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Pulse height tests of a large diameter fast LaBr₃:Ce scintillation detector

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HIGHLIGHTS

• Pulse height tests of 100 mm × 100 mm cylindrical fast LaBr₃:Ce detector.

Prompt gamma ray measurements.

• Water samples with mercury (3.1 wt%), boron (2.5 wt%), cadmium (0.25 wt%), chromium (52 wt%), and nickel (22 wt%) contamination.

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ABSTRACT

The pulse height response of a large diameter fast 100 mm × 100 mm LaBr₃:Ce detector was measured for 0.1–10 MeV gamma-rays. The detector has a claimed time resolution of 608 ps for 511 keV gamma rays, but has relatively poor energy resolution due to the characteristics of its fast photomultiplier. The detector pulse height response was measured for gamma rays from cobalt, cesium, and bismuth radio-isotope sources as well as prompt gamma rays from thermal neutron capture in water samples contaminated with mercury (3.1 wt%), boron (2.5 wt%), cadmium (0.25 wt%), chromium (52 wt%), and nickel (22 wt%) compounds. The energy resolution of the detector was determined from full width at half maximum (FWHM) of element-characteristic gamma ray peaks in the pulse height spectrum associated with the element present in the contaminated water sample. The measured energy resolution of the 100 mm × 100 mm detector varies from 12.7 ± 0.2% to 1.9 ± 0.1% for 0.1 to 10 MeV gamma rays, respectively. The graph showing the energy resolution $\Delta E/E(\%)$ versus $1/\sqrt{E_{\gamma}}$ was fitted with a linear function to study the detector light collection from the slope of the curve. The slope of the present 100 mm × 100 mm detector is almost twice as large as the slope of a similar curve of previously published data for a 89 mm × 203 mm LaBr₃:Ce detector. This indicates almost two times poorer light collection in the 100 mm × 100 mm detector as compared to the other detector.

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1. Introduction

Recently developed lanthanide-based gamma ray detectors have excellent energy and time resolutions (Favalli et al., 2010; Van Loef et al., 2001). They have high potential for applications in studies involving pulse height spectroscopy (Camera et al., 2014; Giaz et al., 2013; Menge et al., 2007; Naqvi et al., 2011, 2012a, 2012b; Owens et al., 2007) and timing analysis (Iltisa et al., 2006; Kuhn et al., 2004, 2005, 2006; Surti et al., 2003). Detectors designed for pulse height spectroscopy are coupled to photomultipliers with excellent energy resolution while those designed for timing analysis have their scintillator crystals coupled to fast photomultipliers with excellent time resolution (Iltisa et al., 2006; Kuhn et al., 2005). Fast photomultipliers have relatively poor energy resolution while spectroscopy photomultipliers have poor time resolution (Kuhn et al., 2004, 2006; Modamio et al., 2015; Surti et al., 2003). Furthermore, the time resolution of LaBr₃:Ce detectors also depends upon the LaBr₃:Ce crystal size and its cerium doping concentration, better time resolution with higher

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http://dx.doi.org/10.1016/j.apradiso.2015.07.009 0969-8043/© 2015 Elsevier Ltd. All rights reserved. cerium doping concentration (Iltisa et al., 2006; Kuhn et al., 2005, 2006). The energy resolution of the LaBr₃:Ce detector deteriorates with increasing cerium doping due to optical self-absorption of cerium. Due to higher cerium concentration of CeBr₃ detectors than LaBr₃:Ce detectors, CeBr₃ detector has poorer resolution of 4.4% as compared to 2.9% measured for LaBr₃:Ce detector for 661 keV gamma rays (Weele et al., 2014). As the detector volume increases its time resolution deteriorates but its detection efficiency for higher energy gamma rays improves. Additional large diameter detectors offer larger solid angles resulting in reduced counting times in the experiments. The energy and time resolutions of large volume LaBr₃:Ce detectors were previously measured (Giaz et al., 2013). For 76 mm \times 76 mm (diameter \times height) and $90 \times 203 \text{ mm}^2$ (diameter × height) LaBr₃:Ce detectors, the measured energy resolutions were 3.0-3.1% for 661 keV gamma rays; but the reported time resolutions of these detectors for 511 keV gamma rays were 671 and 880 ps, respectively (Giaz et al., 2013).

A large diameter $100 \text{ mm} \times 100 \text{ mm}$ (diameter \times height) LaBr₃:Ce detector has been acquired by King Fahd University of Petroleum and Mineral for the time-flight spectroscopy program. Large volume LaBr₃:Ce detectors designed for timing spectroscopy are coupled to fast photomultipliers which have relatively poor energy resolution. In this study the energy resolution of the large diameter LaBr₃:Ce detector was measured for gamma-rays with energies up to 10.0 MeV. The findings of this study are reported in this paper.

2. Experimental methods

2.1. Measurement of the detector intrinsic activity

The LaBr₃:Ce detector (with 100 mm × 100 mm LaBr₃:Ce crystal, coupled to a fast photomultiplier model number R4144) was supplied to us by the manufacturer Saint Gobain, France, as a single unit along with a matching voltage divider. The detector was operated at negative 994 V voltage. The detector body has a common ground connection with dynode and timing output with grounded-anode configuration of the photomultiplier. The detector time resolution warranted by the manufacturer for 511 keV gamma rays was 608 ps. In the present study, the time resolution of the 100 mm × 100 mm detector was not measured. The manufacturer-quoted time resolution of our detector is better than the time resolution for a 89 mm × 203 mm LaBr₃:Ce detector reported earlier (Giaz et al., 2013). The energy resolution of the 100 mm × 100 mm LaBr₃:Ce detector quoted by the manufacturer for 661 keV gamma rays was 5.3%.

The intrinsic activity of the $100 \text{ mm} \times 100 \text{ mm}$ detector was determined using the procedure described for the smaller 76 mm \times 76 mm LaCl₃:Ce (Naqvi et al., 2012a, 2012b) and LaBr₃:Ce detectors (Nagyi et al., 2011). The activity/second was determined from the area under the 1468 keV peak of the detector as shown in Fig. 1. The lack of a bend on the lower slope of the 1468 keV peak in this spectrum (as observed in other smaller sizes LaBr₃:Ce detectors) is due to poorer energy resolution of the detector caused by the coupled fast photomultiplier with poorer energy resolution. The detector activity was measured for 101 s and was found to be 142 ± 1 counts/s. The photopeak efficiency (PE) of the large detector was calculated from the ratio of the measured activity count rate $(142 \pm 1 \text{ counts/s})$ and the calculated activity of 775 Bq for a $100 \text{ mm} \times 100 \text{ mm}$ LaBr detector. The PE of the large detector was found to be 0.183. The activity of the 100 mm \times 100 mm LaBr detector was calculated from extrapolation of Menge et al. data for smaller sizes LaBr₃:Ce detectors to $100 \times 100 \text{ mm}^2$ LaBr detector. For smaller cylindrical LaBr₃:Ce detectors of 38 mm × 38 mm,

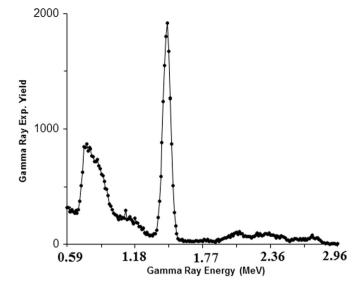


Fig. 1. $100 \text{ mm} \times 100 \text{ mm}$ LaBr₃:C detector intrinsic activity pulse height spectrum exhibiting 1468 keV line from lanthanum.

41 mm × 76 mm, and 51 mm × 76 mm sizes (diameter × height), Menge et al. reported calculated activity of the detector for 1436+32 keV as 42.6 Bq, 99.0 Bq, and 153 Bq, respectively. They also reported photopeak count rates (count/s) for expected/measured count rates for theses detectors as (3.32/3.55), (10.7/10.6) and (21.1/21.8), respectively. From the ratio of measured count rate and calculated activity, the photopeak efficiency was derived (Menge et al., 2007). Previously, the PE of the 76 mm × 76 mm LaBr₃:Ce and LaCl₃:Ce detectors were reported to be 0.191 (Naqvi et al., 2012a) and 0.16 (Naqvi et al., 2012b), respectively.

The energy resolution of the large LaBr₃:Ce detector was measured for 662 keV gamma-rays from ¹³⁷Cs source. The pulse height spectrum of the large detector from ¹³⁷Cs source is shown in Fig. 2. For 662 keV gamma-rays, the energy resolution of the detector was measured to be 5.8%. This is 9% higher than the manufacturer's warranted energy resolution (5.3%) of the detector.

2.2. Measurement of LaBr₃:Ce detector activation spectrum

The gamma-ray tests of the large detector were performed following the method used for the smaller lanthanum-halide

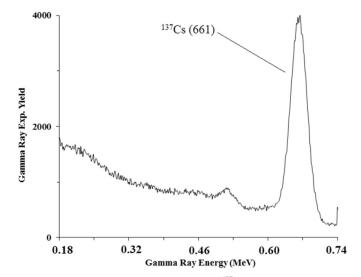


Fig. 2. LaBr₃:Ce pulse height spectrum taken with 137 Cs source exhibiting 662 keV peak.

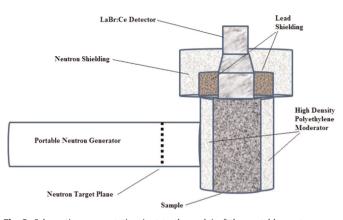


Fig. 3. Schematic representation (not to the scale) of the portable neutron generator-based PGNAA setup used in the prompt gamma-ray tests of the LaBr₃:C detector.

detectors (Naqvi et al., 2011, 2012a, 2012b). The tests were conducted using the PGNAA setup reported earlier (Naqvi et al., 2011, 2012a, 2012b). The setup was modified to accommodate the large LaBr₃:Ce detector. Fig. 3 shows a schematic drawing of the experimental setup used in the present study. Neutrons with 2.5 MeV energy from a D-D portable neutron generator are moderated in a cylindrical high density polyethylene moderator with 250 mm diameter and 140 mm height. The moderator has a 90 mm diameter central cavity drilled through to accommodate a sample bottle with an outer diameter of 90 mm and a height of 140 mm. The axis of the cylindrical sample is parallel to the neutron target plane. The large gamma-ray detector views the sample along its longitudinal axis.

The large detector is shielded against gamma rays and neutrons by 3 mm thick lead shielding and an additional 50 mm thick neutron shielding, built around the lead shield of the LaBr₃:Ce detector. The neutron shielding was made from mixture of paraffin and lithium carbonate (taken in equal weight proportions). The optimum sizes of the moderator and shielding were calculated using Monte Carlo simulations (Naqvi et al., 2011, 2012a, 2012b).

The detector activation spectrum was produced due to the interaction of thermal neutrons with the detector material. The activation spectrum also contains gamma ray peaks due to room background. The energies of the gamma-rays are listed in Table 1 (Choi et al., 2006).

In this study, a flux of 2.5 MeV neutrons was produced via the D (d,n) reaction using a 70 μ A current deuteron beam of 70 keV energy. Due to the extended usage of the neutron generator over more than 1000 h, the neutron source strength was estimated to be 4×10^5 n/s. The beam has a frequency of 250 Hz and a pulse width of 800 μ s. The detector pulse height spectra were acquired over two different energy ranges, a narrow range of 0.09–0.61 MeV for low energy capture gamma-rays from cadmium, boron and mercury and a broader energy range of 1.33–10.0 MeV for high energy gamma rays from nickel, mercury and chromium samples.

Fig. 4 shows a gamma ray spectrum of the large detector over 0.11 to 0.84 MeV energy range exhibiting prominent gamma-ray peaks of lanthanum, cerium and bromine from the detector material. The gamma ray energy data is listed in Table 1 (Choi et al., 2006).

For comparison, the activation spectrum of a smaller 76 mm \times 76 mm LaBr₃:Ce detector (Naqvi et al., 2011) is shown in Fig. 5 over 0.05 to 2.55 MeV range. The energy resolution of the large detector is poorer as compared to the 76 mm \times 76 mm LaBr₃:Ce detector and many peaks could not be resolved from adjacent ones. Fig. 4 shows the lanthanum peaks at 163 and

Table 1

Energies and partial elemental cross section $\sigma_{\gamma}^{z}(E_{\gamma})$ -barns of prominent capture gamma-rays used in this study (Choi et al., 2006).

Element Gamma ray energy (MeV		$\sigma_{\gamma}^{Z}(E_{\gamma})$ -barns	
В	478	716	
Br	196	0.434	
	271	0.462	
	275	0.158	
	315	0.460	
	367	0.233	
	513	0.210	
	661	0.082	
	828	0.285	
	1248	0.0527	
	7577	0.108	
Cd	171	57	
	245	274	
	558	1860	
	651	359	
Ce	475	0.082	
	662	0.241	
	1107	0.040	
Hg	368	251	
	5967	62.5	
Н	2223	0.333	
Ni	8533	0.721	
	8998	1.49	
Cr	7099	0.146	
CI CI	7938	0.424	
	8483	0.121	
	8511	0.233	
	8884	0.780	
	9719	0.260	
La	163	0.489	
Lu	272	0.502	
	288	0.73	
	567	0.335	
	595	0.103	
	789	Intrinsic	
	1436+32	Intrinsic	
	2521	0.212	
	5126	0.212	
	J120	0.114	

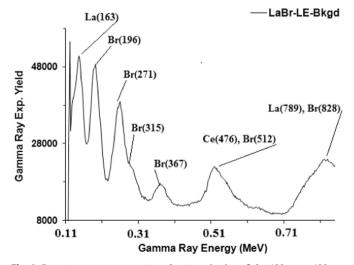


Fig. 4. Prompt gamma-ray spectrum due to activation of the 100 mm \times 100 mm LaBr₃:Ce detector caused by capture of thermal neutrons in La, Br and Ce elements present in LaBr₃:Ce detector.

789 keV along with the bromine peaks at 196, 276, 315, 367, and 512 keV. The Ce peak at 476 keV could not be resolved from the Br peak at 512 keV and from the La peaks at 567 and 595 keV. Also, the La peak at 789 keV could not be resolved from the Br peak at 828 keV. The detector energy resolution can be calculated from the

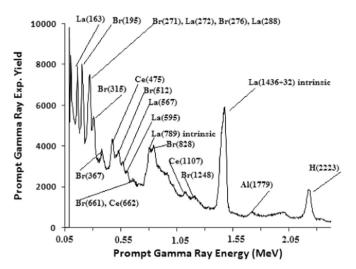


Fig. 5. Prompt gamma-ray spectrum due to activation of the 76 mm \times 76 mm LaBr: Ce detector caused by capture of thermal neutrons in La, Br and Ce elements present in the LaBr₃:Ce detector (Naqvi et al., 2011).

activation spectrum for well resolved peaks of significant intensities.

3. Measurements of prompt gamma-ray spectra from boron-, cadmium-, mercury-, chromium-, and nickel-contaminated water samples

The spectra of the large detector were recorded from mercury, boron-, cadmium-, chromium-, and nickel-contaminated water samples over 0.3 to 10 MeV gamma-ray energies. The detector spectra were acquired with two different energy ranges. The gamma-ray spectra from mercury, boron and cadmium were acquired over 0.09 to 0.61 MeV energy range with data acquisition time listed separately for each sample in Table 2. The gamma-ray spectra from chromium, mercury, and nickel were acquired over 1.33–10.0 MeV energy range. The mercury, boron, cadmium, chromium and nickel samples were prepared by dissolving various chemical compounds, such as boric acid, cadmium acetate, mercuric nitrate, chromium trioxide and nickel nitrate, in water and filling them in the 90 mm diameter × 145 mm long plastic (PET) bottles.

These samples were supplied by the Department of Chemistry, King Fahd University of Petroleum and Minerals, Saudi Arabia. The concentrations of the various elements along with corresponding sample measurement times are listed in Table 2. The samples' measurement times vary from 480 s to 6400 s.

Figs. 6, 8 and 10 show the detector pulse height spectra over 0.09 to 0.61 MeV gamma-rays from the mercury, boron and

Table 2

Chemical composition and concentration of various elements samples used in the present study.

Chemical compound	Element	Concentration (wt%)	Measurement time (s)
Boric acid H ₃ BO ₃	В	2.5	720
Cadmium acetate Cd(CH ₃ CO ₂) ₂	Cd	0.25	480
Mercuric nitrate Hg(NO ₃₎₂	Hg	3.1	1600
Nickel nitrate Ni(NO ₃) ₂	Ni	22	4600
Chromium trioxide CrO ₃	Cr	52	6400

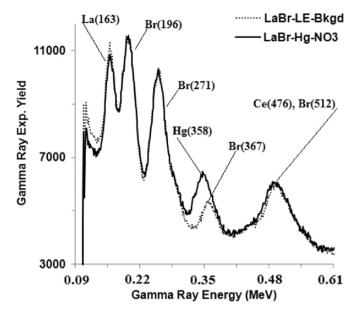


Fig. 6. Prompt gamma-rays pulse height spectra of mercuric nitrate (NiNO₃) contaminated water sample superimposed upon background spectrum.

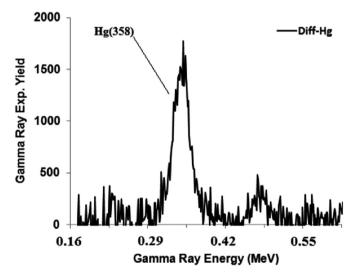


Fig. 7. Difference pulse height spectra of mercuric nitrate contaminated water sample exhibiting 358 keV mercury peak.

cadmium samples superimposed upon background spectra while Figs. 7, 9 and 11 show the background-subtracted peaks of mercury, boron and cadmium samples. In order to superimpose the sample spectra upon the background spectra, both sample and background spectra were normalized to the same counting time and same neutron flux. This was obtained by normalizing both sample and background spectra in the region where background is constant (sample independent). Then, the background spectrum was subtracted from normalized spectrum to generate the difference spectrum.

Figs. 12 and 14 show detector pulse height spectra over 1.33–10.0 MeV gamma-rays from the chromium and nickel samples while Fig. 16 shows the detector pulse height spectrum over 0.54–9.42 MeV gamma rays from the mercury sample. Figs. 13, 15 and 17 are subsections of Figs. 12, 14 and 16 showing enlarged plots of chromium, nickel and mercury peaks.

Fig. 6 shows the spectrum of mercury-contaminated water sample superimposed upon the background spectrum over 0.09 to 0.61 MeV. The mercury peak at 358 keV interferes with the

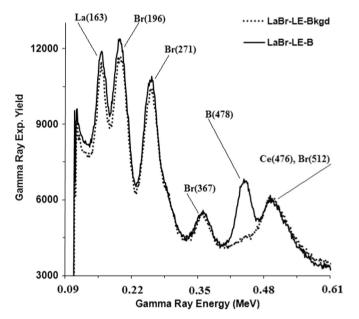


Fig. 8. Prompt gamma-rays pulse height spectra of boric acid contaminated water sample superimposed upon background spectrum.

bromine peak at 367 keV from the detector background. Fig. 7 shows the well resolved 358 keV mercury peak in the difference spectrum, obtained after subtracting background from mercury sample spectrum. Similarly, Fig. 8 shows the boron contaminated water sample spectrum superimposed upon background spectrum. The boron peak at 478 keV is interfering with cerium and bromine peaks at 476 and 512 keV, respectively. Fig. 9 shows the well resolved 478 keV boron peak in the difference spectrum, obtained after subtracting background from boron spectrum. The cadmium spectra acquired by the large detector are shown in Figs. 10 and 11. As shown in Fig. 10, the cadmium peak at 558 keV interferes with the cerium and bromine peaks at 476 and 512 keV, respectively. Fig. 11 shows the well resolved 558 keV cadmium peak in the difference spectrum, obtained after subtracting background after subtracting background spectrum.

Fig. 12 shows the spectrum of chromium contaminated water sample superimposed upon the background spectrum over 1.33 to 10.0 MeV. The chromium peaks at 7099, 7938, 8884 and 9719 keV appear at the end of the spectrum. Also shown in Fig. 12, is the

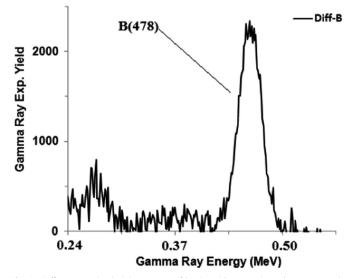


Fig. 9. Difference pulse height spectra of boric acid contaminated water sample exhibiting 478 keV boron peak.

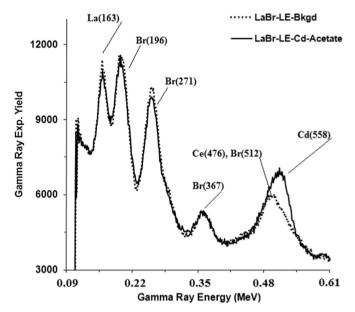


Fig. 10. Prompt gamma-rays pulse height spectra of cadmium-acetate contaminated water sample superimposed upon background spectrum.

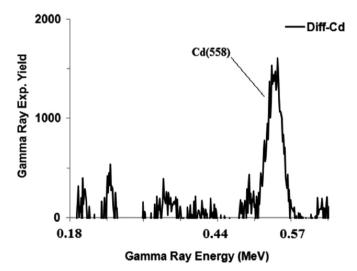


Fig. 11. Difference pulse height spectra of cadmium-acetate contaminated water sample exhibiting 558 keV cadmium peak.

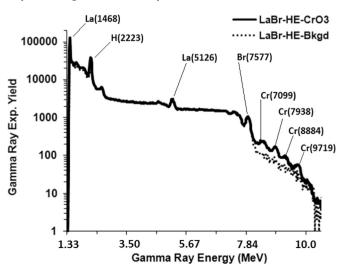


Fig. 12. Prompt gamma-rays pulse height spectra of chromium trioxide (CrO₃) contaminated water sample superimposed upon background spectrum.

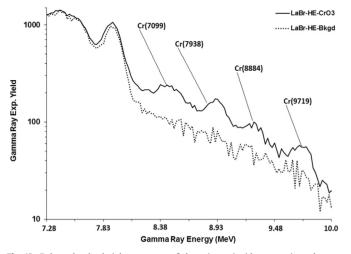


Fig. 13. Enlarged pulse height spectrum of chromium trioxide contaminated water sample exhibiting 7099, 7938, 8884 and 9719 keV chromium peaks superimposed upon background spectrum.

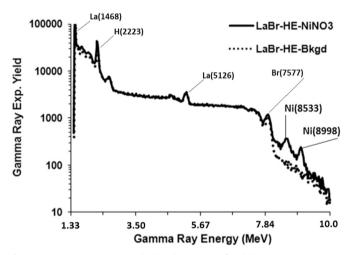


Fig. 14. Prompt gamma-rays pulse height spectra of nickel nitrate (NINO₃) contaminated water sample superimposed upon background spectrum.

hydrogen capture peak at 2223 keV due to capture of thermal neutrons in the high density polyethylene. The lanthanum and bromine peaks from thermal neutron capture in the detector material appear at 5126 and 7577 keV, respectively. Fig. 13 shows the enlarged part of Fig. 12 over 7.28-10.0 MeV exhibiting the chromium peaks at 7099, 7938, 8884 and 9719 keV, along with the background spectrum. Similarly, Fig. 14 shows the spectra of nickel contaminated water sample superimposed upon background spectrum over 1.33-10.0 MeV. The nickel peaks at 8533 and 8998 keV appear at the end of the spectrum. Fig. 15 shows the enlarged part of Fig. 14 over 6.77 to 10.0 MeV exhibiting the nickel peaks at 8533 and 8998 keV along with the single escape peak superimposed upon the background spectrum. The high energy gamma-ray spectrum of mercury-contaminated water sample, superimposed upon background spectrum, is shown in Fig. 16 over 0.54 to 10.0 MeV. The mercury peak could be seen at 5967 keV energy. The 5967 keV mercury peak along with its associated single escape peak, superimposed upon the background, is shown on an enlarged scale in Fig. 17.

4. Results and discussion

The gamma ray peaks data from the detector material, as well as from boron, cadmium, mercury, chromium and nickel samples

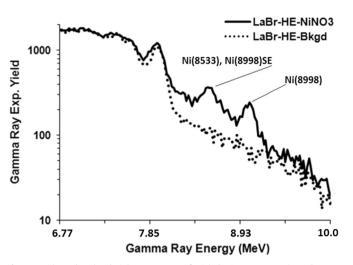


Fig. 15. Enlarged pulse height spectrum of nickel nitrate contaminated water sample exhibiting 8533 and 8998 keV nickel peaks superimposed upon background spectrum.

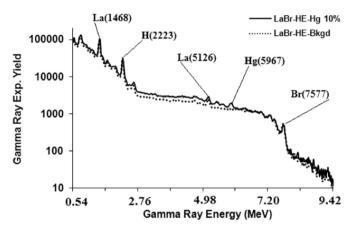


Fig. 16. Prompt gamma-rays pulse height spectra of mercuric nitrate (HgNO₃) contaminated water sample superimposed upon background spectrum.

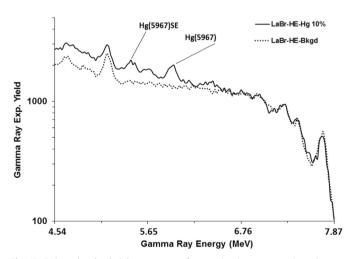


Fig. 17. Enlarged pulse height spectrum of mercuric nitrate contaminated water sample exhibiting 5967 keV mercury peak superimposed upon background spectrum.

was fitted to determine the energy resolution (FWHM) of the detector using the least squares fit method. In order to investigate the effect of light collection from the LaBr₃:Ce crystal, the energy

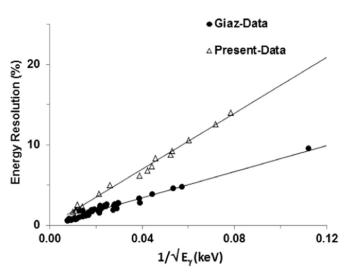


Fig. 18. Energy resolution (%) of the 100 mm × 100 mm LaBr₃:Ce detetor and 89 mm × 203 mm LaBr₃:Ce detetor (Camera et al., 2014; Giaz et al., 2013) plotted as a function of $1/\sqrt{E_{\gamma}}$.

Table 3

Coefficient of fit to energy resolution data of 100 mm × 100 mm and 89 mm × 203 mm LaBr₃:Ce detectors. (Camera et al., 2014; Giaz et al., 2013). Fitted equation: $\Delta E/E(\%) = a/(\sqrt{E_g}) + b$

Detector size	Fit coefficient		
	Coefficient a	Coefficient b	
100 mm × 100 mm 89 mm × 203 mm	174.4 80.9	0.011 0.224	

resolution (%) was plotted against $1/\sqrt{E_{\gamma}}$. The slope of the fit is indicative of the light collection of the detector assembly. For comparison, the energy resolution data of a 89 mm × 203 mm LaBr₃:Ce detector coupled to a photomultiplier with good energy resolution (Camera et al., 2014; Giaz et al., 2013), was also plotted against $1/\sqrt{E_{\gamma}}$. Since both detectors contains 5% Ce each, the only difference between them is more efficient light collection in the 89 mm × 203 mm LaBr₃:Ce detector (Camera et al., 2014; Giaz et al., 2014; Giaz et al., 2013).

Fig. 18 shows the energy resolution data of the 89 mm × 203 mm LaBr₃:Ce and 100 × 100 mm² LaBr₃:Ce detector plotted as a function of $1/\sqrt{E_{\gamma}}$. The solid line through the data represents linear least squares fit to the data of the type:

$$\Delta E/E(\%) = a/(\sqrt{E_g}) + b$$

The values of the coefficients *a* and *b* of the fit to both detectors' energy resolution data are listed in Table 3. The slope of the curve (value of coefficient *a*) for 100 mm × 100 mm detector was calculated to be 174.4 while for the 89 mm × 203 mm LaBr₃:Ce detector its value was 80.9. The almost two times smaller value of the slope for the 89 mm × 203 mm LaBr₃:Ce detector indicates almost two times better light collection for 89 mm × 203 mm LaBr₃:Ce detector. This is due to the fact that the photomultiplier used in the $100 \times 100 \text{ mm}^2 \text{ LaBr}_3$:Ce detector has poorer energy resolution but superior timing resolution. The manufacturer claimed an excellent time resolution of 608 ps for the $100 \times 100 \text{ mm}^2 \text{ LaBr}_3$:Ce detector as compared to 880 ps reported for the 89 mm × 203 mm LaBr₃:Ce detector as more the slope for the 89 mm × 203 mm LaBr₃:Ce detector as compared to 880 ps reported for the 89 mm × 203 mm LaBr₃:Ce detector.

5. Conclusions

Performance tests of a large diameter cylindrical $100 \text{ mm} \times 100 \text{ mm}$ (height × diameter) LaBr₃:Ce detector were carried out for 0.10-10 MeV gamma-rays produced via thermal neutron capture in the detector material as well in mercury-, boron-, cadmium-, chromium- and nickel-contaminated water samples. From the measured gamma-ray spectra, the energy resolution of the 100 mm \times 100 mm LaBr₃:Ce detector was derived. The percentage energy resolution data of the 100 mm \times 100 mm LaBr₃:Ce detector was fitted as a function of $1/\sqrt{E_{\gamma}}$ using a linear least square fit. For comparison, previously published energy resolution data of a 89 mm \times 203 mm LaBr₃:Ce detector (Giaz et al., 2013) was also fitted as a function of $1/\sqrt{E_{\gamma}}$. The difference in the slopes of the fits reveals almost twice poorer light collection from the $100 \times 100 \text{ mm}^2$ LaBr₃:Ce detector. This study has provided useful data on the performance of the large fast $100 \text{ mm} \times 100 \text{ mm} \text{ LaBr}_3$:Ce detector.

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