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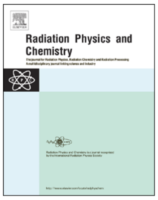

Investigation of $LaBr_3$:Ce probe for gamma-ray spectroscopy and dosimetry

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HIGHLIGHTS

- Development of a radiation detector.
- Monte-Carlo simulation of the $LaBr_3$:Ce detector.
- Use of the $LaBr_3$:Ce detector and comparison to other detectors.

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ABSTRACT

The main thrust of this work is the investigation of performance of relatively new commercial $LaBr_3$:Ce probe (Inspector 1000™ with $LaBr_3$:Ce crystal) for gamma-ray spectroscopy and dosimetry measurements in comparison to $LaCl_3$:Ce and NaI:Tl scintillators. The crystals were irradiated by a wide range of energies (^{57}Co, ^{22}Na, ^{18}F, ^{137}Cs and ^{60}Co). The study involved recording of detected spectra and measurement of energy resolution, photopeak efficiency, internal radioactivity measurements as well as dose rate. The Monte Carlo package, Geant4 Application for Tomographic Emission (GATE) was used to validate the experiments. Overall results showed very good agreement between the measurements and the simulations. The $LaBr_3$:Ce crystal has excellent energy resolution, energy resolutions of $(3.37 \pm 0.05)\%$ and $(2.98 \pm 0.07)\%$ for a ^{137}Cs 662 keV and a ^{60}Co 1332 keV gamma-ray point sources respectively, were recorded. The disadvantage of the lanthanum halide scintillators is their internal radioactivity. Inspector 1000™ with $LaBr_3$:Ce scintillator has shown an accurate and quick dose measurements at Positron Emission Tomography (PET) Units which allows accurate assessment of the radiation dose received by staff members compared to the use of electronic personal dosimeters (EPD).

1. Introduction

There has been considerable research and development of inorganic scintillators for gamma spectroscopy and dosimetry applications over the past several decades and the search for the ideal scintillator is intensifying (Eijk, 2002). The ideal scintillator for such applications should have high light output (for good energy resolution and intrinsic spatial resolution), high density (>3.5 g/cm^3), an emission wavelength well matched to photomultiplier tube readout (300–500 nm), short decay time ($< \mu s$) and off course cost-effective (Moses and Shah, 2005).

Lately cerium-doped lanthanum crystals, in particular $LaBr_3$:Ce, have drawn significant interest due to their high scintillation yields and superior energy resolutions which make them to have the potential to replace NaI:Tl as the material of choice for gamma spectroscopy applications.

Radiation exposure of technical staff operating PET units with the use of ^{18}F has been controversial area of research due to the high energy of the photons (511 keV) as well as sharp increase in the number of scans with limited number of PET/CT facilitates

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(Alsafi et al., 2007. Dalianis et al., 2006). Hence, accurate dose measurements are essential optimising PET/CT unit designed to reduce staff exposure.

The main thrust of this work is the investigation of performance of relatively new commercial $LaBr_3$:Ce probe (Inspector 1000™ with $LaBr_3$:Ce crystal) for gamma-ray spectroscopy and dosimetry measurements. In addition, this work is aimed to compare detection properties of $LaBr_3$:Ce scintillator to NaI(Tl). The detector characterisation involved energy spectra, energy resolution, photo-peak efficiency, internal radioactivity measurement as well as dose rate.

2. Materials and methods

For this study, the Canberra Inspector 1000™ Digital Hand-Held Multichannel Analyser with the IPROL-1 Intelligent Probe with sourceless stabilisation was used. The detector dimensions are 19.0 cm × 16.5 cm × 6.4 cm with a weight of less than 2.4 kg. Canberra Inspector 1000™ probe contains a $\emptyset 38.0 \times 38.0\ mm^2$ $LaBr_3$ scintillation crystal.

A $LaCl_3$ scintillator, doped with 10% of cerium, was supplied by Saint-Gobain™ (BrilLanCeTM, 2012). The $LaCl_3$ crystals are directly coupled to Hamamatsu R6231 photomultiplier tube (PMT). The size and operating voltages for $LaCl_3$ detector were as follows: $\emptyset 44.4 \times 50.8$ mm and 737 V respectively. For the NaI(Tl) measurements, an NaI(Tl) crystal, doped with approximately 0.5% of thallium and directly coupled to PMT (SCIONX 51 B 51/2), was used. The size and operating voltages for the NaI(Tl) detector were $\emptyset 50.8 \times 50.8\ mm^2$ and 540 V respectively.

The crystals were irradiated by a wide range of γ-ray point sources (^{241}Am, ^{57}Co, ^{133}Ba, ^{22}Na, ^{137}Cs and ^{60}Co). These isotopes cover the energy range of approximately 60–1332 keV. Gamma-ray spectra for each source were recorded with these crystals under identical operation conditions, such as acquisition time, a source-to-detector surface distance, surrounding shielding media, shaping time and amplification settings.

In this study, the Monte Carlo package, Geant4 Application for Tomographic Emission (GATE Version 4.0.0) was used to validate the experiments. The $LaBr_3$:Ce (5%), $LaCl_3$:Ce and NaI(Tl) crystals mentioned above, were simulated. GATE allows the modelling of a so-called back-compartment to account for the photomultipliers and electronics located behind the crystal. Hence, for all three detectors, a back-compartment was modelled as a 10 cm Perspex layer (density 2.5 g/cm^3). For a more accurate representation of reality, an aluminium casing was simulated for each detector.

In all the simulations carried out in this study, the point sources were modelled as isotropic emitters with a wide range of energies representing ^{241}Am, ^{57}Co, ^{22}Na, ^{137}Cs and ^{60}Co sources. The physics processes were modelled using the low energy electromagnetic processes package including Rayleigh, photoelectric and Compton interactions.

3. Results and discussion

3.1. Recorded spectra and energy resolution

Fig. 1 shows the measured energy resolution with the gains set to encompass all energies of interest (i.e. up to ≈ 1500 keV). Except for low energies, the energy resolution figures achieved for the lanthanum based-crystals are twice as good as that of NaI(Tl). For instance, for a ^{137}Cs 662 keV γ-ray point source, energy resolutions of (3.05 ± 0.03)%, (3.75 ± 0.03)% and (6.85 ± 0.03) % were achieved for $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) respectively at room temperature.

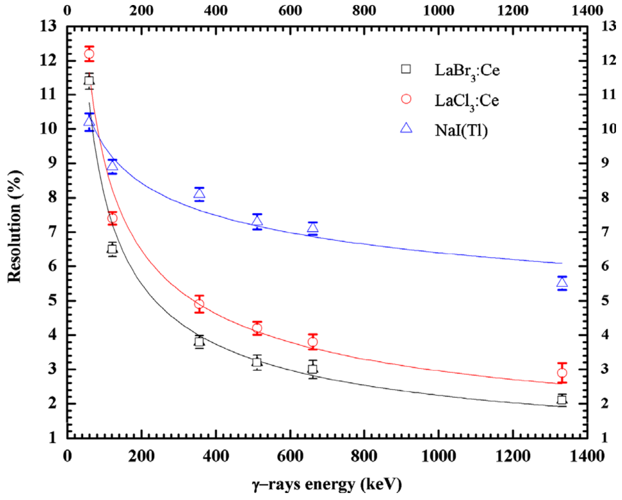

Fig. 1. Measured energy resolution of $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) scintillators versus γ-ray energy.

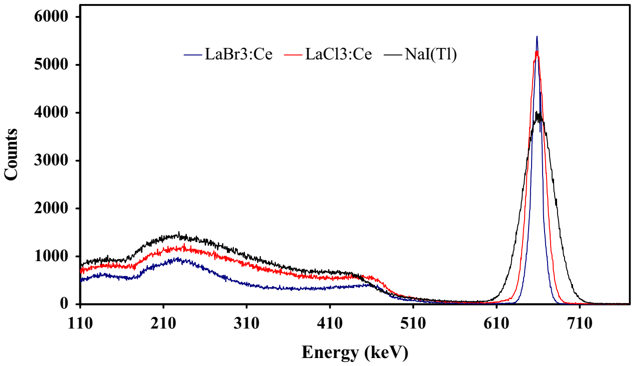

Fig. 2. Comparison of the energy spectra of 662 keV gamma-rays from a ^{137}Cs source, as measured with $LaCl_3$:Ce and NaI(Tl) crystals.

The superiority of the energy resolution of the lanthanum based-crystals is also seen in a comparison of the measured energy spectra of 662 keV γ-rays from a ^{137}Cs source, as shown in Fig. 2. The measured energy resolutions of (3.05 ± 0.03)% and (3.75 ± 0.03)% of 662 keV γ-rays are among the best energy resolution obtained for existing inorganic scintillator crystals (Shah et al., 2004; Eijk, 2001). Fig. 3 shows a comparison of simulated energy spectra of a ^{133}Ba point source obtained from the $LaBr_3$:Ce(5%) and NaI(Tl) detectors demonstrating the superiority of energy resolution of the cerium-doped lanthanum crystals at wide range of low energies (< 400 keV).

It can be deduced from Fig. 1 that the resolution of $LaBr_3$:Ce and $LaCl_3$:Ce scintillator is poorer than that of NaI(Tl) at low energies (< 100 keV). This could be due to the fact that NaI(Tl) has a significantly larger light output below approximately 100 keV (Milbrath et al., 2007).

3.2. Coincidence timing resolution

Timing resolution was measured using the BaF_2 scintillator which is considered to be a benchmark in fast time experiments due to its high decay time (0.8 ns). The BaF_2 detector was operated at −2450 V. The BaF_2 detector formed a "START" channel in the timing circuit, while the $LaBr_3$:Ce detector formed the "STOP" channel. Upon irradiation of each crystal with 511 keV γ-rays from a ^{22}Na point source, the signal from each detector was processed using two channels of a Constant Fraction Discriminator (CFD). The

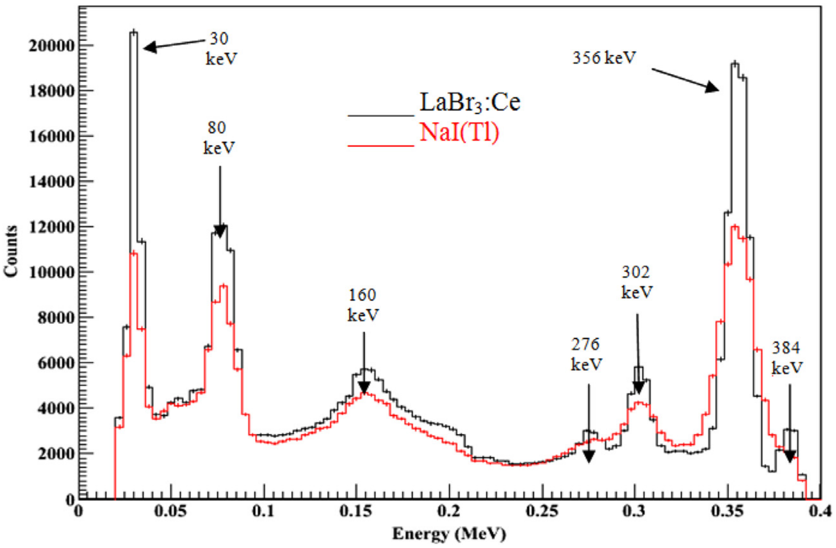

Fig. 3. Comparison of simulated energy spectra of a ^{133}Ba point source obtained from the LaBr$_3$:Ce and NaI(Tl) detectors.

time difference between the “START” and “STOP” signals was digitised with a time-to-amplitude converter (TAC).

The time resolution of the system is conventionally defined as the FWHM of the prompt coincidence peak. The FWHM of the timing spectrum was found to be (332 ± 7.4), (412 ± 6.2) and (1764 ± 12.7) ps for the LaBr$_3$:Ce, LaCl$_3$:Ce and NaI(Tl) detectors respectively. Similar results are reported (320 ps by Nicolini et al., 2007) for the LaBr$_3$:Ce and (264 ps by Shah et al., 2003) for the LaCl$_3$:Ce. Differences in timing resolution from those reported in the literature could be due to differences in measurement conditions such as the distance between the pairs of detector. The excellent timing resolution of LaBr$_3$:Ce, LaCl$_3$:Ce is due to its combination of fast decay and high light output (i.e. approximately 10 times higher than NaI(Tl)).

3.3. *Peak-to-valley ratio*

The energy resolution spectrum of a ^{137}Cs point source was used for this measurement because it is mono-energetic. The positions of the channels are determined by the resolution of the main full energy photopeak energy. The valley is calculated as the average count at the following channels: (1) a distance of 0.5 FWHM from the peak centre channel; (2) a distance of 2 FWHM from the peak centre channel; and (3) a distance of 5 FWHM from the peak centre channel. The associated uncertainties with peak-to-valley ratio results were calculated using error propagation analysis.

The peak-to-valley ratio results are 16.4 ± 0.10, 14.7 ± 0.05 and 8.7 ± 0.04 for the LaBr$_3$:Ce, LaCl$_3$:Ce and NaI(Tl), respectively.

3.4. *Internal radioactivity measurement*

The internal radioactivity spectrum was easily measured by self-counting in a very well shielded chamber for approximately 2 days. The internal radioactivity is due to naturally occurring radioisotopes ^{138}La and ^{227}Ac (Firestone and Ekstrom, (2004)). The main features of this curve are β continuum below 300 keV, sum peak from ^{138}Ce (789 keV + β continuum), and 1436 + 32 keV sum peak from ^{138}Ba. There are strong Ba X-rays from 31–38 keV, but they are not seen due to the discriminator level settings to enable viewing at 1436 keV. Nevertheless, the internal radioactivity drawback is likely to be serious only for the very long times of counting and in low activity measurement applications. Furthermore, energy calibration for the detector itself can be done using its internal radioactivity.

Table 1
Dose rate values using the LaBr$_3$:Ce probe, EPD and GATE.

Dose rate (μSv/h)			
Distance (cm)	Inspector 1000^{TM} with LaBr3:Ce	GATE	EPD
25	9.82 ± 0.73	10.20 ± 0.54	11.80 ± 1.12
50	3.20 ± 0.41	2.83 ± 0.62	4.27 ± 1.06
100	0.67 ± 0.34	0.73 ± 0.19	1.07 ± 0.87

3.5. *Dose rate*

In order to investigate the potential use of the Canberra Inspector 1000^{TM} LaBr$_3$:Ce probe for dosimetry application, the dose rate was directly recorded for activity of ^{18}F point source located at different distances as listed in Table 1. The measured results were compared to measured dose rate from Electron Personal Dosimeter (EPD) as well as the calculated results from the GATE Monte Carlo code. Dose rate results show very good agreement between GATE calculations and experimental measurements with Inspector 1000^{TM}. In general, EPD overestimates the dose rate compared to Inspector 1000^{TM} with LaBr$_3$:Ce crystal.

4. Conclusions

Experimental results on the scintillation properties of the Canberra Inspector 1000^{TM} LaBr$_3$:Ce probe for γ-ray spectroscopy were compared to the relatively new cerium-doped lanthanum crystals, LaCl$_3$:Ce, and conventional scintillator, NaI(Tl). Overall, the measured and simulated results indicate that the lanthanum scintillators are promising, in particular, LaBr$_3$:Ce which shows superior energy resolution above 100 keV. Unfortunately, the lanthanum scintillators have a few drawbacks of their own: internal radioactivity and a relatively low response at low energies. The second part of this research investigated the potential use of the LaBr$_3$:Ce probe for dosimetry applications. In summary, because the relatively new cerium-doped scintillators, particularly $LaBr_3$ have excellent detection properties, they have the potential to be the scintillator of choice in γ-ray spectroscopy and dosimetry measurements.

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