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Delayed gamma-ray spectroscopy with lanthanum bromide detector for non-destructive assay of nuclear material

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ABSTRACT

High-energy delayed γ-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 γ-ray detection system, up to ~6 MeV, can be summarized as follows: high efficiency at high γ-ray energies, high energy resolution, good linearity between γ-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 ($LaBr_3$) scintillation detectors offer significantly higher count rate capabilities at lower energy resolution. Thus, $LaBr_3$ 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) × 2" (diameter) of $LaBr_3$ scintillation detector system, coupled to a negatively biased PMT and a tapered active high voltage divider, with count-rates up to ~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 γ-ray spectra were acquired with the $LaBr_3$ detector system at the Idaho Accelerator Center, Idaho State University, where samples of ~3g of ^{235}U were irradiated with moderated neutrons from a photo-neutron source. Results of the spectroscopy characterization and analysis of the delayed γ-ray spectra acquired indicate the possible use of $LaBr_3$ scintillation detectors when high count rate capability may outweigh the lower energy resolution.

1. Introduction

High-energy, beta-delayed γ-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 γ rays extends up to ~6 MeV, thus high-energy delayed γ-ray spectroscopy is needed. Requirements for the γ-ray detection system can be summarized as follows: high efficiency at high γ-ray energies, high energy resolution, good linearity between γ-ray energy and output signal amplitude, ability to operate in a very high count rate γ-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} γ-rays per second [7], resulting in count rate capability being an important driving criterion among the requirements. Lanthanum Bromide ($LaBr_3$) 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

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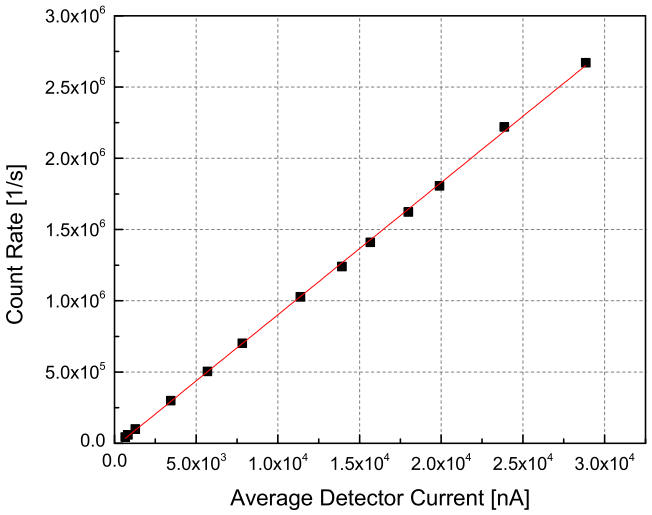


Fig. 1. Measured anode current vs. estimated incoming count rate values as reported by the nanoMCA. The line is a linear fit to data with $R^2 = 0.9996$.

a cooling system and long cooldown periods but have a significantly reduced energy resolution in comparison with HPGe detectors [8–10]. Thus $LaBr_3$ detectors may be an effective alternative when the high count rate capability outweighs the lower energy resolution.

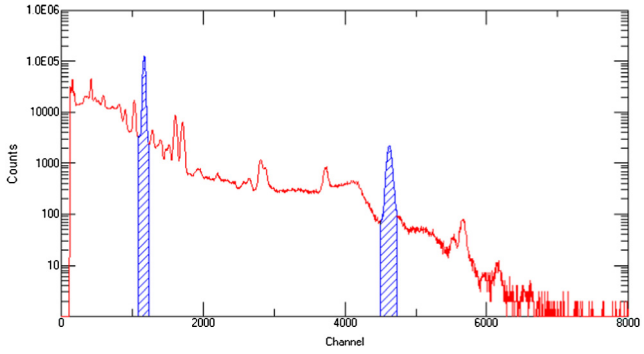
2. $LaBr_3$-based-spectrometer characterization

2.1. Experimental set up

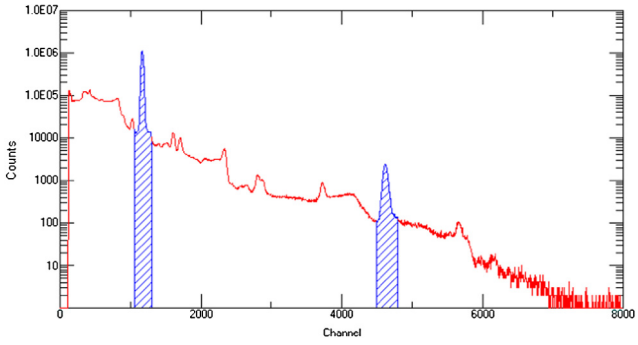
In the first research phase, the performance of a 2″ (length) × 2″ (diameter) $LaBr_3$ scintillation counter was evaluated at high γ-ray count rates as may be encountered in spent nuclear fuel applications. The measurement setup was optimized for high count rate studies. It included a negatively biased 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 γ-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 μs, 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 $LaBr_3$ detector, a fast operational amplifier, an ADA4817 with a 1 GHz bandwidth and 870 $V \cdot \mu s^{-1}$ slew rate, was selected for the preamplifier.

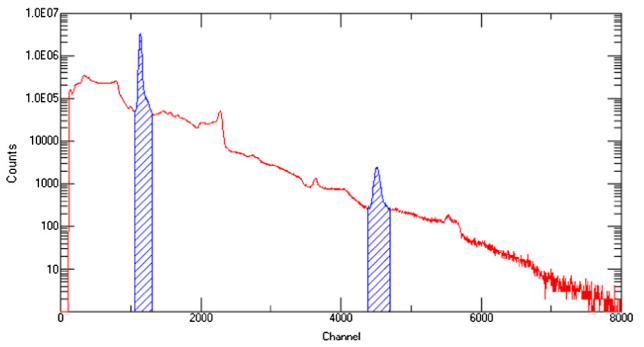
An MCA, called nanoMCA, produced by LabZY LLC [12] (firmware version April 2013, software ver.1.15) was used for the spectroscopic measurements. An important feature specific to this MCA is the availability of two compensation filters that allow the removal of some detector pulse tails caused by slow-rise 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.



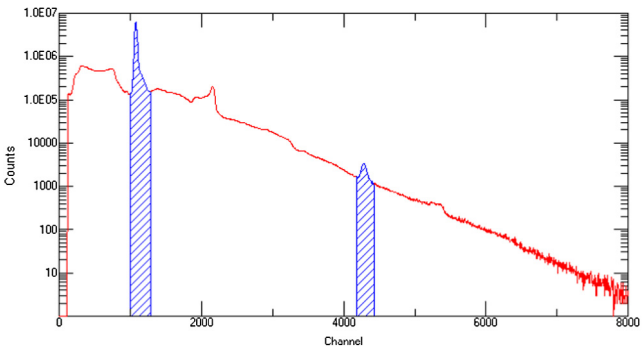
(a) Count rate 100 kcps, dead time 7.5%.



(b) Count rate 500 kcps, dead time 32%.



(c) Count rate 1.4 Mcps, dead time 67%.



(d) Count rate 2.7 Mcps, dead time 88%.

Fig. 2. γ-ray spectra collected using ^{137}Cs and ^{232}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}Cs) and 2614 keV (^{232}Th) are indicated in each spectrum (colors in the online version).