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Assessment of LaBr$_3$:Ce and CeBr$_3$ scintillators for low-energy ^{55}Fe X-ray detection

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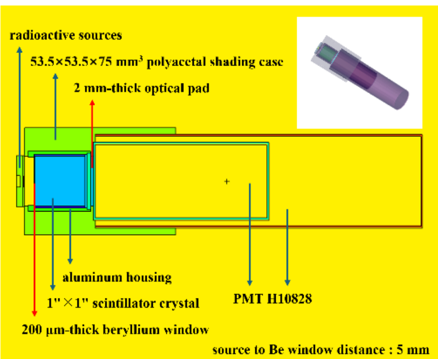
1. Introduction

With the increasing decommissioning of aging nuclear power plants, attention has turned to the detection of radioactive aerosols generated during these processes. Among the prevalent radionuclides in decommissioning waste, ^{55}Fe is particularly significant due to its emission of low-energy (5.9 keV) X-rays, which contribute to radiation exposure risks [1-2]. Conventional ^{55}Fe detection methods, such as liquid scintillation counting and accelerator mass spectrometry, require complex chemical separation steps and are not suitable for field applications [3-4]. Given the widespread use of LaBr$_3$:Ce and CeBr$_3$ scintillators in gamma-ray spectroscopy, this study aims to compare their capabilities in detecting ^{55}Fe, considering the influence of intrinsic background radiation from ^{138}La.

2. Materials and methods

The study employed 1" × 1" cylindrical LaBr$_3$:Ce and CeBr$_3$ scintillators, manufactured by Epic Crystal, enclosed in aluminum housing with a 200 μm-thick beryllium window to enhance low-energy X-ray detection. A Hamamatsu H10828 photomultiplier tube (PMT) was used for light detection, with an optical pad (EJ-560, Eljen Technology) ensuring optimal transmission. The output signal was processed using a charge-sensitive preamplifier (CR-113, Cremat Inc.) and recorded via a digitizer (DT5725, CAEN). Experiments were conducted in a darkroom without additional radiation shielding. The detection efficiency and minimum detectable activity (MDA) were analyzed, and the significance method was applied to assess peak observability. The PMT bias voltage was set to -740 V for LaBr$_3$:Ce and -790 V for CeBr$_3$, with all measurements taken over a 1200-s duration.



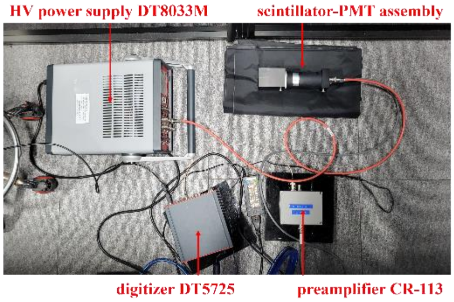


Fig. 1. The scintillator-PMT assembly configuration and the experimental setup.

3. Experimental results

Fig. 2 shows the intrinsic background spectra of LaBr$_3$:Ce and CeBr$_3$, as well as the pulse height spectrum obtained for the ^{55}Fe source. The ^{55}Fe source (4.077 μCi) was positioned 5 mm from the beryllium window of the scintillator, ensuring proper alignment with the detector. The comparison of spectra with and without the ^{55}Fe source revealed a distinctive 5.9 keV peak, which was confirmed by differentiating it from the 5.6 keV peak originating from ^{138}La decay in LaBr$_3$:Ce [5].

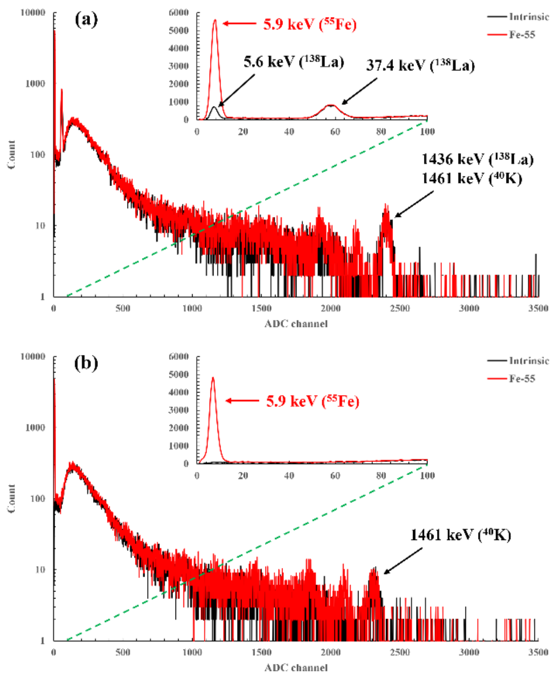
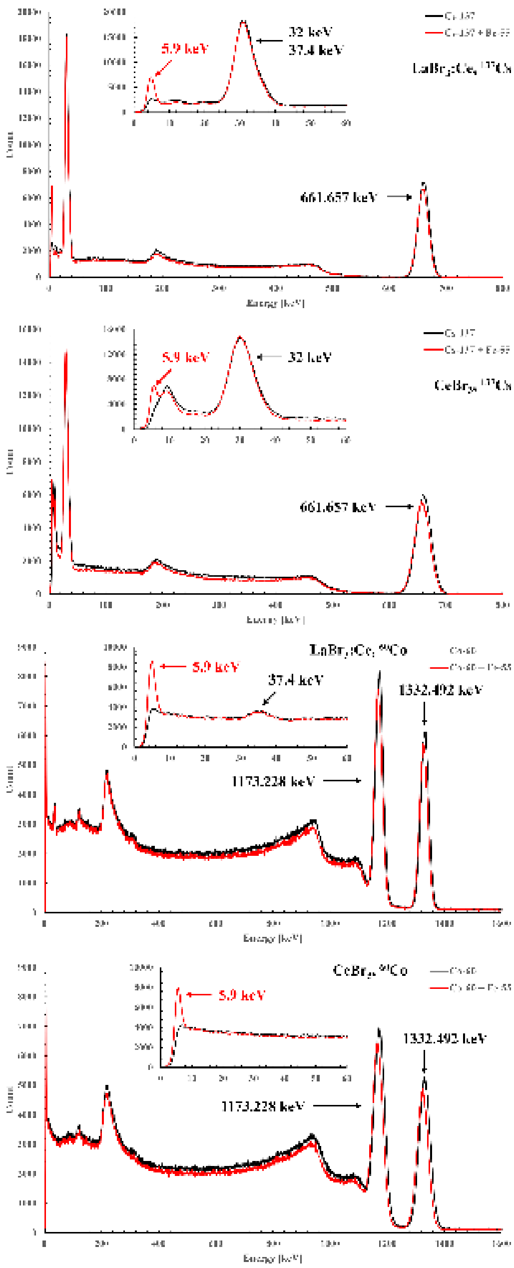


Fig. 2. Pulse height spectra of intrinsic background and ^{55}Fe collected with (a) LaBr$_3$:Ce and (b) CeBr$_3$.

Since real-world decommissioning sites often contain multiple radionuclides, simultaneous measurements of ^{55}Fe with ^{137}Cs and ^{60}Co were conducted to evaluate detector performance under multi-source conditions. As shown in Fig. 3, distinct differences emerge between measurements with and without the ^{55}Fe source. The primary photopeaks, including the low-energy 5.9 keV peak, remain clearly identifiable across all experimental conditions, demonstrating that both LaBr$_3$:Ce and CeBr$_3$ scintillators are capable of simultaneous detection of low-energy radiation even in the presence of high-energy emitters. However, when measuring ^{137}Cs with CeBr$_3$, an additional peak appears around 10 keV, which causes distortion of the 5.9 keV peak. This feature is presumed to result from scattering or sum peaks associated with low-energy X-rays emitted by ^{137}Cs.



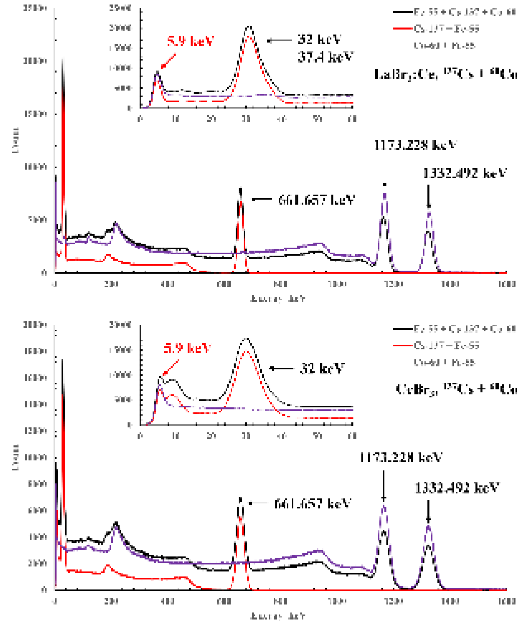


Fig. 3. Energy spectra under multi-source conditions with LaBr$_3$:Ce and CeBr$_3$.

To evaluate the detection capability of LaBr$_3$:Ce and CeBr$_3$, the significance method introduced by Milbrath et al. was applied [6]. The significance (Sig_{quan}) is defined as:

$$(1) \quad Sig_{quan} = Net\ counts/Net\ error,$$

where the net error is given by:

$$(2) \quad Net\ error \approx \sqrt{Background + Gross\ area}.$$

Table 1 presents a comparison of significance values for LaBr$_3$:Ce and CeBr$_3$ under identical conditions (1200 s measurement). The results indicate that LaBr$_3$:Ce exhibited higher significance values across all conditions. However, due to intrinsic background from ^{138}La decay (5.6 keV X-ray), this significance may be slightly overestimated. Additionally, the full width at half maximum (FWHM) of the 5.9 keV peak was found to be better for LaBr$_3$:Ce in most cases.

Table 1. Significance comparison for ^{55}Fe measurements.

Factor	LaBr$_3$:Ce			CeBr$_3$		
	^{55}Fe	^{55}Fe, ^{137}Cs	^{55}Fe, ^{60}Co	^{55}Fe	^{55}Fe, ^{137}Cs	^{55}Fe, ^{60}Co
FWHM	2.37	2.38	2.55	2.63	1.51	2.57
Gross area	23224	34940	48143	19610	28671	45989
Net area	22448	25387	30027	18385	9236	24527
Background	776	9553	18116	1225	19435	21462
Error	154.92	210.93	257.41	144.34	219.33	259.71
Sig_{quan}	144.90	120.36	116.65	127.37	42.11	94.44

To assess the effect of intrinsic background, significance values were recalculated after background subtraction, as shown in Table 2. The recalculated significance (Sig) is defined as follows:

$$(3) \quad Sig = net\ counts/\sqrt{gross\ area}.$$

The recalculated significance revealed that $LaBr_3$:Ce showed a notable decrease compared to its initial significance, confirming that its intrinsic background had contributed to its originally higher values. In contrast, $CeBr_3$ exhibited improved significance, particularly in the presence of ^{137}Cs, reducing the performance gap between the two scintillators. Despite this reduction, $LaBr_3$:Ce still maintained a slight advantage, particularly in simultaneous measurement scenarios. These results suggest that while $CeBr_3$ provides better sensitivity in low-background conditions, $LaBr_3$:Ce remains a more effective choice for field applications where multiple sources coexist.

Table 2. Recalculated significance after background subtraction.

Factor	$LaBr_3$:Ce			$CeBr_3$		
	^{55}Fe	^{55}Fe, ^{137}Cs	^{55}Fe, ^{60}Co	^{55}Fe	^{55}Fe, ^{137}Cs	^{55}Fe, ^{60}Co
Gross area	23224	34940	48143	19610	28671	45989
Net area	19951	16413	19046	18576	14057	17068
Background	3273	18527	29097	1034	14614	28921
Sig	130.92	87.81	86.80	132.65	83.02	79.59

The minimum detectable activity (MDA) was calculated using the Currie method, where the MDA for peaked background scintillators ($LaBr_3$:Ce) is given by:

$$(4) \quad MDA_{peaked} = \frac{2.71+3.29\sqrt{B+B\cdot N/2m}}{\varepsilon\cdot p\cdot t},$$

and for non-peaked background scintillators ($CeBr_3$) is:

$$(5) \quad MDA_{non-peaked} = \frac{2.71+3.29\sqrt{2B}}{\varepsilon\cdot p\cdot t},$$

where B is the background count, ε is the detection efficiency, p is the emission probability, t is the measurement time, N is the number of channels in the ROI, and m is the number of channels used for background estimation [7-8].

Table 3 shows that $LaBr_3$:Ce demonstrated slightly higher detection efficiency than $CeBr_3$; however, due to its intrinsic background, it had a higher MDA, making $CeBr_3$ more sensitive for detecting weak ^{55}Fe activity. Since MDA decreases with extended measurement time, this limitation does not significantly impact long-term monitoring applications.

Table 3. Measurement capabilities of each scintillator for ^{55}Fe.

Factor	$LaBr_3$:Ce	$CeBr_3$
Measurement live time	1200 sec	1200 sec
Detection efficiency	4.04E-04 ± 8.18E-05	3.77E-04 ± 7.62E-05
MDA	1557 Bq	1134 Bq

4. Conclusion

The study demonstrates that $LaBr_3$:Ce and $CeBr_3$ both effectively detect ^{55}Fe, with $LaBr_3$:Ce showing better detection capability despite its intrinsic background. The intrinsic background effect can be mitigated through extended measurement times, making $LaBr_3$:Ce a viable option for long-term field applications. Given its lower cost and better simultaneous measurement performance with other radionuclides, $LaBr_3$:Ce is considered more suitable for portable radioactive aerosol monitoring systems in decommissioning sites. Future studies will focus on evaluating detection performance in real aerosol filter samples.

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