Collimated LaBr3 detector response function in radioactivity analysis of nuclear waste drums (Postprint)

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Abstract

The properties of a Lanthanum bromide (LaBr3) detector and its response functions were investigated via experiments and simulations in this paper. The LaBr3 detector had good relative energy resolution and higher efficiency than a high-purity germanium detector. Monte Carlo and other numerical methods were used to calculate the efficiencies of a LaBr3 detector with a square collimation window. A model of the numerical method was established based on a pure geometric model that was consistent with the experimental situation. The results showed that the detector response functions calculated by these methods were in great agreement with experimental results.

Full Text

Preamble

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Collimated $LaBr_3$ Detector Response Function in Radioactivity Analysis of Nuclear Waste Drums

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Abstract

This paper investigates the properties of a lanthanum bromide ($LaBr_3$) detector and its response functions through experiments and simulations. The $LaBr_3$ detector exhibited good relative energy resolution and higher efficiency compared to a high-purity germanium detector. Monte Carlo and other numerical methods were employed to calculate the efficiencies of a $LaBr_3$ detector with a square collimation window. A numerical method model was established based on a pure geometric model consistent with the experimental configuration. The results demonstrated that the detector response functions calculated by these methods showed excellent agreement with experimental results.

Key words: Nuclear waste, $LaBr_3$ detector, Detector response, Gamma scanning, Energy resolution

Introduction

The radionuclide activity in radioactive waste drums must be characterized prior to handling and disposal. Segmented gamma scanning (SGS) and tomographic gamma scanning (TGS) are two widely used non-destructive characterization methods. While the accuracy of the SGS method is lower than that of TGS when the waste drum contains heterogeneous radioactive materials, SGS requires significantly less measurement time compared to TGS [?]. At present, long measurement time represents the primary constraint on applying TGS in nuclear power plants. Two approaches have been employed to reduce measurement time in TGS: establishing new reconstruction algorithms and utilizing detectors with higher efficiency. Liu et al. used dynamic grids in TGS to reconstruct source distribution [?], reducing measurement time by half while maintaining accuracy. To detect prompt gamma rays from neutron capture, lanthanum bromide scintillators ($LaBr_3$) have been applied to PGNAA [?]. In the present study, a $LaBr_3$ detector was applied to TGS. Its efficiency and energy resolution are described in Section 2, while Section 3 discusses the efficiencies determined by Monte Carlo and deterministic calculation methods. Conclusions are presented in Section 4.

2 Properties of $LaBr_3$ Crystal and Its Detector

2.1 Properties of $LaBr_3$ Crystal

Lanthanum bromide scintillators ($LaBr_3$) with energy resolution of approximately 3% at 662 keV provide substantial improvement over sodium iodide scintillators in resolution [?]. Unlike HPGe detectors, which require low-temperature operating conditions, $LaBr_3$ detectors can be operated at room temperature. However, $LaBr_3$ has several disadvantages, including internal radioactivity and low-energy response. The internal radioactivity arises from naturally occurring radioisotopes ^{138}La and decay products of ^{227}Ac [?]. Here, we discuss the following properties of a $LaBr_3$ detector: energy linearity, relative energy resolution, and detection efficiency as a function of γ-ray energy.

2.2 Properties of LaBr$_3$ Detector

The LaBr$_3$ crystal was manufactured by Saint-Gobain. The following set of sources was used in the experiment: ^{133}Ba, ^{152}Eu, ^{137}Cs, ^{60}Co, ^{226}Ra, and ^{241}Am. These sources were positioned 25 cm from the detector center, with a measurement time of 3600 s. These sources provided homogeneous and wide-range γ-ray energies for analyzing the performance of LaBr$_3$ detectors and distinguishing between different radionuclides. The experiments primarily aimed to analyze absolute efficiency, energy linearity, and full width at half maximum of the LaBr$_3$ detector. [Figure 1: see original paper] shows a typical spectrum of ^{137}Cs (661.7 keV) and ^{60}Co (1173.21 keV and 1332.47 keV) measured by the LaBr$_3$ detector. Both radioactive sources could be identified; however, the spectrum was affected by the detector' s intrinsic activity (1430 keV from ^{138}La and 1465 keV from ^{40}K), and the 789 keV peak from ^{138}Ce was identified from β-decay.

[Figure 2: see original paper] demonstrates good energy linearity from ^{241}Am (59.54 keV) to ^{152}Eu (1408 keV), covering the most important radioactive sources found in nuclear waste drums. [Figure 3: see original paper] shows the relative energy resolution from ^{241}Am (59.54 keV) to ^{60}Co (1332 keV), fitted by the function:

The relative energy resolution improves from 13% at 59.54 keV to 2% at 1332 keV, which represents a satisfactory value for scintillation detectors. [Figure 4: see original paper] compares the full absorption efficiency of LaBr$_3$ and HPGe detectors from 121 to 1332 keV. While ^{60}Co provided high γ-ray energies, ^{133}Ba and ^{137}Cs supplied low and medium energies, respectively. The measurement time was 3600 s for both detectors, with each peak reaching 10,000 counts, resulting in statistical errors of less than 1% for each peak. The full absorption efficiency was fitted by Eq.(2).

The ratio of LaBr$_3$ detector efficiency to HPGe detector efficiency ranged from 1.3 to 2 as energy increased from 121 to 700 keV, and remained at 2 for energies exceeding 700 keV. Consequently, the measurement time for the LaBr$_3$ detector was half that required for the HPGe detector.

3 Experimental and Simulation Studies of LaBr$_3$ Detector Response

3.1 Experimental Description

In the experiment, the detector was collimated by a square window. The source was distributed on a 9-cm radius circle (r) in front of the detector. The distance (d$_0$) between the circle center and the detector was 52 cm. [Figure 5: see original paper] schematically illustrates the experimental setup of the collimated detector system. The crystal has a radius (r_d) of 1.96 cm and length of 3.91 cm. The collimator length (l_{col}) is 12 cm, with a square window (a_{col}) width of 4.8 cm. The drum was rotated in steps. The source was located at radial positions of 9 and 20 cm, shifted 3.5 cm (l) from the drum center. Photon count rates were measured at each 15° rotation step.

3.2 MCNP Simulation of $LaBr_3$ Detector Response

This section presents simulation results for the $LaBr_3$ detector response using the Monte Carlo N-particle (MCNP) method. The efficiencies of sources at different positions are fundamental parameters in gamma scanning. It is difficult to determine these efficiencies experimentally because energies from experimental sources cannot cover all energies present in waste drums. In contrast, simulation models can easily establish energies and positions of point sources as well as void sources, allowing experiments to be partially replaced by simulations.

[Figure 6: see original paper] compares simulated values with experimental results, showing good agreement. The average error is less than 3%, indicating that the simulation model reproduces experimental results very well.

3.3 Numerical Method Simulation of $LaBr_3$ Detector Response

Although MCNP simulation can reproduce experimental results, it requires considerable computational time to calculate detector efficiency. In contrast, the numerical method significantly reduces calculation time. For example, calculating the efficiency at one point takes approximately 9 minutes with MCNP, while the numerical method calculates efficiencies at 90 points in only 5 minutes using the same computer. Thus, the numerical method may serve as a replacement for MCNP.

A calculation model for a point source is shown in [Figure 5: see original paper]. The detector efficiency can be calculated based on the solid angle subtended by the collimated detector to the point source. The geometry presented in [Figure 5: see original paper] follows Krings and Mauerhofer (2011) [**?**] and Nan et al. (2012) [**?**]. The absolute detector efficiency (d_n) to a point source can be calculated by:

where (d_n) is the absolute detector efficiency for a point source located at distance d_n from the detector; (d_0) is the absolute detector efficiency for a point source located at the drum center; S_{d_n} is the portion of the active detector surface illuminated by the photon beam due to collimation; S_{det} is the area of the active detector surface; C is the correction factor for edge penetration effects; and m is the power of (d_0/d_x).

Efficiencies were calculated based on the source efficiency at Point A. First, the efficiency at point A was transformed to Point B located at d_n ([Figure 5: see original paper]). This transformation primarily depended on the distance between the point source and the detector surface. Then, the efficiency at

Point B was transformed to the target location (Point C). This transformation mainly depended on the active detector surface illuminated by the photon beam, S_{d_n}. The factor C corrected the absolute efficiency for edge penetration effects. Unscattered photon beams passing through the collimator and reaching the active detector volume were disregarded due to their negligible effects on efficiency transformation. When the source was a volume source, it was divided into numerous point sources. The efficiency of this volume source represents the average efficiencies of the point sources, calculated by:

where k is the number of point sources in the volume source.

Illuminated Active Detector Surface In this section, r_d represents the radius of the active detector surface. [Figure 7: see original paper] presents six cases for schematic calculation of S_{d_n}, following Nan et al. (2012) [?]. Before calculating S_{d_n} for these cases, the arc segment areas (S) must be determined. The area of an arc segment according to the notation in [Figure 5: see original paper] is given by:

where g is the chord length defined by the intersection of the active detector surface and one side of the collimated photon beam. The central angle of the sector is expressed as 2 arcsin(g/2r_d). The side length of the photon beam at the plane defined by the active detector surface is given by:

where d_x is the projection of d_n on the X-axis.

Based on Eqs. (5)-(7) and [Figure 7: see original paper], S_{d_n} for these cases can be calculated by:

Case 1:
Case 2:
Case 3:
Case 4:
Case 5:
Case 6:

Edge Penetration Correction [Figure 8: see original paper] schematically illustrates different photon path lengths in a detector crystal from a point source. The correction factor was calculated as the ratio of the photon beam absorption probability to the average absorption probability of a photon beam from a point source facing the detector. The average absorption probability calculation relied on a mesh of n_0 virtual points distributed homogeneously across the active detector surface. Penetration lengths l_{i_r}s of photons from a point source were calculated for each photon beam entering the active detector surface at one of the n points within the illuminated portion of the active detector surface. The correction factor can be expressed as:

where n is the number of virtual points distributed across the active detector surface under different conditions; is the linear attenuation coefficient of Ge; is the density of Ge; and n_0 is the maximum number of virtual points distributed across the active detector surface.

Reliability Analysis of Numerical Method The count rate distribution of ^{137}Cs point sources was simulated under the same experimental conditions. Experimental and calculated count rate results are shown in [Figure 9: see original paper]. The calculated results agree well with experimental results for all simulated source positions, with an average error of less than 3%. This verifies the validity of the numerical method for simulating $LaBr_3$ detector response.

4 Conclusion

This study demonstrated that the $LaBr_3$ detector can be applied to SGS and TGS techniques. The error between experimental results and MCNP simulation results for detector efficiencies was less than 3%. The numerical method used to calculate the $LaBr_3$ detector response for gamma scanning system geometry agreed perfectly with experimental data. Therefore, the numerical method for $LaBr_3$ detector response can be effectively employed in SGS and TGS techniques.

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