MDA Assessment of Liquid Radioactive Waste for On-Site Application using $CeBr_3$, $LaBr_3(Ce)$, and NaI(Tl) Scintillators

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1. Introduction

In situ gamma spectroscopy enables rapid decision-making by providing real-time results on-site without the need for sample extraction. This technique allows the measurement of radionuclide concentrations and isotopic compositions without altering the sample, minimizing both environmental and human-induced errors. The portability of the equipment facilitates analysis at various locations while significantly reducing transportation and laboratory preparation costs. [1, 2]

However, detectors commonly used for field analysis, such as NaI(Tl), have poorer energy resolution compared to High Purity Germanium (HPGe) detectors, making isotope identification more challenging. To address these limitations, cerium-doped lanthanum bromide ($LaBr_3(Ce)$) and cerium bromide ($CeBr_3$) scintillators have been introduced. NaI(Tl) not only has inferior energy resolution but also exhibits higher temperature dependency than $LaBr_3(Ce)$ and $CeBr_3$.

Among these alternatives, $LaBr_3(Ce)$ offers superior energy resolution and lower temperature dependency than both NaI(Tl) and $CeBr_3$. However, its use is limited by relatively high intrinsic background radiation due to natural radionuclides within the scintillator material. In contrast, $CeBr_3$ provides better energy resolution than NaI(Tl), though slightly lower than $LaBr_3(Ce)$, while maintaining a lower intrinsic background than $LaBr_3(Ce)$ [3].

The detection capabilities of scintillation detectors vary depending on the properties of the scintillator material and the energy of the incident gamma rays, as summarized in the table below. Therefore, to ensure rapid and accurate field measurements, it is crucial to evaluate the minimum detectable activity (MDA) in advance.

2. Methods and Results

2.1 Experimental setup

In this study, we aims to compare the MDA of NaI(Tl), $LaBr_3(Ce)$, and $CeBr_3$ scintillators under identical condition. To ensure a comparison, all detectors ware size of 1”Ø x 1”.

The samples were obtained from liquid waste containing ^{137}Cs and ^{60}Co, which were generated during the decommissioning or operation of a research reactor at KAERI. Three different samples with varying concentrations were selected for measurement. Since these liquid radioactive samples are not standard reference materials, they were measured for 3600 seconds using identical size of Marinelli beakers. The analysis results are shown in Table 1. The analysis program Genie 2000 (Canberra Co., Ltd, USA) employs the Currie MDA method for each radionuclide. The gamma-ray spectrum for each scintillator were measured under the same conditions as the aforementioned sample. The analysis was conducted by defining regions of interest (ROI) for the photopeak. The background gamma-ray spectrum was measured for 600 seconds, and the MDA for each detector was calculated under the same conditions. The energy range of the spectrum was obtained from both the scintillation detector covering 20 – 1800 keV.

Table 1. The samples obtained from KAERI were measured for 3600 seconds using an HPGe detector (Canberra Co., Ltd, USA).

Samples	Sample weight (g)	661.6 keV (^{137}Cs)		1173 keV (^{60}Co)		1332 keV (^{60}Co)	
		MDA (Bq/g)	Activity (Bq/g)	MDA (Bq/g)	Activity (Bq/g)	MDA (Bq/g)	Activity (Bq/g)
231-10	326.7	0.0133	11.579	0.0022	0.0357	-	0.03407
231-20	401.1	0.0171	21.959	0.0045	0.4424	-	0.4475
233-10	282.8	0.125	393.99	0.0403	14.838	-	15.025

2.2 Calculation of Minimum Detectable Activity

In gamma-ray spectroscopy, the term used to describe the detection limit in units of activity is commonly referred to as the MDA. When calculated per unit mass, MDA is defined as

$$MDA_{Currie} = \frac{k^2 + 2k\sqrt{B + B\frac{N}{2m}}}{\varepsilon \cdot i \cdot t \cdot v}$$ (for the identified peak), (1)

where

$$B = \left(\frac{N}{2m}\right)(B_1 + B_2),$$ (2)

and B_1 and B_2 are the sums of the counts in m channels to the left and right of the peak width. B was defined as the background and was calculated based on the ROI of spectrum on specificed gamma-ray photopeak, with a width corresponding to 0.85 times the full width at half maximum (FWHM). The number of channels used to define the peak width, N, is proportional to the photopeak Full Width at Half Maximum (FWHM). In Eq. (1), i is the emission intensity, ε is the detection efficiency, v is the sample mass, and t is the measurement time.

2.3 Comparison of minimum detectable activity according to scintillators

Table 2, 3, and 4 show the MDA for three different samples (231-10, 231-20, and 233-10) measured using $CeBr_3$, $LaBr_3(Ce)$, and NaI(Tl) scintillators. The MDA are provided at three characteristic gamma-ray energies 661.7 keV, 1173 keV, and 1332 keV from ^{137}Cs and ^{60}Co. For ^{137}Cs at 661.7 keV, $LaBr_3(Ce)$ exhibited the lowest MDA across all samples, followed by $CeBr_3$ and NaI(Tl) in increasing order. Similarly, for ^{60}Co at 1173 keV, $LaBr_3(Ce)$ showed the lowest MDA across all samples, with $CeBr_3$ and NaI(Tl) following in the same order. Overall, in all gamma-ray energy, $LaBr_3(Ce)$ demonstrated the best performance across all scintillators in terms of MDA. Meanwhile, NaI(Tl) consistently exhibited the highest MDA across all gamma-ray energy, confirming its relatively lower detection capability.

Table 2. MDA for varying scintillators at 661.7 keV of ^{137}Cs. ROI in spectrum for each radionuclide was the 0.85 times FWHM.

Scintillators	Samples of liquid radioactive waste		
	231-10 (Bq/g)	231-20 (Bq/g)	233-10 (Bq/g)
$CeBr_3$	2.58	3.17	15.73
$LaBr_3(Ce)$	1.95	2.22	10.24
NaI(Tl)	3.21	3.67	17.40

Table 3. MDA for varying scintillators at 1173 keV of ^{60}Co. ROI in spectrum for each radionuclide was the 0.85 times FWHM.

Scintillators	Samples of liquid radioactive waste		
	231-10 (Bq/g)	231-20 (Bq/g)	233-10 (Bq/g)
$CeBr_3$	2.20	1.85	3.21
$LaBr_3(Ce)$	1.65	1.44	2.11
NaI(Tl)	2.32	2.81	10.59

Table 4. MDA for varying scintillators at 1332 keV of ^{60}Co. ROI in spectrum for each radionuclide was the 0.85 times FWHM.

Scintillators	Samples of liquid radioactive waste		
	231-10 (Bq/g)	231-20 (Bq/g)	233-10 (Bq/g)
$CeBr_3$	1.01	1.12	2.87
$LaBr_3(Ce)$	0.88	0.91	1.84
NaI(Tl)	1.29	1.25	4.09

The $LaBr_3(Ce)$ exhibited the lowest MDA**,** while the $CeBr_3$, despite having a slightly higher MDA than $LaBr_3(Ce)$, demonstrated stable detection performance even in the low-energy region due to its relatively low intrinsic background. In contrast, the NaI(Tl), despite having the lowest intrinsic background among the three scintillators, showed higher MDA than $LaBr_3(Ce)$ and $CeBr_3$. This was primarily due to its relatively poor energy resolution, which limits its ability to effectively distinguish low-activity signals. For low-activity samples, it was initially expected that the effect of $LaBr_3(Ce)$'s intrinsic background would be significant. However, since the sample's contribution to the total spectrum was dominant compared to the intrinsic background, $LaBr_3(Ce)$ achieved the lowest MDA values due to its superior energy resolution**.**

3. Conclusions

In this study, we compared the MDA values and radioactivity concentration measurement performance of 1-inch NaI(Tl), $LaBr_3(Ce)$, and $CeBr_3$ scintillator-based detectors under identical conditions for key gamma-emitting radionuclides in liquid radioactive waste.

The detection performance of each detector varied depending on the energy of the radionuclide and the intrinsic background level. Notably, energy resolution and relative efficiency had a significant impact on the measurement results, leading to meaningful differences among the detectors.

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