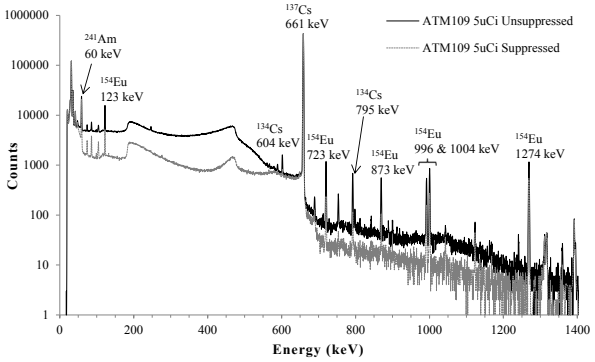

FIGURE 3: Unsuppressed and suppressed measured spectra from the 5µCi ATM 109 spent fuel samples using the HPGe-based Compton suppression system.

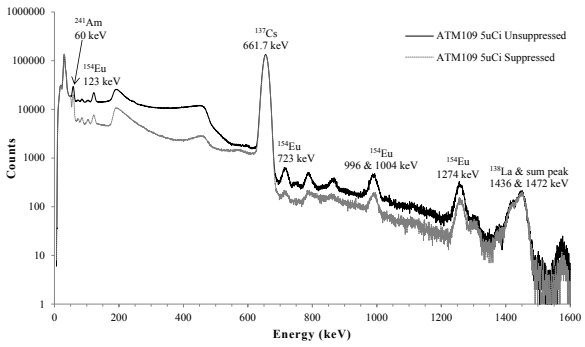

FIGURE 4: Unsuppressed and suppressed measured spectra from the 5µCi ATM 109 spent fuel samples using the $LaBr_3$-based Compton suppression system.

In measured spectra using either detector, the continuum from the ^{137}Cs 661.7 keV photopeak is suppressed, but many other peaks have significant losses in the number of recorded counts. These photopeak losses were observed because spectra were recorded with the source in very close proximity to the primary and annular guard detectors, so the coincidence detection probability was very high. This leads to sum peaks in both the suppressed and unsuppressed spectra and losses to photopeak counts in the suppressed spectra. The ^{154}Eu and ^{152}Eu peaks were affected the most by the peak losses with Compton suppression activated. These nuclides are cascade decay emitters that give off several photons during de-excitation, which can be rejected during Compton suppression counting as scattered photons. The photopeak losses to cascade decay emitters was not observed in the PSBR fuel element spectra because of the differing measurement geometries. Though the enclosed source geometry is optimal for a Compton suppression system for the measurement of low count rate samples, measurement of high count rate samples leads to sum peaks in both the suppressed and unsuppressed spectra and losses to photopeak counts in the suppressed spectra.

These experiments show that $LaBr_3$-based Compton suppression systems would improve measured spectral results if oriented such that the gamma-ray photons are collimated to impinge the primary detector face as a beam. Photopeak losses as a result of accidental coincidences were minimal and the reduction in the Compton continuum allowed several additional photopeaks to be resolved. While HPGe-based Compton suppression systems resolve many more photopeaks, $LaBr_3$ is suitable for applications that require room-temperature operation and fewer opportunities to perform maintenance. Larger volume $LaBr_3$ detectors are available which would record higher unsuppressed peak-to-Compton ratios and may increase the number of resolved photopeaks.

Conclusions

The $LaBr_3$-based Compton suppression system demonstrated a significant improvement over unsuppressed $LaBr_3$ measurements through appreciably lower Compton continua of dominant photopeaks, such as ^{137}Cs and ^{140}La. While HPGe-based Compton suppression systems resolve many more photopeaks, $LaBr_3$ is a better candidate for applications where maintenance is difficult because it operates at room-temperature.

The enclosed geometry of a typical, low count rate Compton suppression system was shown to be ineffective for the measurement of spent nuclear fuel. While the Compton continuum from the ^{137}Cs 661.7 keV peak was largely suppressed, many large peaks have significant losses in the number of recorded counts due to the large solid angle, and therefore detection probability, of the measurement geometry. This leads to sum peaks in both the suppressed and unsuppressed spectra and losses to photopeak counts in the suppressed spectra.

Compton suppression would be a beneficial addition to NDA process monitoring systems if oriented so that the gamma-ray photons are collimated to impinge the primary detector face as a beam. The analysis has shown that peak losses through accidental coincidences are minimal and the reduction in the Compton continuum allows additional peaks to be resolved.

Acknowledgements

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Publications

S. Bender, B. Heidrich, K., Unlu, "Compton Suppressed $LaBr_3$ Detection System for use in Nondestructive Spent Fuel Assay" *Nucl. Instrum. Meth. A.* **784**,474-481 (2015).