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Article *in* IEEE Transactions on Nuclear Science · January 2007

DOI: 10.1109/TNS.2006.885385 · Source: IEEE Xplore

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Comparison of a $LaBr_3$(Ce) Scintillation Detector With a Large Volume CdZnTe Detector

Agnieszka Syntfeld, *Associate Member, IEEE*, Rolf Arlt, Vladimir Gostilo, Alexander Loupilov, Marek Moszyński, *Fellow, IEEE*, Antoni Nassalski, Martha Swoboda, and Dariusz Wolski

***Abstract*—The performance of hand-held radioisotope identification devices (RIDs) is still hampered by the performance of the NaI(Tl) detectors, which are commonly used in such instruments. In this paper, we continue the search for better detector options. One of the largest single elements ever made, a coplanar CdZnTe (CZT) detector ($30 \times 15 \times 12.1$ mm^3 volume 5.45 cm^3 designed by University of Michigan) is compared with a commercially available $LaBr_3$(Ce) detector ($\oslash 1'' \times 1''$ volume 12.9 cm^3.) Parameters that are relevant to the performance of isotope identification devices, such as resolution and efficiency as function of the γ-ray energy, temperature shift, linearity and others are measured and compared. According to measurement results, it seems that for this application $LaBr_3$(Ce) detectors are a viable alternative to CZT detectors; even more so if one bears in mind that $LaBr_3$(Ce) became commercially available only recently and detectors with larger volumes are likely to appear in the near future.**

***Index Terms*—Border monitoring, CdZnTe, $LaBr_3$(Ce), room temperature semiconductor detectors, scintillators.**

I. Introduction

GAMMA spectrometry is the only method that allows for determining, under field conditions, the isotope that had caused an alarm of the border monitor or radiation pager. The ideal detectors for the determination of gamma energies and intensities are high-purity germanium detectors (HPGe), which, however, require liquid nitrogen cooling. Therefore, the market for field-usable instruments is still dominated by the use of the classic NaI(Tl) scintillation detector. These detectors have a typical energy resolution of about 7.5% at 662 keV (^{137}Cs) and show a considerable nonlinearity of the energy scale and a significant temperature drift. For several decades, the only alternative detectors option that could be used at room temperature were CdZnTe (CZT) detectors. In [1] and [2] their performance was compared with various scintillation detectors, with the result that CZT detectors provide the best resolution, compared to all other options. Recently, however, $LaBr_3$(Ce) scintillation detectors became commercially available and the first comparative studies of these detectors with the commonly used NaI(Tl) detectors are presented in [3]. These preliminary results show superior performance of $LaBr_3$(Ce) over NaI(Tl) in RIDs applications. The $LaBr_3$(Ce) scintillators already have a volume of about 13 cm^3 and larger volumes are expected in the future. Their energy resolution of about 3.0%—3.2% for ^{137}Cs [4] comes close to that of large volume CZT detectors.

The CZT detectors exhibit a high energy resolution well below 2% for ^{137}Cs gamma rays; however, this is mainly reported for detectors with small volumes below 1 cm^3 [5]. An application of so-called coplanar grid structures for detector contacts allowed for the production of larger detectors of 1–2 cm^3 with energy resolution of 2% to 4% [6]. Recent studies of a $15 \times 15 \times 10$ mm^3 CZT detector (2.25 cm^3) [7] resulted in a measured energy resolution as good as 1.6% on an energy of 662 keV.

The aim of this paper was to study one of the largest CZT detectors developed to date, with a volume of 5.45 cm^3, in comparison to the $LaBr_3$(Ce) detector (volume 12.9 cm^3). Such large volume CZT detectors are of great importance for their application in the RID devices.

It is worth noting, however, that the studies of CZT detectors of a similar size lead to a rather poor performance concerning energy resolution [8], [9].

Apart from good energy resolution and high efficiency, there are, however, many other parameters that need to be checked to compare the expected performance of both detector options for use in a hand-held isotope identification device.

In this paper we describe the comparison of a commercially available $\oslash 1'' \times 1''$ $LaBr_3$(Ce) detector and a large volume, multi-pair CoPlanar grid CZT (CP CZT) detector ($30 \times 15 \times 12.1$ mm^3) with respect to their use in hand-held isotope identification devices. We use the following criteria for the evaluation:

- Energy resolution as a function of γ-ray energy;
- Peak shape;
- Relative gamma detection efficiency as a function of energy;
- Low energy threshold;
- Linearity of the energy scale;
- Peak shift as a function of temperature;
- Power requirements;
- Size of the detector assembly;
- Ruggedness;
- Price.

II. Experimental Details

The $LaBr_3$(Ce) detector and CP CZT room temperature semiconductor-based detector were studied.

Manuscript received February 16, 2006; revised August 7, 2006. This work was supported in part by the International Atomic Energy Agency under the Coordinated Research Project "The Improvement of Technical Measures to Detect and Respond to Illicit Trafficking of Nuclear and other Radioactive Materials" Research Contract 12596/Nuclear Security Multi-donors Fund, and by the Polish Committee for Scientific Research Grant 8 T10C 002 20 and SPUB 621/E-78/SPB/IAEA/0-13/DWM21/2004-2006.

A. Syntfeld, M. Moszyński, A. Nassalski, and D. Wolski are with the Soltan Institute for Nuclear Studies (SINS), PL-05-400 Świerk-Otwock, Poland (e-mail: syntfeld@ipj.gov.pl).

R. Arlt and M. Swoboda are with the International Atomic Energy Agency (IAEA), Wagramer Strasse 5, 1400 Vienna, Austria (e-mail: r.arlt@iaea.org).

V. Gostilo and A. Loupilov are with the Baltic Scientific Instruments (BSI), LV-1005 Riga, Latvia (e-mail: bsi@bsi.lv).

Digital Object Identifier 10.1109/TNS.2006.885385

The $LaBr_3$ (5%Ce) crystal $\oslash 1'' \times 1''$ in size was grown and cut by the crystals and detectors division of Saint Gobain, France. The scintillator was surrounded with reflecting material and encased in an aluminium container that was hermetically sealed with a glass window.

The scintillator was coupled to a calibrated 50-mm diameter Photonis XP5212 photomultiplier (PMT) with a photocathode blue sensitivity of 12.2 μA/LmF. Silicon grease was used for optical coupling between the crystal and the PMT window. The anode signal was preamplified using an ORTEC 113 unit.

Then the pulse was shaped at 2 μs by an internal amplifier built in the miniature multichannel analyzer (MMCA) [10] or by an external amplifier Tenelec TC244. In the latter case, the 3 μs shaping time constant was used in most of the measurements.

The following set of test sources was used:

- Standard calibration sources including ^{137}Cs, ^{152}Eu, ^{60}Co, ^{133}Ba, ^{241}Am, ^{57}Co, ^{22}Na, ^{93}Mo, ^{51}Cr, ^{207}Bi, ^{54}Mn, ^{203}Hg.
- Naturally occurring radioactive materials (NORM) ^{226}Ra, ^{232}Th, ^{238}U.
- Nuclear materials (NM) such as a 2.6 kg depleted uranium (DU) cube, CBNM plutonium standards (6.6 g PuO_2), and uranium samples with different enrichments.

Energy spectra were recorded by the data acquisition systems of MMCA and TUKAN8K [11] in the IAEA, Austria, and SINS, Poland, respectively.

The spectra were analyzed using the TUKAN8K analysis module, where all photopeaks were fitted with Gaussian functions.

Measurements of fissile nuclear materials were carried out in the Safeguards Instrumentation Laboratory (SIL) of the IAEA in Seibersdorf, Austria.

Fabrication and room temperature tests of the CP CZT detector ($30 \times 15 \times 12.1$ mm^3 volume 5.45 cm^3) were performed at the BSI, Latvia. The crystal of initial dimensions $30.5 \times 32.9 \times 17.3$ mm^3 was cut from the ingot CZT3-18 grown by Yinnel Tech. with purity grade P-2 of the source material. The multipair coplanar grid structure was formed on both sides of the crystal. The contact topology used in this paper is presented in more detail in [8]. The crystal inspection tests resulted in nonuniformity of the material and a significant amount of surface defects. During the fabrication, the detector was remade four times to obtain better performance. Finally, the crystal thickness was reduced from 17.3 to 12.1 mm. The best performance was obtained for the detector with a size of $30 \times 15 \times 12.1$ mm^3 where two independent grids (two quadrants) were employed (see the results obtained by Gostilo *et al.* in [9]). The evaluated registration efficiency of the CP CZT detector was about 11% for irradiation from the cathode. The optimal voltage supplied to the cathode was -1700 V and $+30$ V on grids, the shaping time constant being 3 μs.

Besides the detector, the Detector Probe also contained a thermoelectric cooler (TEC) for detector temperature stabilization, a two-channel differential charge sensitive preamplifier with resistive feed back, the detector's thermostabilisation device, an HV filter and the HV power supply with voltage divider.

III. Results and Discussion

This section is divided into several subsections. First, characteristics such as the number of photoelectrons produced per 1-MeV energy deposited in the crystal, absolute light yield, and the nonproportional response of $LaBr_3$(Ce) versus γ-ray energy are described. Next, the ^{137}Cs spectra are demonstrated, showing an energy resolution of the detectors as well as good peak-to-Compton and peak-to-valley ratios for both detectors. Energy resolutions of the $LaBr_3$(Ce) and the CP CZT as a function of γ-ray energy, measured with standard calibration sources, are presented compared to the reference NaI(Tl) crystal. Then relative photopeak efficiencies for both $LaBr_3$(Ce) and CP CZT detectors measured with ^{226} Ra are illustrated followed by sections concerning the long term and temperature stability measurements. Finally, aspects of the high energy resolution of the detectors in a NORM and NM identification are described.

A. Light Output and Nonproportionality

The $LaBr_3$(Ce) crystals seem to be the most efficient scintillators compared to all recently available ones on the market. In this paper, the photoelectron yield N_{phe} per 1-MeV γ deposited in the crystal volume was measured on the basis of single photoelectron and ^{137}Cs spectra. The N_{phe} value is defined as the ratio between the position of the given γ-ray fully-energy peak and the position of the single photoelectron peak. The number of photoelectrons was equal to $18\,000 \pm 500$ phe/MeV with a shaping time constant of 3 μs as measured by the XP5212 PMT. At the quantum efficiency of the photocathode, about 30% at 360 nm, the absolute light output of $60\,000 \pm 6\,000$ photons per MeV was obtained. This is significantly larger than the yield of 38 000 ph/MeV found for the NaI(Tl) scintillator (e.g., [12]).

The light yield as a function of γ and X-ray energy relative to the yield at 662 keV is presented in Fig. 1. The nonproportional response of $LaBr_3$(Ce) is compared to that of NaI(Tl). For the energies above 200 keV, both scintillator responses are quite proportional; whereas in the low energy region, the $LaBr_3$(Ce) detector is more proportional than the NaI(Tl) detector. In contrast to NaI(Tl), the $LaBr_3$(Ce) nonproportionality curve bends down when energy decreases, and the light production is much lower compared to NaI(Tl) (see Fig. 1). The nonproportionality curve presented in Fig. 1 for the $LaBr_3$(Ce) detector is in agreement with the nonproportionality reported by Dorenbos *et al.* [4]. However, one should be careful in determination of the light yield in the high energy region (above $\sim$1 MeV), owing to a possible nonlinearity of the PM tube.

Independent nonlinearity measurements of $LaBr_3$(Ce) and CP CZT were performed in parallel. The results of measurements of the nonlinear response of $LaBr_3$(Ce) and CP CZT are presented in Fig. 2. First, the measurements with a ^{226}Ra source emitting γ radiation in a wide energy region, from 186 to 2448 keV, were carried out. The positions of different photopeaks were normalized to the 609–keV line. Fig. 2(b) shows that the nonlinearity for $LaBr_3$(Ce) is on the order of 3% for the whole energy region presented; above 200 keV, it is comparable to that of the large volume CP CZT. The nonlinearity curve of $LaBr_3$(Ce) matches well with the nonproportionality of the crystal shown in Fig. 1. Next, the nonlinearity in the low-energy region (below 200 keV) was determined using the spectra measured with ^{241}Am and/or ^{133}Ba. Unfortunately, in the low energy region Fig. 2(a) a significant deterioration of the linearity

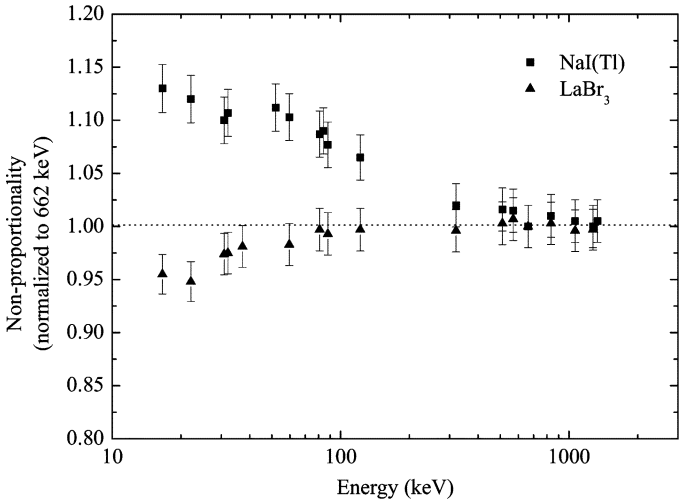

Fig. 1. A comparison of the nonproportionality curves measured for the $LaBr_3$(Ce) and NaI(Tl) ($\oslash 25 \times 31$ mm) crystals, respectively.

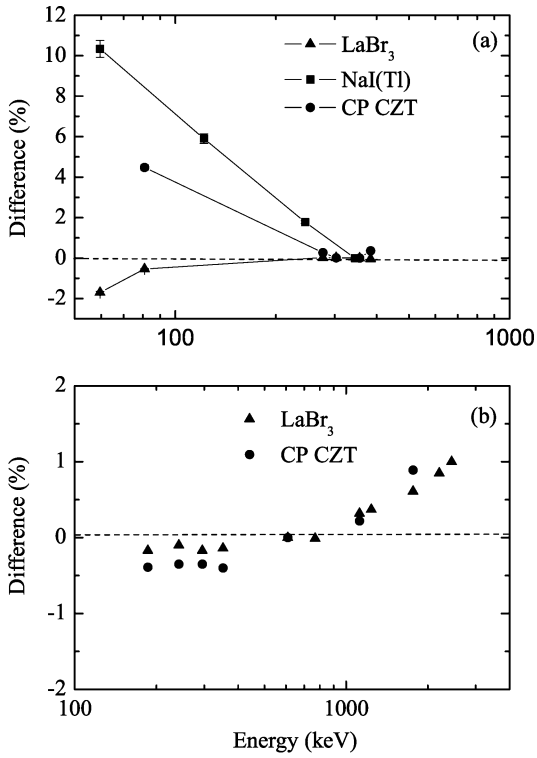

Fig. 2. Deviations from linearity measured with ^{241}Am and ^{133}Ba for the $LaBr_3$(Ce), CP CZT and $\oslash 1'' \times 1''$ NaI(Tl) detectors (a) and with ^{226}Ra source for $LaBr_3$(Ce) and CP CZT detectors (b). The error bars are of the data points size.

for the CP CZT detector was observed. This is probably due to a more efficient charge collection for low energy γ-rays detected in the front area of the studied CP CZT detector. Such a deterioration of the linearity has not been measured for smaller volume CZT detectors.

In conclusion, both $LaBr_3$(Ce) and CP CZT detectors are more linear in the energy scale than common NaI(Tl) crystals used in RIDs. However, the nonlinearity of the CP CZT detector is surprisingly high, especially in the low energy region. The nonlinearity of CP CZT at lower energies together with poor energy resolution (see Section III-B) does not allow highly selective detection of these materials (e.g., nuclear materials) that emit γ-rays clustered in this energy region.

B. Energy Resolution

The measured energy resolution of the full-energy peak registered in a scintillator can be expressed as [13], [14]:

$$(\Delta \mathrm{E}/\mathrm{E})^2 = (\delta_{\mathrm{sc}})^2 + (\delta_{\mathrm{st}})^2 + (\delta_{\mathrm{t}})^2 \quad (1)$$

where δ_{sc} is the intrinsic resolution of the crystal, δ_{st} is the statistical contribution associated with variation of the number of photoelectrons produced at the photocathode, and δ_{t} is the transfer resolution associated with the variation of light and photoelectron collection [14], [15].

The intrinsic resolution is a property characteristic of the crystal itself and follows the crystal nonproportional response to X-rays and γ-rays (see [14] and references therein.) It is believed that other effects, such as inhomogeneities in the scintillator, cause local variations in the scintillation efficiency and nonuniform reflectivity at the surface of the crystal affect the intrinsic resolution.

Assuming Poisson statistics for scintillation light produced in the crystal, the statistical term δ_{st} is expressed as:

$$\delta_{\mathrm{st}} = 2.35 \times 1/\mathrm{N}^{1/2} \times (1+\varepsilon)^{1/2} \quad (2)$$

where N is the number of photoelectrons produced at the photocathode and ε is the variance of the electron multiplier gain, 0.1 for the XP5212 PMT.

The δ_{t} term was not taken into account in the calculations of the intrinsic resolution since its contribution to the overall energy resolution was not estimated in this paper. On the other hand, in modern scintillation detectors the transfer component can be neglected since its value is small compared to the other components of the energy resolution [14].

The overall energy resolution for $LaBr_3$(Ce) crystal connected to a PM tube was measured as 3.2% at 662 keV (see Fig. 3) with the shaping time of 3 μs. In the same figure, the spectrum of ^{137}Cs measured with CP CZT is included and compared to $LaBr_3$(Ce). An energy resolution of 5.1% was measured for the CP CZT detector with the 1 μs shaping time constant. The energy resolution of $LaBr_3$(Ce) is much better than that obtained for the tested CP CZT detector. On the other hand, all smaller volume (up to 2.25 cm^3) CZT detectors had excellent energy resolutions, less than 2% of the energy at 662 keV [7]. For the measured ^{137}Cs spectra the peak-to-Compton and peak-to-valley ratios for $LaBr_3$(Ce) and CP CZT were calculated and are presented in Table I.

Fig. 4 shows an overall energy resolution as a function of γ-ray energy measured for $LaBr_3$(Ce), CP CZT, and NaI(Tl) detectors. The CP CZT exhibits poor energy resolution in the whole energy region (18.7% and 5.1% at 81 and 662 keV, respectively.) The energy resolution measured for $LaBr_3$(Ce) is extremely good for γ-ray energies above 100 keV. The tendency of NaI(Tl) light output to increase while the γ-ray energy goes

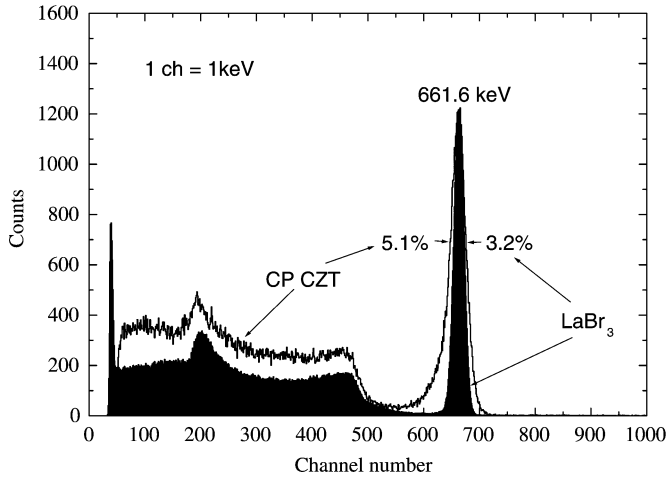

Fig. 3. The γ-ray spectra from a ^{137}Cs source measured with $LaBr_3$(Ce) and CP CZT.

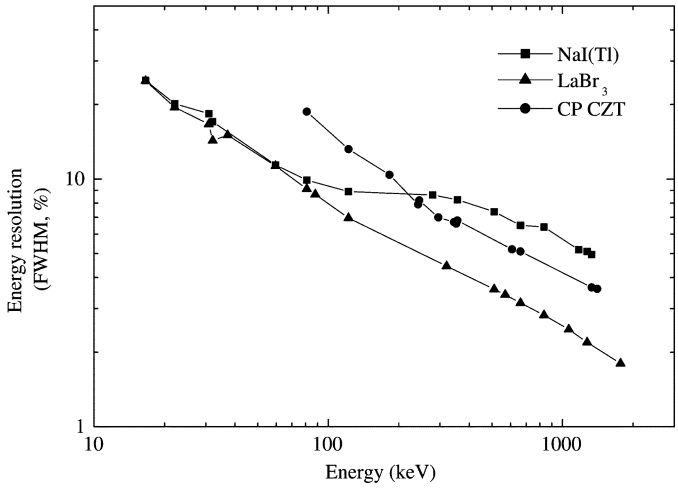

Fig. 4. The overall energy resolution measured for $LaBr_3$(Ce) and CP CZT (compared to $\oslash 25 \times 31$ mm NaI(Tl).)

TABLE I
PROPERTIES OF $LaBr_3$(CE), CP CZT, AND NAI(TL) DETECTORS

Parameter	$LaBr_3$(Ce)	CP CZT	NaI(Tl)
Volume (cm^3)	12.9	5.45	15.2
Area (cm^2)	4.9	4.5	4.9
Density (g/cm^3)	5.3	6.2	3.67
Decay time constant (ns)	18	-	230
Light output (ph/MeV)	60 000	-	38 000[a)]
Effective atomic number	46.9	48.5	50
Energy resolution at 81 keV/662 keV	9.1/3.2	18.7/5.1	9.9/6.5
Intrinsic resolution at 81 keV/662 keV	6.3/2.2	-	5.3/5.7
Temperature drift (%/°C)	0.01[b)]	0.01[c)]	±0.2[b)]
Peak-to-Compton ratio at 662 keV	5.8	5.1	3.2
Peak-to-valley ratio at 662 keV	137	32	40
Natural radioactivity	^{138}La	No	No

a) from Ref. [12],
b) without PMT contribution, from Ref. [16],
c) from Ref. [2].

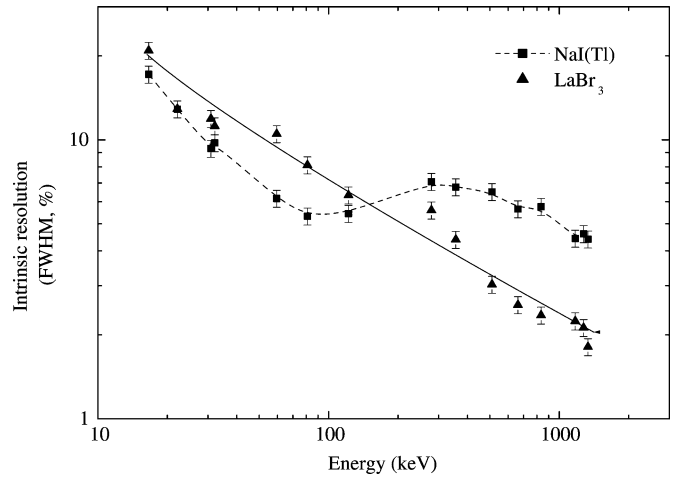

Fig. 5. The intrinsic resolution measured for $LaBr_3$(Ce) and compared to NaI(Tl) ($\oslash 25 \times 31$ mm).

down makes NaI(Tl) still competitive amongst scintillators in the low energy region.

According to (1) and (2), with the transfer term δ_t being neglected, the intrinsic energy resolution was calculated for $LaBr_3$(Ce) and is presented in Fig. 5. The intrinsic contribution to the overall energy resolution is 2.2% at 662 keV. As the γ-ray energy decreases, the intrinsic resolution is strongly increased, thereby deteriorating the overall energy resolution. Thus, it is evident that the nonproportional response at lower energies correlates with the intrinsic resolution of the crystal.

C. Photopeak Efficiency

In efficiency measurements, the $LaBr_3$(Ce) and CP CZT detectors were fixed on a measurement bench at the same distance from a source. The CP CZT detector was irradiated from the cathode and, in the case of the $LaBr_3$(Ce) detector, the 1''-diameter entrance surface was exposed to the source. The relative efficiency (slope) presented in Fig. 6 was measured with a weak ^{226}Ra source that directly touched the entrance surface of a detector. The points were normalized to the strongest 609–keV line in ^{226}Ra. Photopeak areas and absolute intensities of γ-lines from ^{226}Ra were incorporated into the calculations. Fig. 6 shows that photopeak efficiencies in the low energy region, compared to 609–keV efficiency, are higher for the CP CZT detector than for the $LaBr_3$(Ce) detector. Therefore the CP CZT detector exceeds the $LaBr_3$(Ce) detector with respect to the detection of nuclear materials.

One should note that the efficiency depends on geometric factors such as volume and area of the front side of the crystal. The relative efficiencies calculated in this paper were not corrected for the geometric factors.

D. Long-Term Stability

Long-term stability was measured for $LaBr_3$(Ce) for about 15 h with a ^{232}Th source. The observed instability was -0.6% for the $LaBr_3$(Ce) detector, assuming that the instability is mainly determined by the detector/preamplifier and the contribution of the MMCA to the long-term drift is negligible.

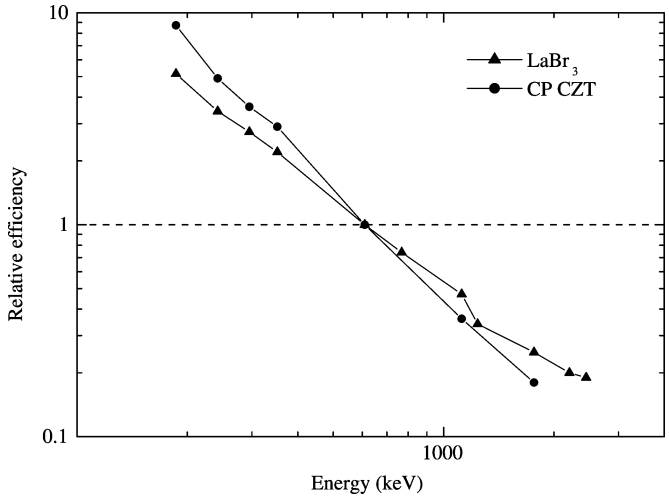

Fig. 6. Relative (to 609 keV) efficiency versus γ-ray energy determined for the $LaBr_3$(Ce) and compared to NaI(Tl) ($\varnothing 25 \times 31$ mm).

Additional measurement of the $LaBr_3$(Ce) time stability was performed using ^{226}Ra and an external Tenelec TC244 amplifier. The strongest γ-lines were marked and their positions were checked every hour. The measurement lasted 15 h and the observed relative drift of the marked full-energy peaks was less than about $+0.5\%$.

The long-term drift of CZT detectors is negligible as it was reported by Arlt *et al.* [1].

E. Temperature Drift

The $LaBr_3$(Ce) Detector Probe (crystal coupled to PMT) was placed into an environmental chamber. The temperature was slowly changed. The extreme points were -10 and $+40$ °C. The 662-keV peak drift was observed between these extreme temperatures. Its relative value was about -0.2% per degree. This drift is mainly due to the PMT instability with changing temperature. Independent measurements of the temperature stability were performed at Target GmbH, Solingen [16] demonstrated very stable intrinsic light output of the $LaBr_3$(Ce) crystal in the temperature range from -30 to $+60$ °C. High thermal stability of the light output of $LaBr_3$(Ce) of about $+0.01\%/$C was found after correction for thermal dependences of the PMT gain and the quantum efficiency of the photocathode [16].

CZT detectors are more stable with respect to ambient temperature changes. Typically, the temperature drift is as low as 0.01% per degree (see [2]).

F. Identification of NORM and NM

Both $LaBr_3$(Ce) and CP CZT detectors are considered here as the detectors dedicated for border monitoring and shipment container survey. CP CZT has already been used in RIDs (see [10]). Due to the low efficiency and high price of CP CZT, we searched the market for a new crystal that would operate at room temperatures. $LaBr_3$(Ce) scintillators met the requirements as the detectors in hand-held radioisotope identifiers. Its high light output (high scintillation efficiency), extreme energy resolution, good linearity in the energy scale and lower price makes this crystal competitive with CP CZT. High energy resolution assures proper identification of a sample; whereas high efficiency allows shorter detection time and faster recognition of illegal sources being trafficked.

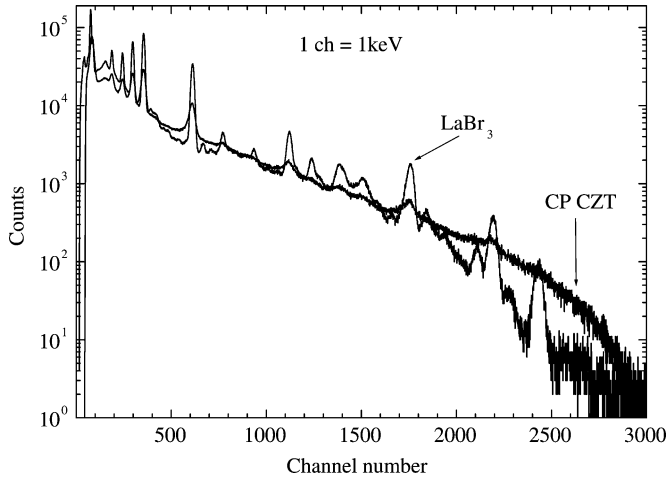

Fig. 7. The ^{226}Ra spectra taken for the $LaBr_3$(Ce) and CP CZT detectors.

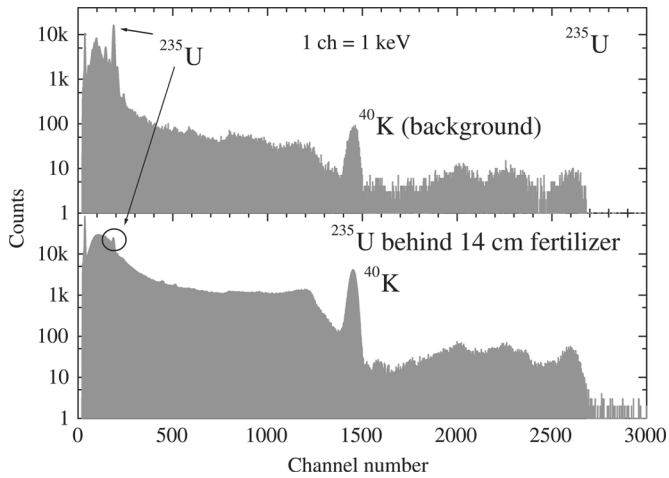

Fig. 8. The ^{235}U spectra taken for the $LaBr_3$(Ce) detector.

Fig. 7 presents a comparison of γ spectra of the ^{226}Ra NORM-type source measured for both detectors. Much better performance of $LaBr_3$(Ce) over the large volume CP CZT allows more efficient detection and higher selectivity in identification of materials emitting γ-rays in the wide energy region. Fig. 7 also shows that the CP CZT detector fails when detecting higher energy γ-rays due to poor energy resolution and worse peak-to-total ratio (photofraction.) This may also suggest a further deterioration of energy resolution because of the detection of secondary gamma rays that create the full energy peaks (Compton scattered and 511-keV annihilation quanta produced in the stopping process by the pair production) in the whole volume of the detector.

Fig. 8 shows γ spectra for the enriched ^{235}U. The lower spectrum shows data for the ^{235}U source behind fertilizer, 14 cm in thickness, taken with the $LaBr_3$(Ce) detector. Since ^{235}U emits γ-radiation mainly in the low energy region, good energy resolution is crucial to γ-ray separation and identification in this energy region. The 186–keV peak is still observed even if the ^{235}U sample is masked by the natural radioactive shield.

IV. Summary

In this paper, the comparison between a $LaBr_3$(Ce) scintillation detector and a large volume coplanar grid CZT detector is presented. The studies were prompted by a search for a new detector with a performance best fitted to RIDs requirements. Commonly exploited NaI(Tl) scintillators are characterized by a poor energy resolution and temperature instability.

Presently, the new $LaBr_3$(Ce) scintillation crystals represent a compromise between NaI(Tl) and CZT. Recent samples of a volume of about 13 cm^3 can register γ radiation with an energy resolution as high as 3.2% at 662 keV, somewhat worse than that of 1.6% for CP CZT (volume 2.25 cm^3), and much better than 6.5% for the selected NaI(Tl) samples. In the low energy region (<100 keV) $LaBr_3$(Ce) has poorer energy resolution and is comparable to NaI(Tl). On the other hand, $LaBr_3$(Ce) is more linear in the whole energy region. The high detection efficiency of $LaBr_3$(Ce) scintillators is due to their high density and recently available larger crystals.

Unfortunately, CZT detectors of comparable volume are still not an alternative to $LaBr_3$ because of lower energy resolution in the whole range of gamma ray energies. Moreover, even larger degradation of the energy resolution seems to be observed at energies above 1 MeV. This is a conclusion of the study of one of the first developed detectors of such a large size. Its performance is no doubt limited by the quality of the crystal which is very difficult to grow to such a large volume. Smaller detectors of 2.25 cm^3 previously showed good performance. More efforts are necessary to further develop large-volume CZT detectors up to 10 cm^3, with an energy resolution similar to that of smaller ones.

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