

THE USE OF LANTHANUM BROMIDE DETECTORS FOR NUCLEAR SAFEGUARDS APPLICATIONS

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ABSTRACT

Lanthanum Bromide (LaBr₃) is a type of scintillation detector that has become commercially available recently. According to the manufacturer and studies conducted at a laboratory level, this new gamma-ray detector presents improved characteristics in comparison with traditional sodium iodine (NaI) detectors. Better energy resolution, relative efficiency and stability with temperature are some of the features that may impact positively the performance of the measurements made with this detector, which works at room temperature. Uranium enrichment measurements performed in the field is of special interest at nuclear fuel cycle facilities. Typical applications include quality control, U-235 inventory verification and nuclear safeguards. The Safeguards Laboratory of the Brazilian Nuclear Energy Commission (CNEN) and the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) have initiated the evaluation of the performance of a LaBr₃ probe model BrillLanceCe 380 (B380) for U-235 enrichment determination in uranium compounds typically found in nuclear facilities under safeguards. This activity is routinely performed by national and international safeguards inspectors in both Brazilian and Argentine facilities. Depending on the characteristics of the material to be verified and the conditions at the measurement location, NaI or high-purity germanium detector (HPGe) are usually selected as the standard gamma-ray spectroscopic system for enrichment determination. This paper presents the conclusions of the initial studies jointly conducted by CNEN and ABACC regarding the use of a LaBr₃ detector for enrichment determination, based on the comparison with NaI and HPGe typical performances. It also discusses the possibility to use this new gamma-ray detector as a standard technique for safeguards applications, compliant with national and international performance values.

1. INTRODUCTION

Verification of the ²³⁵U enrichment in items (cylinders, drums, pellets, rod etc) is routinely performed in nuclear facilities using portable instrumentation, for material control and accountability, as well as quality control purposes. Standard methods for this verification are based on non-destructive (NDA) gamma-ray spectroscopy using high-purity germanium (HPGe) or sodium iodine detectors (NaI) [1]. If measurement conditions are not favorable, i.e. high background levels, limited measurement time, low enrichment, then HPGe is usually selected.

Recently, Lanthanum Bromide (LaBr₃) detectors have become commercially available. Based on the technical specifications reported by vendors, there is an expectation that this type of detector will bring significant performance improvements into NDA based methods routinely used for ²³⁵U enrichment determination. It is also expected that new gamma-ray analysis algorithms will be developed in order to properly analyze gamma-ray spectra obtained with this new type of detector. Some studies [2] have been conducted in order to assess the performance of such a detector for ²³⁵U enrichment determination, most of them under laboratory conditions. The goal of this study is to perform the evaluation of a LaBr₃ detector in the field. Enrichment measurements of UF₆ cylinders have been conducted, with data analysis using currently available software tools. As a reference for the evaluation, the observed performance has been compared to the values obtained using a standard NaI based system.

2. EXPERIMENTAL EQUIPMENT AND SETUP

Two different NDA systems were used for simultaneous spectra acquisition from the same batch of UF₆ cylinders enriched at 4% U₂₃₅. For comparison purposes, a system based on NaI (2"x2" Canberra model 802-2x2) detector was used as reference. A LaBr₃ (1.5"x1.5" Saint Gobain model Brilliance 380) based system was used for testing and evaluation purposes.

Two versions of the NaIGEM code (1.52a and 2.1) were used for data analysis. This code was developed to be a software tool dedicated to determine U₂₃₅ enrichment in infinitely thick [1] samples using NaI detectors (version 1.52a). Version 2.1 has been recently developed as an option capable of analyzing LaBr₃ spectra. Therefore, all LaBr₃ spectra were analyzed using this version. In addition, version 2.1 has the ability to account for significant differences between the wall thickness of the calibration standard and sample. An independent thickness calibration routine must be run for this purpose. Therefore, two calibration methods were tested: using only a calibration standard (NBS-969); and adding an additional absorber (steel, 12 mm thick) so that geometry and attenuation conditions could be very close to those associated with a 30B cylinder.

Collimator dimensions (circular 1"x1"), multichannel analyzer instrument (GBS model MMCA-166) and calibration conditions were the same.

Figure 1 shows low energy spectra from photons emitted by uranium decay in the calibration standard (4,46% U₂₃₅). The total efficiency for both NaI and LaBr₃ systems were similar in this study. It can be also observed by that energy calibration for LaBr₃ is more linear than NaI, as expected.

3. RESULTS

Eight UF₆ 30B cylinders were measured in the same area where they are weighted. The background level in this area is significantly smaller than at the external yard. Figure 2 shows typical NaI and LaBr₃ spectra for a cylinder measurement conducted in the weighing area (600 sec counting time). Higher Compton background for the NaI system can be observed.

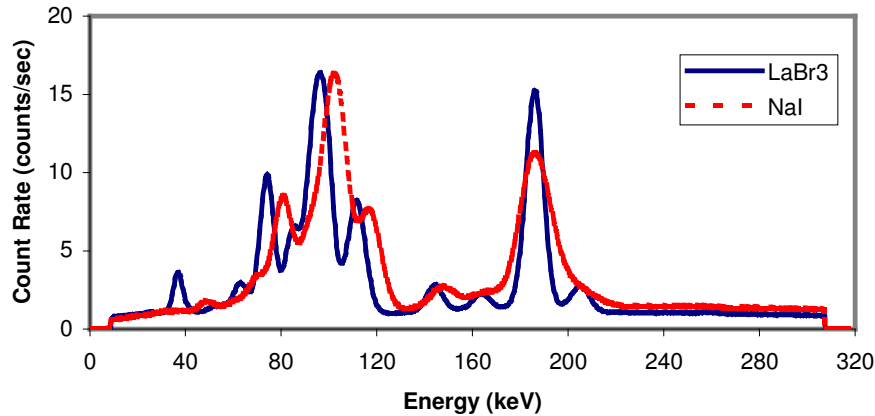


Figure 1: Uranium Spectra of LaBr3 and NaI Detectors for the Calibration Standard

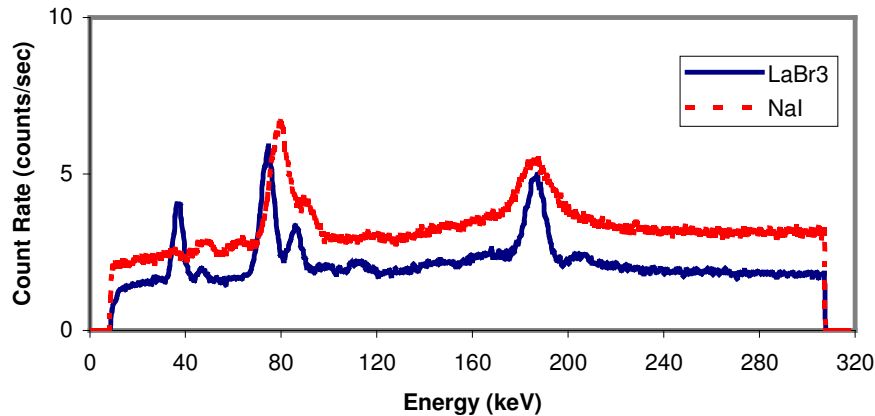


Figure 2: UF6 Spectra of LaBr3 and NaI Detectors for a 30B Cylinder in the Weighing Area

Three measurements at different positions on the cylinder surface were performed, with counting time always set at 600 seconds. The wall thickness for each measurement position was determined by using an ultrasonic thickness gage (model Panametrics 26MG) adjusted for 0.1 mm reading resolution. Both NaI and LaBr3 detectors were placed on the same points for spectra acquisition.

For the evaluation of the calibration uncertainty, a set of 20 repeated calibration measurements were collected with both systems under two different attenuation conditions: with and without an additional steel absorber disk. The calculated relative standard deviation for the enrichment measurement of a reference sample when the absorber was added was 0.34% for the NaI and 0,48% for the LaBr3 based system. In the case that no absorber was used, 0.14% and 0.12%rsd values were obtained, respectively.

In the field, a set of 16 repeated enrichment measurements of the same item were simultaneously collected using both systems so that repeatability could be evaluated and

compared. For the NaI based system, the calculated relative standard deviation was 1.44%, while for the LaBr₃ based system this value was 1.86%. These values are within the 2% limit usually observed during measurement campaigns using an NaI based system.

Figures 3 and 4 show the measured error for each measurement (every cylinders was measured three times, at different positions on its external surface). NaI data is shown in Fig. 3, while LaBr₃ data is in Fig. 4. NaI data was evaluated under four different conditions, depending on the version of the NaIGEM code and the use or not of the additional calibration absorber. Meanwhile, LaBr₃ data was evaluated under two different conditions, depending on the use or not of the additional calibration absorber.

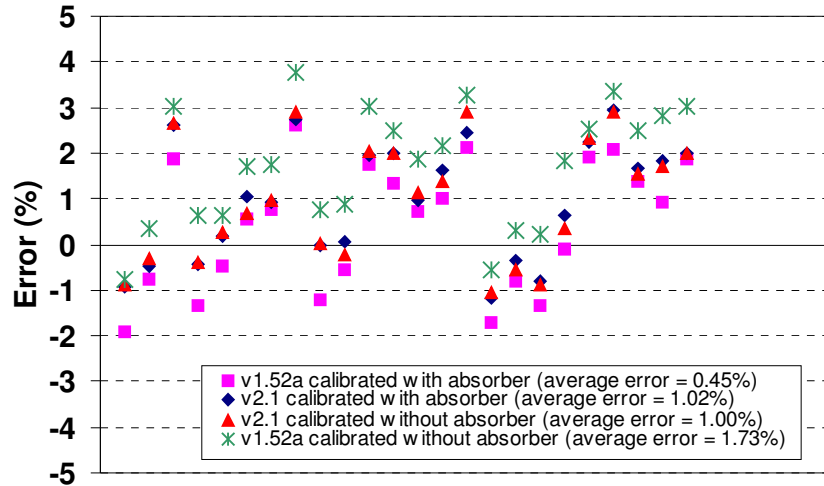


Figure 3: Relative Errors for the NaI Based System

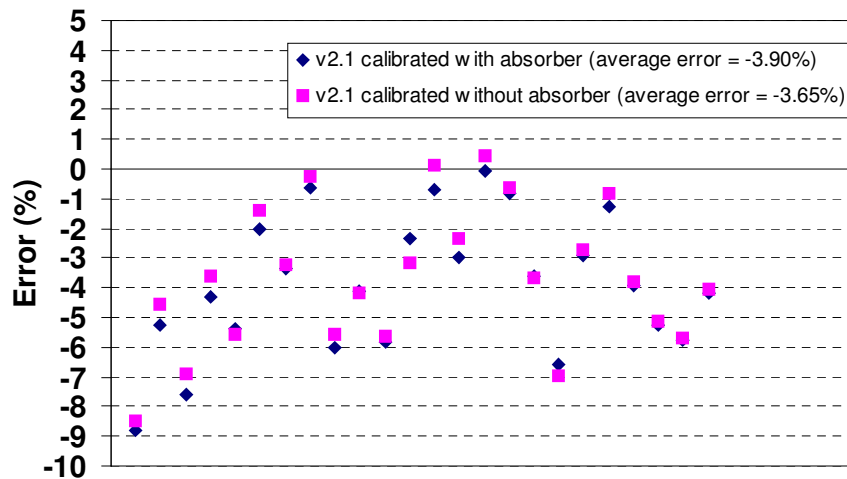


Figure 4: Relative Errors for the LaBr₃ Based System

4. CONCLUSIONS

Two factors were considered in this study: the LaBr₃ detector performance and the NaIGEM capability to properly make corrections and calculate the enrichment of a sample. Previous studies conducted at the laboratory environment have provided clear evidence that the LaBr₃ detector under evaluation may contribute to improving the quality of enrichment measurements. In this study, we also observed better temperature stability and repeatability in results obtained with LaBr when thinner containers were measured, which cause lower gamma-ray attenuation in the 186 keV region.

Regarding NaIGEM version 2.1, the capability to compute significant differences between the attenuation features of the standard for calibration and samples was observed to be effective. Version 1.52a does not have this capability and then significant biases are usually observed for measurement of samples that have thicker containers in comparison to calibration standards.

Some bias was observed for both NaI and LaBr₃ enrichment results using version 2.1. The authors are investigating possible causes for this apparent problem.

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