

LA-UR-17-26601 (Accepted Manuscript)

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Provided by the author(s) and the Los Alamos National Laboratory (2017-12-08).

**To be published in:** Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment

**DOI to publisher's version:** 10.1016/j.nima.2017.09.054

**Permalink to record:** <http://permalink.lanl.gov/object/view?what=info:lanl-repo/lareport/LA-UR-17-26601>

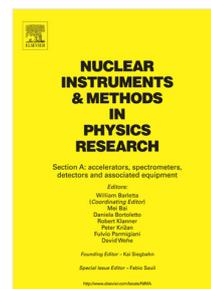
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PII: S0168-9002(17)31024-0  
DOI: <https://doi.org/10.1016/j.nima.2017.09.054>  
Reference: NIMA 60133

To appear in: *Nuclear Inst. and Methods in Physics Research, A*

Received date: 1 August 2017  
Revised date: 13 September 2017  
Accepted date: 21 September 2017

Please cite this article as: A. Favalli, M. Iliev, K. Ianakiev, A.W. Hunt, B. Ludewigt, Delayed gamma-ray spectroscopy with lanthanum bromide detector for non-destructive assay of nuclear material, *Nuclear Inst. and Methods in Physics Research, A* (2017), <https://doi.org/10.1016/j.nima.2017.09.054>

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**DELAYED GAMMA-RAY SPECTROSCOPY WITH LANTHANUM BROMIDE DETECTOR  
FOR NON-DESTRUCTIVE ASSAY OF NUCLEAR MATERIAL****Andrea Favalli<sup>1\*</sup>, Metodi Iliev<sup>1</sup>, Kiril Ianakiev<sup>1</sup>, Alan W.Hunt<sup>2</sup>, Bernhard Ludewigt<sup>3</sup>**<sup>1</sup>Los Alamos National Laboratory, Los Alamos, New Mexico, USA<sup>2</sup>Johns Hopkins University Applied Physics Laboratory, Laurel, Maryland 20723<sup>3</sup>Lawrence Berkeley National Laboratory, Berkeley, California, USA\*[afavalli@lanl.gov](mailto:afavalli@lanl.gov); phone +1 505 667 1429**ABSTRACT**

High-energy delayed  $\gamma$ -ray spectroscopy 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. Requirements for the  $\gamma$ -ray detection system, up to  $\sim 6$  MeV, can be summarized as follows: high efficiency at high  $\gamma$ -ray energies, high energy resolution, good linearity between  $\gamma$ -ray energy and output signal amplitude, ability to operate at very high count rates, and ease of use in industrial environments such as nuclear facilities. High Purity Germanium Detectors (HPGe) are the state of the art and provide excellent energy resolution but are limited in their count rate capability. Lanthanum Bromide ( $\text{LaBr}_3$ ) scintillation detectors offer significantly higher count rate capabilities at lower energy resolution. Thus,  $\text{LaBr}_3$  detectors may be an effective alternative for nuclear spent-fuel applications, where count-rate capability is a requirement. This paper documents the measured performance of a 2'' (length)  $\times$  2'' (diameter) of  $\text{LaBr}_3$  scintillation detector system, coupled to a negatively biased PMT and a tapered active high voltage divider, with count-rates up to  $\sim 3$  Mcps. An experimental methodology was developed that uses the average current from the PMT's anode and a dual source method to characterize the detector system at specific very high count rate values. Delayed  $\gamma$ -ray spectra were acquired with the  $\text{LaBr}_3$  detector system at the Idaho Accelerator Center, Idaho State University, where samples of  $\sim 3$ g of  $^{235}\text{U}$  were irradiated with moderated neutrons from a photo-neutron source. Results of the spectroscopy characterization and analysis of the delayed  $\gamma$ -ray spectra acquired indicate the possible use of  $\text{LaBr}_3$  scintillation detectors when high count rate capability may outweigh the lower energy resolution.

**KEYWORDS:** delayed gamma-ray; ultra-high count rate; gamma spectroscopy;  $\text{LaBr}_3$  detector; nondestructive assay of spent nuclear fuel.

## Introduction

High-energy, beta-delayed  $\gamma$ -ray spectroscopy is a potential, non-destructive assay technique for the independent verification of declared quantities of special nuclear materials at key stages of the fuel cycle and for directly assaying 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. Neutron induced fission generates a distribution of short-lived fission fragments with half-lives ranging from seconds to tens of minutes. Measured spectra contain unique actinide-specific signatures that can be exploited to nondestructively determine isotopic composition [1-6]. The energy range of the useful delayed  $\gamma$  rays extends up to  $\sim 6$  MeV, thus high-energy delayed  $\gamma$ -ray spectroscopy is needed. Requirements for the  $\gamma$ -ray detection system can be summarized as follows: high efficiency at high  $\gamma$ -ray energies, high energy resolution, good linearity between  $\gamma$ -ray energy and output signal amplitude, ability to operate in a very high count rate  $\gamma$ -ray field, and ease of use in industrial environments such as nuclear facilities. High Purity Germanium (HPGe) Detectors are the state of the art; however, an important limitation of HPGe detectors is the inability of a single-crystal detector to handle high count rates. Note that fully-burned spent-fuel assemblies produce over  $10^{15}$   $\gamma$ -rays per second [7], resulting in count rate capability being an important driving criterion among the requirements. Lanthanum Bromide ( $\text{LaBr}_3$ ) scintillation detectors offer significantly higher count rate capability due to the fast decay time (16 ns) of the emitted light and they do not need a cooling system and long cooldown periods but have a significantly reduced energy resolution in comparison with HPGe detectors [8-10]. Thus  $\text{LaBr}_3$  detectors may be an effective alternative when the high count rate capability outweighs the lower energy resolution.

## $\text{LaBr}_3$ -based-spectrometer characterization

### Experimental set up

In the first research phase, the performance of a 2'' (length) x 2'' (diameter)  $\text{LaBr}_3$  scintillation counter was evaluated at high  $\gamma$ -ray count rates as may be encountered in spent nuclear fuel applications. The measurement setup was optimized for high count rate studies. It included a negatively biased photomultiplier (PMT) (PM R6231-100 Hamamatsu with 8.5 ns rise time), to avoid additional long decay time constants and baseline shifts caused by a coupling capacitor. The PMT was coupled with a tapered

active high-voltage divider [11] in order to preserve good photoelectron collection (for good resolution) and gain linearity at high  $\gamma$ -ray energies and high count rates.

The PMT signal was amplified by a charge sensitive RC preamplifier. The decay time constant was chosen to be low, 40  $\mu$ s, and the feedback capacitor was chosen as 1.2 nF in order to avoid preamplifier saturation at high count rates. This time constant is about the minimum that most Multichannel Analyzers (MCAs) can compensate using Pole-Zero (P/Z) compensation. Additionally, in order to preserve the fast rise time of the LaBr<sub>3</sub> detector, a fast operational amplifier, an ADA4817 with a 1 GHz bandwidth and 870 V $\cdot\mu$ s<sup>-1</sup> slew rate, was selected for the preamplifier.

An MCA, called nanoMCA, produced by LabZY LLC [12] (firmware version April 2013, software ver.1.15) was used for the spectroscopic measurements. An important feature specific to this MCA is the availability of two compensation filters that allow the removal of some detector pulse tails caused by slow-rise time scintillator components [13]. The parameters for these filters were adjusted to minimize the duration of both fast and slow channel pulses since they directly affect pileup rejection and dead time performance. The nanoMCA has a slow shaper for spectroscopy and a fast shaper for pileup detection. Both shapers are trapezoidal. For the fast shaper, the rise time and flat top durations were set both to 12.5 ns, which is the minimum that the MCA could be set to. The shaping times for the slow shaper were set to rise time of 250 ns, and flat top of 12.5 ns. Other important parameters include detection thresholds (for slow and fast channel), which were set manually at about 50% above the automatically detected noise level at low rates. Automatic threshold setting was disabled to avoid unknown behavior in high count rate conditions. Spectra were acquired at 16k channels.

### **Spectroscopy performance at high $\gamma$ -ray input count rate.**

As  $\gamma$ -ray sources for testing, we used a combination of <sup>137</sup>Cs and <sup>232</sup>Th sources. The use of <sup>232</sup>Th provided a way to investigate  $\gamma$ -ray energies up to 2614 keV using a sealed radioactive source [8]. To achieve higher energies would require producing materials activated by nuclear reaction with an accelerator [9]. We used the dual source method to determine the dead time losses in the <sup>232</sup>Th peak: a set of <sup>137</sup>Cs sources to generate different count rates up to ~2.7 Mcps in the detector system, while observing the highest energy  $\gamma$ -line at 2614 keV from <sup>232</sup>Th. The <sup>232</sup>Th source had a fixed position relative to the detector, and therefore produced a constant count rate.

The spectroscopy parameters (FWHM, net peak area, peak position) were studied as function of the input count rate. We used the MCA's estimated input count rate as the reported value. At high count rates,

MCA's may introduce count-rate bias due to imperfect dead-time correction. In order to assess the MCA's performance in these conditions, we directly measured the current from the PMT's anode, which is proportional to the charge produced by the detector in a given time, independent of the pileups. Therefore, as long as the PMT remains linear and the average energy distribution of the incoming  $\gamma$ -rays is constant, the current value is proportional to the  $\gamma$ -ray count rate. In our setup a Keithley 6497 pico-ampere meter was used to measure the current from the PMT.

Figure 1 shows that the incoming count rate estimation of the MCA grows linearly with the current produced by the detector, as shown by the linear fit to data in the plot with  $R^2=0.9996$ . In this set of measurements the count rate due to  $^{232}\text{Th}$  is fixed at about 40 kcps, while the variation in count rate is due to the  $^{137}\text{Cs}$  source. It is worth highlighting that when the count rate is high, where we are assessing the effect of dead-time correction in the MCA, the count-rate contribution of  $^{232}\text{Th}$  is negligible, so the average energy distribution, that produced the detector current, is dominated by the  $^{137}\text{Cs}$   $\gamma$ -ray source. The linear behavior exhibited is an indication of good dead-time correction in the nanoMCA for the input count rates studied. Thus in the following part of the paper when we refer to count rate we refer to estimated input count rate from the nanoMCA.

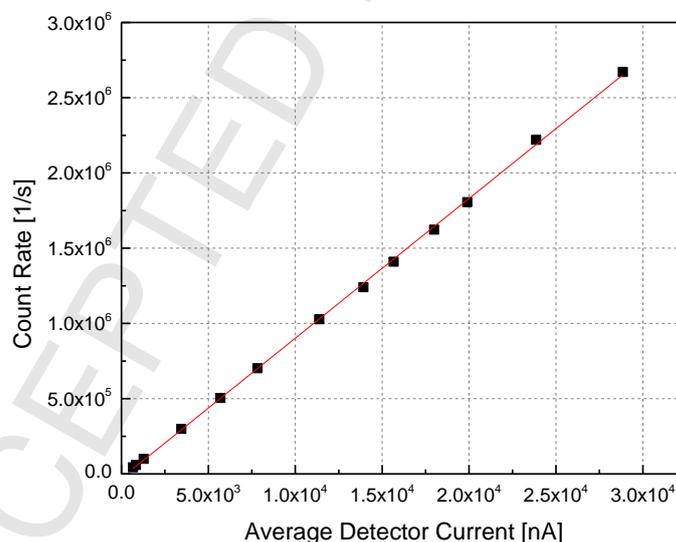


Fig.1. Measured anode current vs. estimated incoming count rate values as reported by the nanoMCA. The line is a linear fit to data with  $R^2=0.9996$ .

At each count rate (data points in Fig.1) spectra were collected with dual source method, and spectroscopy characteristics extracted. Spectra were acquired for MCA live times of 200 s.

Sample spectra from the two sources experiment are shown on Fig 2, for count rates of 100 kcps, 500 kcps, 1.4 Mcps, and 2.7 Mcps. Dead time values are measured from MCA. The effects of pile up at higher input count rates are visible, as well as a shift in the peak position.

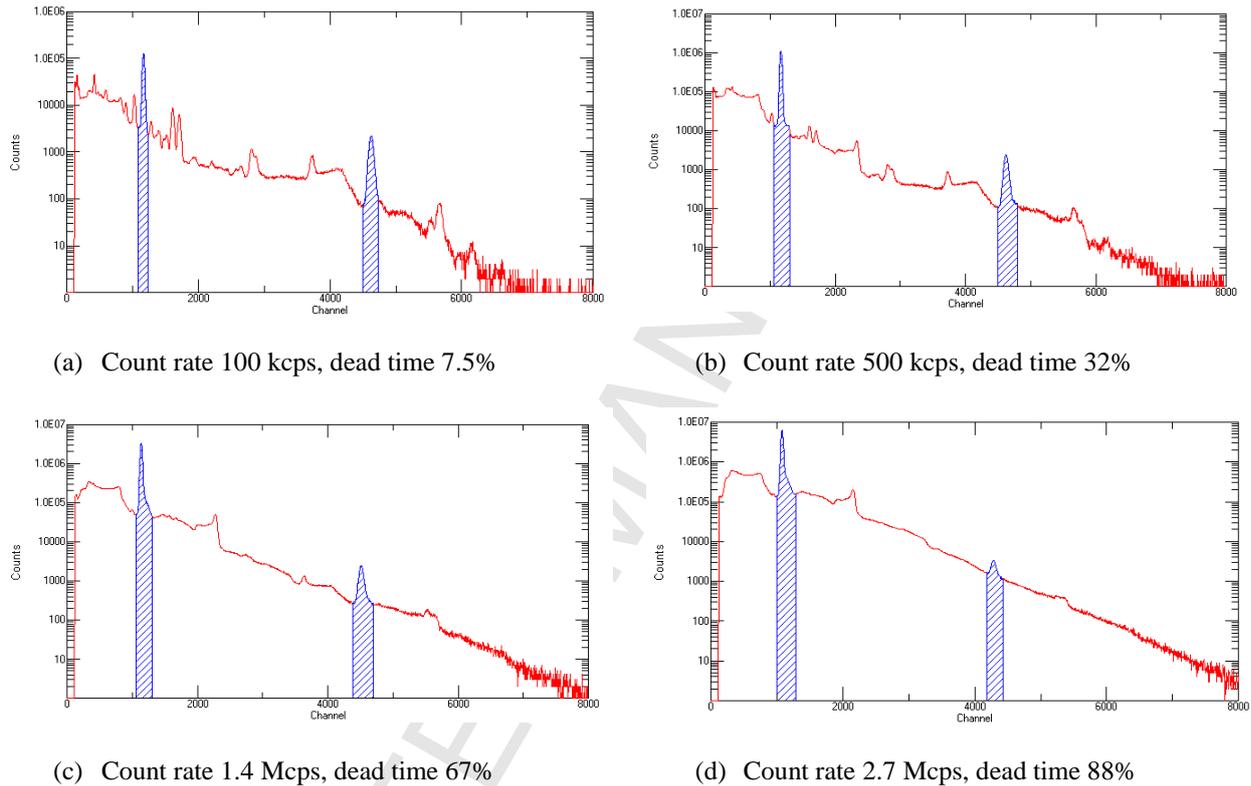


Fig.2.  $\gamma$ -ray spectra collected using  $^{137}\text{Cs}$  and  $^{232}\text{Th}$  sources at different count rates. From (a) to (d) count rate range from 100 kcps to 2.7 Mcps. ROIs for the 661.7 keV ( $^{137}\text{Cs}$ ) and 2614 keV ( $^{232}\text{Th}$ ) are indicated in each spectrum (colors in the online version).

For each spectrum, we extracted the variation in the net peak area of the 2614 keV  $\gamma$ -line from the  $^{232}\text{Th}$  source as a function of count rate. In addition, peak positions (in channels) and energy resolutions (measured as FWHM in %) were determined as a function of count rate of both the  $^{137}\text{Cs}$  661 keV  $\gamma$ -line and the 2614 keV  $\gamma$ -line of  $^{232}\text{Th}$ . The results are summarized in figures 3-5.

The unity normalized net area of the  $^{232}\text{Th}$   $\gamma$ -line as a function of the background count rate is shown in Fig. 3. Up to 2.2 Mcps the decrease in integrated peak counts is less than 5 %, at 2.7 Mcps the loss is less than 20%. Fig. 4 shows the shift in peak position for the  $^{137}\text{Cs}$  and the  $^{232}\text{Th}$   $\gamma$ -ray peaks as a function of count rate. At the highest count rate the shift to lower energy is about 8% for both peaks. Fig.5. shows that the energy resolution (reported FWHM in %) changes only slightly. Even at the highest rate, the

FHWM of the  $^{137}\text{Cs}$  line increased only from 2.9% to 3.1%, while the Th line was unchanged with a width of 1.5% FWHM. This is an indication of good pileup-rejector operation and well-functioning auxiliary MCA services, such as base line restoration, pulse shaping, and pulse tail compensation.

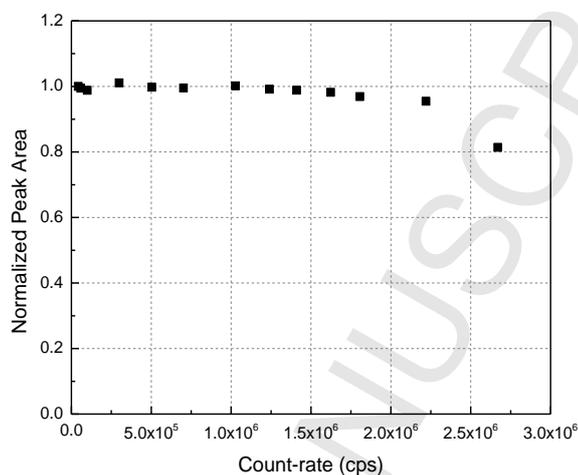


Fig.3.  $^{232}\text{Th}$  2614-keV  $\gamma$ -line net area (normalized) as a function of count rate.

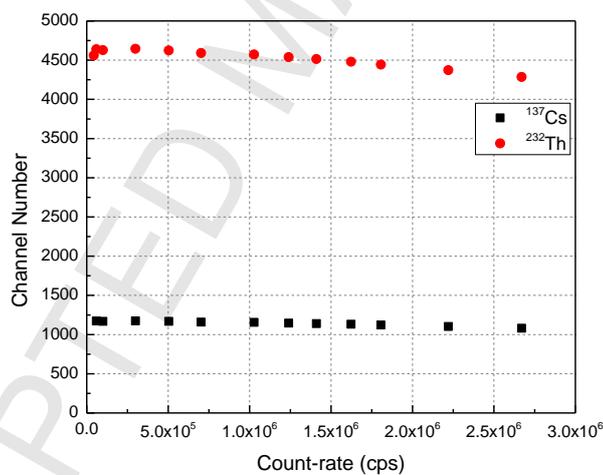


Fig.4. Peak positions, in channel number, of  $^{137}\text{Cs}$  and  $^{232}\text{Th}$   $\gamma$ -lines as function of count rate.

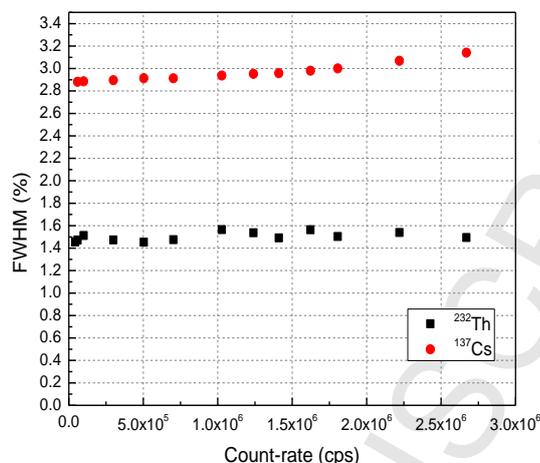


Fig.5. Variation of the energy resolution, measured as FWHM in %, of the  $^{137}\text{Cs}$  and  $^{232}\text{Th}$   $\gamma$ -lines with increasing of count rate.

It is worth mentioning that the setup used with  $^{137}\text{Cs}$  and  $^{232}\text{Th}$  for this experiment simulates the scenario in spent nuclear fuel (surrogate scenario) where the high passive  $\gamma$ -ray background is mainly due to  $^{137}\text{Cs}$ , and its effect on the delayed  $\gamma$ -ray peaks, at energies of several MeV, must be understood to effectively perform delayed  $\gamma$ -ray spectroscopy of nuclear material.

#### Delayed $\gamma$ -ray measurements on $^{235}\text{U}$ targets

Experimental measurements were conducted at the Idaho Accelerator Center (IAC), where a 2.8 g  $^{235}\text{U}$  sample was irradiated with neutrons from a photo-neutron source. At the IAC, a pulsed radiofrequency linac accelerator was used to produce 21 MeV electrons at 90 Hz pulse rate. Using Figure 6 as a guide, accelerated electrons produced a bremsstrahlung spectrum by impinging on a 2.2 mm tungsten radiator. Electrons that were not absorbed in the radiator struck a 5.08 cm thick aluminum beam stop immediately following the radiator. The high-energy photon beam traversed through a 15.2 cm thick Pb collimator with a 6.03 cm inner diameter that was inset in a large lead wall that shielded the fissile target from direct bremsstrahlung photons to prevent photofission. The bremsstrahlung beam then impinged on a 25.4 cm thick and 8.9 cm diameter beryllium cylinder located 58.8 cm from the bremsstrahlung radiator. This Be neutron converter was surrounded by a large amount of neutron moderating polyethylene, measuring  $\sim 57.8$  cm wide by  $\sim 40.6$  cm deep and  $\sim 60$  cm tall. The  $^{235}\text{U}$  target was located in a small cavity inside this moderator  $\sim 17.5$  cm from the Be converter, resulting in its irradiation by a moderated neutron spectrum.

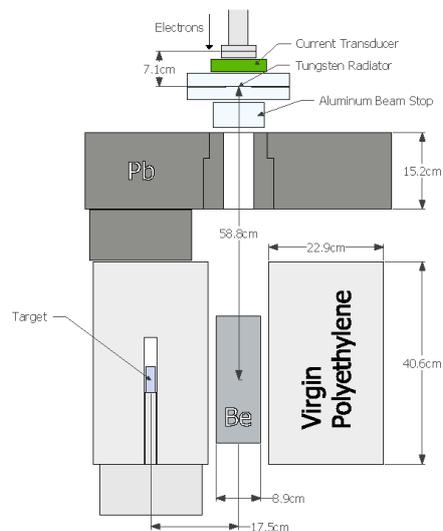


Fig.5. Schematic diagram of the irradiation set up.

The  $\text{LaBr}_3$  scintillation was located in a separate well-shielded experimental room, and the sample, after irradiation, was manually transferred to the detector. The detector was surrounded by 3.2 mm of Pb to filter out low energy  $\gamma$ -rays. Delayed  $\gamma$ -ray spectra were acquired for energies up to 6 MeV. For the delayed  $\gamma$ -ray measurements employing the  $\text{LaBr}_3$  detector consist of (15/15 pattern) 15 minutes of neutron irradiation, followed by 15 minutes of  $\gamma$ -ray spectroscopic measurements. Transfer time for the nuclear material samples from the irradiation location to the measurement station was  $\sim 20$  s. Delayed  $\gamma$ -ray spectra were acquired by repeating irradiation and spectrum collection 4 times and summing the spectra, which were corrected for any gain drifts. At the beginning of the 15 minutes measurement period count rate and dead time per each spectrum were on the order of  $\sim 1$  Mcps and  $\sim 50\%$ , respectively. Fig. 7 shows the  $^{235}\text{U}$  delayed  $\gamma$ -ray spectrum in the energy range of 2 MeV to 6 MeV: the energy resolution of the  $\text{LaBr}_3$  detector is sufficient to obtain a spectrum of useful  $\gamma$ -lines over the entire range of measurement. The  $\gamma$ -ray lines were identified using the evaluation library of T. R. England and D. F. Rider [14], now in ENDF/B-VII.1 [15]. The  $\gamma$ -line of the  $^{95}\text{Y}$  fission product is well defined in the center of the spectrum, the  $\gamma$ -lines that arise from the  $^{90}\text{Rb}$  fission fragment, which is an indicator of  $^{235}\text{U}$  fission, can be identified. With increasing energy, the density of the  $\gamma$ -lines decreases substantially, and the spectrum is dominated by the  $\gamma$ -lines of the  $^{90}\text{Rb}$  fission fragment. A decreased density in  $\gamma$ -lines translates also decreased necessity regarding the energy resolution of the detector. In the inset of Fig.7, the high energy region of the  $^{235}\text{U}$  spectrum from 5 to 6 MeV is shown: two  $\gamma$ -lines from  $^{90}\text{Rb}$  and two from  $^{86}\text{Br}$  are observed. While these highest-energy, discrete  $\gamma$ -lines have low yield [14,15], they are in a region with low background, and clear of interferences. It is worth noting here that the 15/15 pattern, i.e. 15 minutes of neutron irradiation and 15 minutes of  $\gamma$ -ray spectroscopic measurements, is not optimized

for a  $\text{LaBr}_3$  detector system, as the detector features a very short light decay constant that enables the detector to work with short irradiation/measurement cycles.

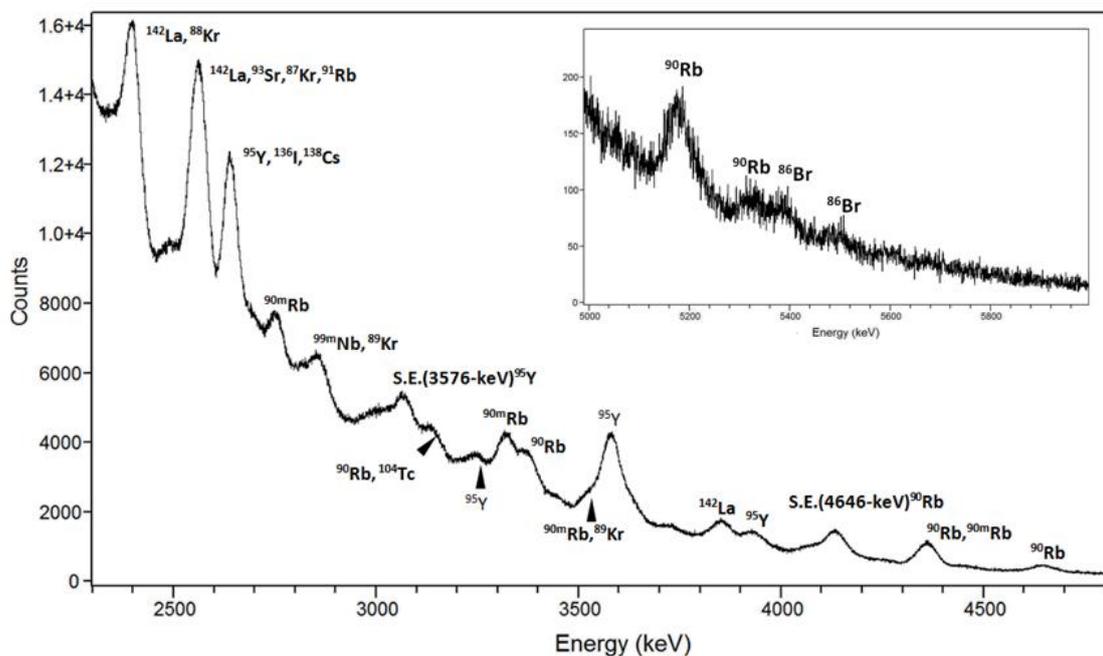


Fig.7. Delayed  $\gamma$ -ray spectrum of  $^{235}\text{U}$  acquired with the (15/15) pattern. Inset spectrum presents range from 5 MeV to 6 MeV. Spectrum acquisition time: Real Time 3600 s, Live Time 2766 s. The labels identify the fission fragment mainly responsible for the  $\gamma$ -ray peak, and S.E. indicates the single escape peak.

## Conclusion

Results of the characterization of a  $\text{LaBr}_3$   $2'' \times 2''$  scintillation detector coupled to (1) a negatively biased PMT PM R6231-100 Hamamatsu, (2) a tapered active high voltage divider, and (3) the nanoMCA displayed excellent performance at high count rates up to 2.7 Mcps, for delayed  $\gamma$ -ray spectroscopy applications. Delayed  $\gamma$ -ray spectra from neutron interrogation of  $^{235}\text{U}$  targets were acquired, and the analysis identified significant  $\gamma$ -ray lines from the  $^{235}\text{U}$  fission products. The spectroscopy characterization at high count rates and the analysis of delayed  $\gamma$ -ray spectra of  $^{235}\text{U}$  indicate that a  $\text{LaBr}_3$  detector system could be a useful alternative to an HPGGe detector for delayed  $\gamma$ -ray spectroscopy in applications, where the high count rate capability outweighs the lower-energy resolution.

## Acknowledgments

This work was sponsored by the U.S. Department of Energy (DOE), National Nuclear Security Administration (NNSA), Office of Nonproliferation Research and Development (NA-22).

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