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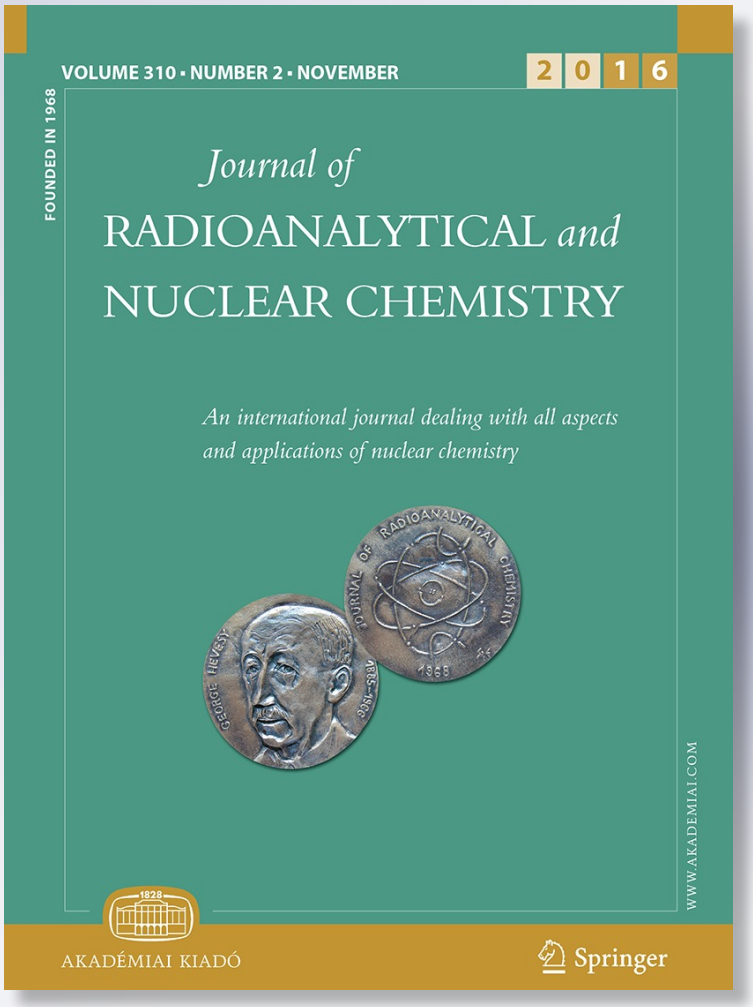

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Energy resolution measurements of $CeBr_3$ and $LaCl_3$:Ce detectors

A. A. Naqvi[1] · F. Z. Khiari[1] · F. A. Liadi[1] · Khateeb-ur-Rehman[1] · A. A. Isab[2]

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Abstract The energy resolution of $LaCl_3$:Ce and $CeBr_3$ detectors were measured from gamma rays produced due to thermal neutron capture in $CeBr_3$ and $LaCl_3$:Ce detector's material using a portable DD neutron generator-based prompt gamma ray neutron activation analysis (PGNAA) setup. The energy resolution of $CeBr_3$ detector varies from 6.64 ± 0.04 to 2.75 ± 0.04 % over 368–2223 keV gamma rays while for $LaCl_3$:Ce it varies from 7.09 ± 0.02 to 0.90 ± 0.01 % over 272–8579 keV gamma rays. The energy resolution data of both detectors exhibit $1/\sqrt{E_\gamma}$ dependence.

Keywords Energy resolution measurements of $LaCl_3$:Ce and $CeBr_3$ detectors · Thermal neutron activation spectra of $LaCl_3$:Ce and $CeBr_3$ detectors · Portable neutron generator-based prompt gamma ray neutron activation analysis (PGNAA) setup

Introduction

In prompt gamma neutron activation analysis (PGNAA) studies based upon thermal neutron capture reactions or neutron inelastic scattering, both sample as well as detector are exposed to thermal as well as fast neutron beams [1–5]. Due to the interaction of the neutron beams with elements present in the detector material, the detector records prompt gamma rays associated with detector material along with the prompt gamma rays associated with the sample to be analyzed [3–5]. Activation gamma rays from the detector material along with any other intrinsic activity gamma rays from the detector material are main source of beam-associated background in prompt gamma ray studies [6–8]. This background sources put major limitations on the use of specific detectors in PGNAA applications. It is highly desirable to use a gamma ray detector with excellent energy and time resolutions in prompt gamma applications [4–8]. Furthermore, to reduce the beam associated background of the detector, it should be made of neutron radiation hardened material (material with insignificant neutron interaction cross section) and free of intrinsic activity. Recently neutron radiation hardened lanthanum-hallide ($LaBr_3$:Ce and $LaCl_3$:Ce) detectors have been developed. $LaBr_3$:Ce and $LaCl_3$:Ce detectors offer excellent energy resolution but they have high intrinsic activity of radioactive lanthanum material in $LaBr_3$:Ce and $LaCl_3$:Ce detectors [1–8]. Studies for minimizing the intrinsic activities of lanthanum-hallide detectors have led to the development of cerium tribromide ($CeBr_3$) gamma ray detectors [9–16]. Like $LaCl_3$:Ce detectors, $CeBr_3$ detectors have also about 4.4 % energy resolution for 661 keV gamma rays [11]. Since the $CeBr_3$ detector does not contain Lanthanum, it has less activity as compared to $LaCl_3$:Ce detector. Ideally the $CeBr_3$ detector should be free of intrinsic activity because Ce and Br are both non-radioactive. The residual intrinsic activity of $CeBr_3$ detector is due to impurity of ^{227}Ac in the detector raw material. Its intrinsic activity is associated with gamma rays emitted by the alpha emitter of ^{227}Ac impurity in the detector (produced by alpha particles of energies 5716 keV (^{223}Ra); 6000 keV (^{227}Th); 6623 keV (^{211}Bi); 6819 keV (^{219}Rn); 7386 keV (^{215}Po); 5716 keV (^{223}Ra)) [11]. The

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energies of intrinsic gamma rays of the $CeBr_3$ detector, produced due to the activity of ^{227}Ac impurity, are spread over the 1400–2200 keV energy range. The $LaCl_3$:Ce has an intrinsic gamma ray background spread over 0.09–1.600 MeV with a lanthanum decay associated peak at 1468 keV [3–8]. The $CeBr_3$ detector has a 1.33 times lower alpha/gamma ratio as compared to $LaBr_3$:Ce/$LaCl_3$:Ce detectors [11]. The higher Ce doping concentration in the $CeBr_3$ detector has resulted in a poorer energy resolution of 4.4 % as compared to $LaBr_3$:Ce detector (2.9 %) for 661 keV gamma rays [14]. This excessive cerium doping has also resulted in a poorer time resolution of $CeBr_3$ detector as compared to $LaBr_3$:Ce and $LaCl_3$:Ce detectors [14].

Although Ce:Br_3 detector has 25 % poorer energy resolution for 661 keV ^{137}Cs gamma rays as compared to $LaBr_3$:Ce, yet it has a reduced intrinsic activity and one-order of magnitude increased detection efficiency around 1461 and 2641 keV gamma ray energies as compared to $LaBr_3$:Ce and $LaCl_3$:Ce detectors [11]. These gamma ray energy ranges are particularly important for detection of ^{40}K and ^{208}Tl(^{232}Th) in environmental samples [11]. Due to their lower intrinsic activity, PGNAA set ups employing Ce:Br_3 detectors, are expected to have better signal to noise ratio in gamma ray spectra and hence better performance compared to those employing $LaBr_3$:Ce and $LaCL_3$:Ce detectors.

A cylindrical 3 × 3 inches (76 × 76 mm) (height × diameter) $CeBr_3$ has been acquired by King Fahd University of Petroleum and Minerals (KFUM), Dhahran, Saudi Arabia from Scionix Holland BV, The Netherland for its prompt gamma analysis program. In this study the activation spectra of 76 × 76 mm (height × diameter) $CeBr_3$ and $LaCl_3$:Ce detectors were measured using a portable neutron generator-based PGNAA setup. Finally, the energy resolution data of both detectors were determined for the resolved prompt gamma rays in the activation spectrum. The results of this study are presented in the following paragraphs.

Experimental

Intrinsic activity spectra of Ce:Br_3 and $LaCl_3$:Ce detectors

The intrinsic activity spectrum of the Ce:Br_3 detector was recorded using standard NIM electronics modules, used in previous studies of $LaCl_3$:Ce and [8] and $LaBr_3$:Ce [4] detectors. For continuity sake it will be briefly described here. The detector signal, which was routed through a preamplifier, was processed through a spectroscopy amplifier with a shaping time of 1 μs. The amplifier signal was processed by a Multichannel Buffer (ADC) for subsequent storage in a personal computer.

Figure 1 shows the Ce:Br_3 detector intrinsic activity spectrum over 1200–2200 keV energy range with gamma ray yield (counts) plotted as a function of gamma ray energy. The intrinsic activity spectrum of the $CeBr_3$ detector is superimposed upon the ^{207}Bi gamma ray source spectrum taken with the detector. The detector intrinsic activity spectrum a significantly reduced background below 1.2 MeV energy. Figure 1 shows three gamma rays peaks at 1510, 1715 and 1990 keV associated with the ^{227}Ac contaminants intrinsic activity of the $CeBr_3$ detector. This agrees with the results of intrinsic activity of a $CeBr_3$ detector due to ^{227}Ac contaminants published earlier by Quarati et al. [11].

The intrinsic activity of the cylindrical (with 76 × 76 mm dimension) $LaCl_3$:Ce detector was also recorded following a procedure similar to the one used for the $CeBr_3$ detector. Figure 2 shows the intrinsic spectrum of the $LaCl_3$:Ce detector showing the 1468 keV intrinsic activity peak due to the decay of radioactive lanthanum impurity in the $LaCl_3$:Ce detector superimposed upon ^{207}Bi source spectrum taken with the detector. Details of the origin of this activity have been published earlier [8]. Quarati et al. [11] have found that the intrinsic activity of a Ce:Br_3 detector is an order of magnitude less than the intrinsic activities of the $LaBr_3$:Ce and $LaCl_3$:Ce detectors of equivalent size.

Measurement of activation spectrum of $CeBr_3$ and $LaCl_3$:Ce detectors

In the present study activation spectra of $CeBr_3$ and $LaCl_3$:Ce detectors were acquired following the procedure described earlier for a 100 × 100 mm (diameter × height)

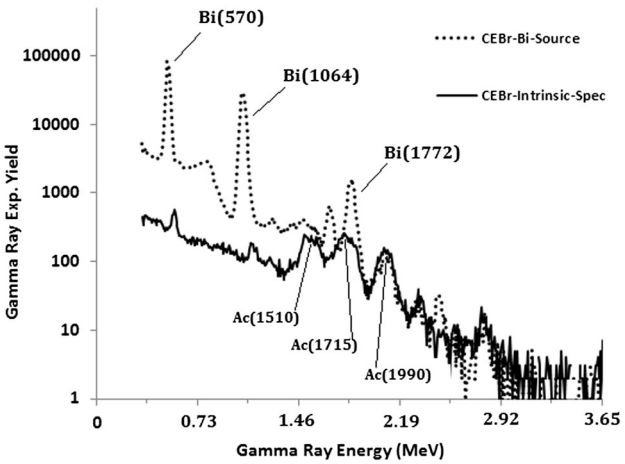

Fig. 1 $CeBr_3$ detector intrinsic activity spectrum superimposed upon ^{207}Bi source spectrum

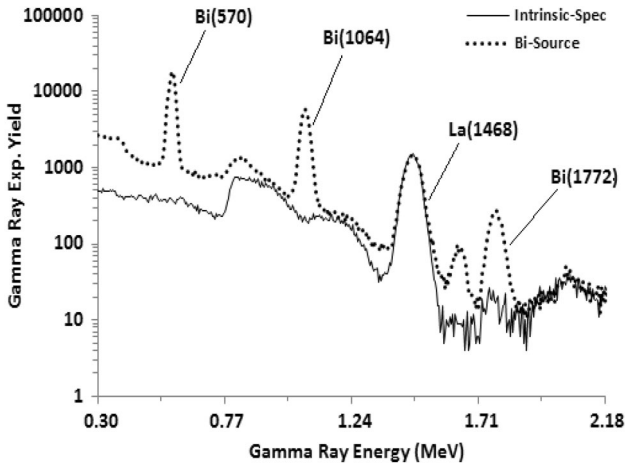

Fig. 2 $LaCl_3$:Ce detector Intrinsic activity spectrum superimposed upon ^{207}Bi source spectrum

$LaBr_3$:Ce detector [3]. For continuation sake it will be described briefly. The PGNAA setup consists of a cylindrical specimen placed inside a cylindrical high density polyethylene moderator. The moderator has a central cylindrical cavity that can accommodate a specimen with a maximum diameter of 9.0 cm and a height of 14 cm. A cylindrical 76 × 76 mm (diameter × length) Ce:Br_3 gamma-ray detector, with its longitudinal axis aligned along the moderator and sample's major axis, was used in this study. The longitudinal axis of the cylindrical sample is also at right angle with respect to the neutron beam axis. In order to prevent undesired gamma-rays and neutrons from reaching the detector, 3 mm thick lead shielding and 50 mm thick paraffin shielding were provided around the gamma-ray detector. The neutron shielding is made of a mixture of paraffin and lithium carbonate mixed in equal weight proportion. The moderator and shielding size was determined from the results of Monte Carlo calculations. Figure 3 shows the PGNAA setup with the high density polyethylene cylindrical moderator and portable neutron generator.

In order to record the activation spectrum of the $CeBr_3$ and $LaCl_3$:Ce detectors, they were irradiated with a mixture of fast as well as thermal neutrons produced in the portable neutron generator-based PGNAA set up. The neutron generator produces 2.5 MeV neutrons via D (d, n) reaction which are thermalized by the moderator. Due to the finite efficiency of the moderator, the neutron flux in the moderator cavity is a mixture of fast and thermal neutrons. The activation spectrum of the detector was recorded without sample. A pulsed beam of 2.5 MeV neutrons was produced via the D (d, n) reaction using a 70 μA deuteron beam of 70 keV energy. The pulsed deuteron beam had a pulse width of 800 μs and a frequency of 250 Hz. The detector pulse height spectrum was acquired using standard NIM electronics modules. The detector signal, which was routed through a preamplifier, was processed through a spectroscopy amplifier with a shaping time of 1 μs. The logical gate signal was generated for each signal processed by the amplifier using a single channel analyzer and gate and delay generators modules.

The activation spectrum of the $CeBr_3$ detector was recorded for 40 min runs. It contains prompt gamma-ray peaks due to capture of thermal neutrons in Br and Ce elements present in the $CeBr_3$ detector, along with small intrinsic activity peaks due to ^{227}Ac contamination. Energies of prominent prompt gamma-rays due to the capture of thermal neutrons in bromine and cerium in the $CeBr_3$ detector were taken from ref [17]. Figure 4 shows the activation spectrum of the $CeBr_3$ detector measured over

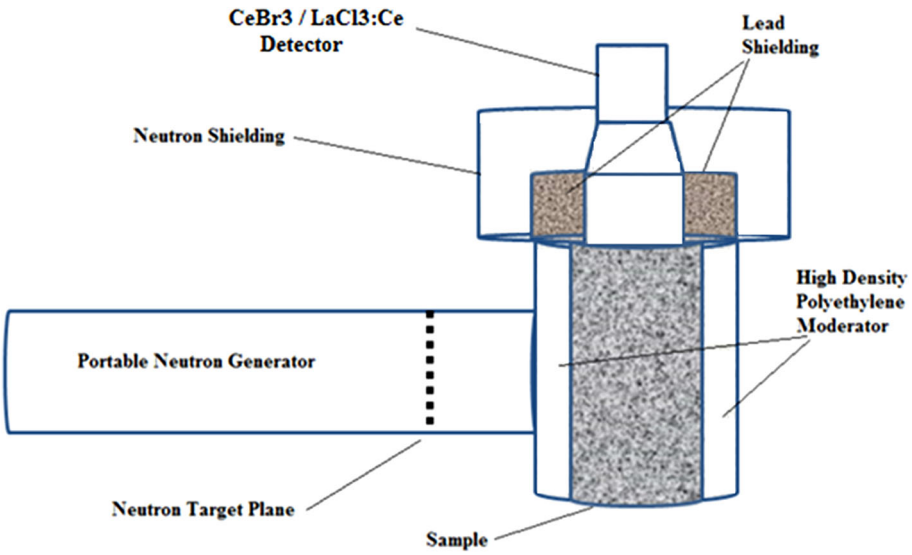

Fig. 3 Schematic diagram of the portable neutron generator based PGNAA setup

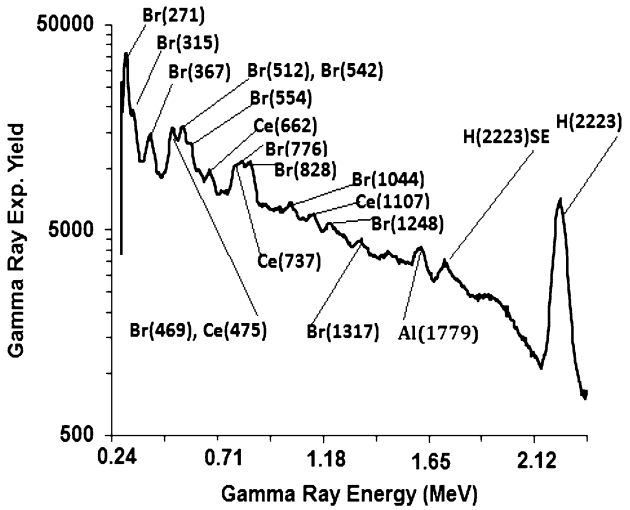

Fig. 4 Activation spectrum of the $CeBr_3$ Detector

0.24–2.35 MeV energy. It contains most of the prompt gamma-ray lines of bromine and cerium in the detector along with the 2.22 MeV hydrogen capture peak of thermal neutrons in the moderator.

Figure 5 shows an enlarged part of the $CeBr_3$ detector activation spectrum over 0.0–1.10 MeV superimposed upon ^{137}Cs source spectrum. The spectrum shows well resolved Br(196), Br(271) and Br(367) peaks due to capture of thermal neutrons in bromine within the $CeBr_3$ detector material. Similar bromine energy peaks were also observed before along with La(163) peak in the activation spectrum of a 100 × 100 mm $LaBr_3$:Ce detector, as shown in Fig. 6 [3].

The activation spectrum of the $LaCl_3$:Ce detector was recorded for 30 min. Figure 7 shows the $LaCl_3$:Ce detector activation spectrum over 0.20–2.40 MeV gamma rays. The

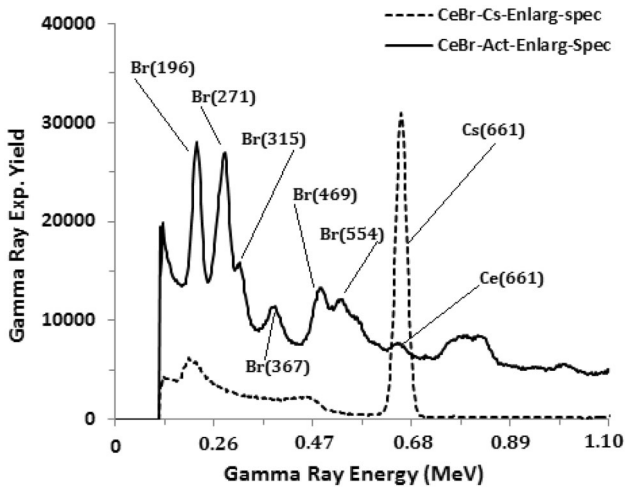

Fig. 5 Portion of $CeBr_3$ activation spectrum superimposed upon ^{137}Cs Source spectrum

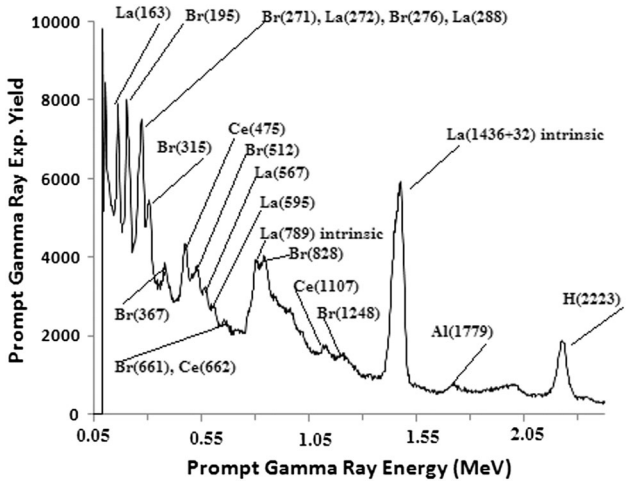

Fig. 6 Activation spectrum of LaBr3:Ce detector taken from ref. [4]

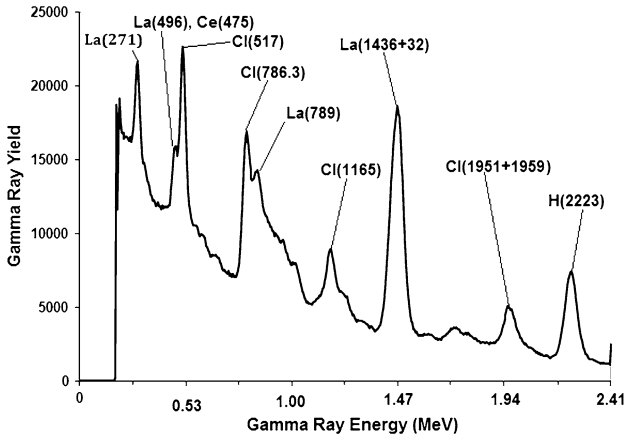

Fig. 7 Activation spectrum of $LaCl_3$:Ce detector

spectrum shows well resolved peaks of La(271), Cl(517), Cl(1165), Cl(1951–1959) and H(2223) peaks. Similar lanthanum and cerium peaks were reported in the activation spectrum of a 76 × 76 mm (diameter × height) $LaBr_3$:Ce detector shown in Fig. 6 from Ref. [4].

Due to short irradiation times, delayed gamma-rays from ^{140}La (half life = 40.3 h) could not be detected. Figure 8 shows an enlarged part of the $LaCl_3$:Ce detector activation spectrum over 2.13–9.69 MeV. The spectrum shows well resolved Cl(2470), Cl(2864), Cl(6111), Cl(6620–6628), Cl(6978) and Cl(8679) peaks due to the capture of thermal neutrons in chlorine in the $LaCl_3$:Ce detector material.

Figure 6 Activation spectrum of $LaBr_3$:Ce detector taken from Ref. [4].

Later on, all the well resolved energy peaks of the $CeBr_3$ and $LaCl_3$:Ce detectors in the full as well as in the enlarged activation spectra were used to calculate each detector energy resolution for the corresponding gamma ray energy.

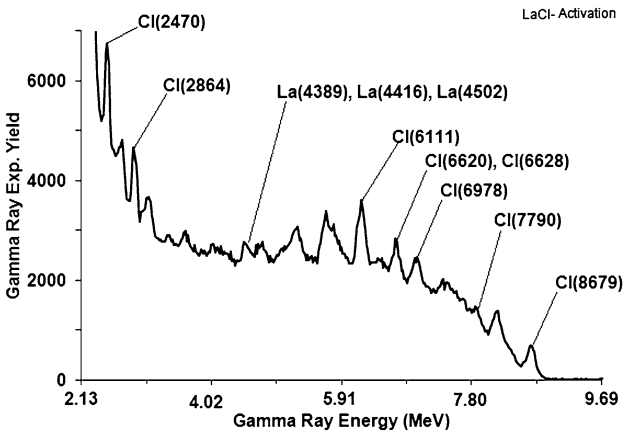

Fig. 8 Portion of Activation spectrum of $LaCl_3$:Ce detector

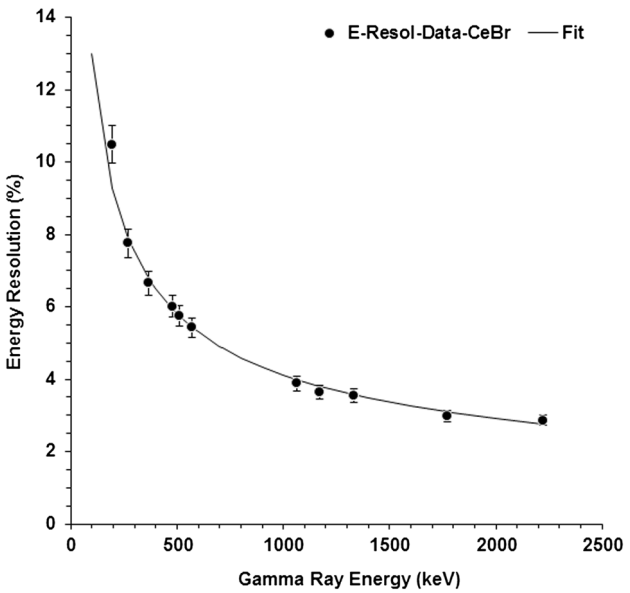

Fig. 9 Energy resolution ΔE/E (%) data of the $CeBr_3$ detector superimposed upon least square fit of type ΔE/E (%) = a/E^b

Energies of prominent prompt gamma-rays due to the capture of thermal neutrons in lanthanum, chlorine, and cerium in the $LaCl_3$:Ce detector were taken from ref [17].

Results and discussion

The energy resolution of the $CeBr_3$ detector was measured over 368 to 2223 keV energy using ^{207}Bi and ^{137}Cs gamma ray sources peaks. Also used for this purpose were the Br(196), Br(271), Br(367), Ce(475), Br(512), Br(554), Br(1064), Ce(1107), and Br (1317) peaks of the activation spectrum from the $CeBr_3$ detector, along with the 2223 keV hydrogen capture peak from the moderator as well as the single escape peak of H (2223). For

Table 1 Coefficients of fit of type ΔE/E(%) = a/E^b to the energy resolution data of the $CeBr_3$, $LaCl_3$:Ce, and $LaBr_3$:Ce detectors

Detector type	Detector size	ΔE/E(%) = a/E^b	
		a	b
$CeBr_3$	76 × 76 mm	130 ± 0.2	0.507 ± 0.025
$CeBr_3$[12]	50 × 50 mm	108	0.498
$LaBr_3$:Ce[12]	50 × 50 mm	81	0.501
$LaCl_3$:Ce	76 × 76 mm	122 ± 0.1	0.505 ± 0.010
$LaBr_3$:Ce[3]	100 × 100 mm	174	0.484

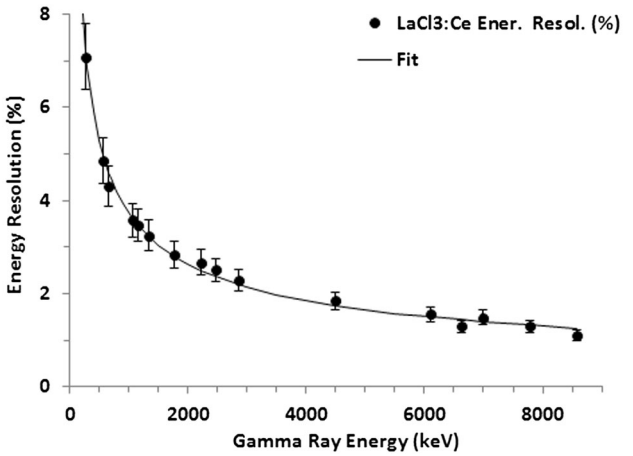

Fig. 10 Energy resolution ΔE/E (%) data of the $LaCl_3$:Ce detector superimposed upon least square fit of type ΔE/E (%) = a/E^b

368–2223 keV gamma rays, the energy resolution of the $CeBr_3$ detector varies from 6.64 ± 0.04 to 2.75 ± 0.04 %. In Fig. 9, the energy resolution data of the $CeBr_3$ detector is plotted as a function of gamma ray energy. A least square fit of type ΔE/E (%) = a/E^b was used to fit the data. The program was developed following the procedure given in a standard textbook on data reduction and error analysis techniques. [18]. Figure 9 shows experimental data points superimposed upon the solid line fit. In the fitted function, a and b are constants and E_γ is the gamma ray energy given in keV. The values of the constants a and b of the fit to the measured energy resolution data of the 76 × 76 mm $CeBr_3$ detector are listed in Table 1.

The energy resolution of the $LaCl_3$:Ce detector was measured over 272–8579 keV energy using 570, 1064 and 1770 keV gamma rays from ^{207}Bi source along with 2223 keV hydrogen capture peaks during prompt gamma rays studies. The activation spectrum of the $LaCl_3$:Ce detector, namely La(272), La(4502), Cl(1165), Cl(2470), Cl(2864), Cl(6111), Cl(6620), Cl(6978), Cl(7790) and Cl(8579) peaks were also used for the measurement. For 272–8579 keV gamma rays, the energy resolution of the

Table 2 Ratios of Energy resolution (@661 keV) and ***a*** Coefficients from Energy Resolution Fit of $CeBr_3$, $LaCl_3$:Ce and $LaBr_3$:Ce detectors

Detector ratio[a]	Energy resolution ratio for 661 keV gamma rays	Ratio of corresponding a coefficients of the fit
$CeBr_3$ (50 mm)/$CeBr_3$(76 mm)	1	0.830
$CeBr_3$(76 mm)/$LaBr_3$Ce(50 mm)	1.517	1.333
$CeBr_3$(76 mm)/$LaCl_3$:Ce (76 mm)	1.071	1.066
$LaCl_3$:Ce (76 mm)/$LaBr_3$:Ce (100 mm)	0.774	0.701

[a] Detector is marked by its diameter (× mm) assuming a cylindrical (× mm, × mm) detector

$LaCl_3$:Ce detector varies from 7.09 ± 0.02 to 0.90 ± 0.01 %. In Fig. 10, the energy resolution data of the $LaCl_3$:Ce detector is plotted as a function of gamma ray energy. Also in Fig. 10, the energy resolution data is fitted with a least square fit of type $\Delta E/E\ (\%) = a/E^b$, where a and b are constants and E is the gamma ray energy in keV. The fit is drawn with a solid line.

The values of the constants a and b of the fit to the measured energy resolution data of the $LaCl_3$:Ce detector are listed in Table 1. Furthermore, the energy resolution data of the 50 × 50 mm (height × diameter) $CeBr_3$ and $LaBr_3$:Ce detectors [11] has been included in Table 1 for comparison with the present energy resolution data of the 76 × 76 mm (diameter × height) $CeBr_3$ and $LaCl_3$:Ce detectors. Also, the energy resolution data of a large 100 × 100 mm $LaBr_3$:Ce [3] detector has been included in Table 1.

As expected, the fit coefficient b for both types of detectors has a value of about 0.5. The value of the coefficient a shows dependence on detector volume and energy resolution at 661 keV. For detectors of the same type its value increases with detector volume. For detectors of different types but with the same volume, its value is proportional to the energy resolution of the detector for 661 keV gamma rays.

A comparison of the coefficient a values of the energy resolution fit to the data of the 50 × 50 mm $CeBr_3$ detector [11] and that of the 76 × 76 mm $CeBr_3$ detector of the present study shows a 20 % larger value of the coefficient a for our 76 × 76 mm detector. The larger volume of the 76 × 76 mm $CeBr_3$ detector used in the present study might have resulted in increasing light absorption with increasing Ce contents and hence poorer light collection. Table 2 shows the energy resolution ratios of the $CeBr_3$, $LaBr_3$:Ce and $LaCl_3$:Ce detectors of various sizes for 661 keV energies and ratios of corresponding a coefficients of the fits to the respective detector's energy resolution data.

As expected, the detector's a-coefficient ratios are in good agreement with the detector energy resolution ratio for 661 keV gamma rays. The disagreement between the two a values is only in the case when the two detectors under comparison have different volumes.

Conclusions

In this study, the energy resolution of 76 × 76 mm $CeBr_3$ and $LaCl_3$:Ce detectors were measured using gamma rays produced due to capture of thermal neutrons in elements present in the $CeBr_3$ and $LaCl_3$:Ce detectors material. The measured energy resolution of the $CeBr_3$ detector for 368–2223 keV gamma rays varies from 6.64 ± 0.04 to 2.75 ± 0.04 %. For the $LaCl_3$:Ce detector, the energy resolution for 272–8579 keV gamma rays varies from 7.09 ± 0.02 to 0.90 ± 0.01 %. The energy resolution data of both detectors has been fitted with a function of type $\Delta E/E\ (\%) = a/E^b$. For both types of the detectors, the fit coefficient b has a value of about 0.5. The value of coefficient a is very sensitive to detector volume and its energy resolution. This study has provided useful data about the energy resolution and activation spectra of $CeBr_3$ and $LaCl_3$:Ce detectors.

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