New Scintillators for the Border Monitoring Equipment

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Performance of new scintillators characterized by a high energy resolution (selectivity), as LaCl$_3$:Ce and LaBr$_3$:Ce, and high detection efficiency (sensitivity), as CdWO$_4$ and CaWO$_4$, are discussed. Particularly, the properties of LaBr$_3$ crystal are presented in details.

1. Introduction

At present there is a continuous need for highly efficient and good energy resolution (selectivity) detectors to use them in the border monitoring instrumentation to fight illicit trafficking of nuclear materials [1]. There are at least two major technical obstacles: sensitivity and selectivity. The sensitivity is of great importance parameter, which allows finding nuclear material, while the selectivity is required to avoid a misinterpretation of the benign radioactivity as being a threat. As of now, the small hand-held instrumentation utilize scintillation detectors NaI(Tl) and semiconductor detectors CZT. The former one is characterized by a very high detection efficiency but poor energy resolution, while the latter is of better selectivity but lower efficiency due to its small size.

New scintillation detectors are needed to significantly improve current border monitoring instrumentation. Particularly, new scintillators as LaCl$_3$, LaBr$_3$, CdWO$_4$ (CWO), and CaWO$_4$ seem to be good candidates and their advantages and drawbacks should be evaluated. High energy resolution (selectivity) of LaCl$_3$ and LaBr$_3$ and the high detection efficiency of CWO and CaWO$_4$ for high-energy gamma rays can improve the border monitoring. The performances of new scintillators are discussed below with a special emphasis to the LaBr$_3$ crystal.

2. Scintillators with a High Selectivity

Recently new chloride and bromide compounds, as LaCl$_3$:Ce [2] and LaBr$_3$:Ce [3], showed both a very high light output and good energy resolution. The value of 2.9% measured for 662 keV $\gamma$-rays from $^{137}$Cs with LaBr$_3$ scintillator is unequalled [3]. A good density of both crystals equal to 3.86 g/cm$^3$ for LaCl$_3$ and 5.3 g/cm$^3$ for LaBr$_3$ assure a comparable or better sensitivity.

![Figure 1: The comparison of energy spectra of 662 keV $\gamma$-rays from a $^{137}$Cs source, as measured with LaCl$_3$ and NaI(Tl) crystals under the same gain of spectroscopy amplifier.](image-url)
than that of NaI(Tl). Fast light pulses with the decay time constant of about 25 ns and 18 ns, respectively, allow for high counting rate measurements.

2.1. LaCl$_3$:Ce Crystal

The LaCl$_3$:Ce crystal was proposed first by van Loef et al [2]. In the study [4], the commercial sample of LaCl$_3$: (9 ± 1)%Ce with size of 25 mm was compared to NaI(Tl). A good light output of 9400 ± 100 photoelectrons per MeV and energy resolution of 4.2 ± 0.2% for 662 keV $\gamma$-rays were measured with the LaCl$_3$ crystal coupled to the XP3212 photomultiplier with bialkali photocathode. Below 122 keV, the energy resolution of LaCl$_3$ was unexpectedly worse than that of NaI(Tl).

Figure 1 presents the comparison of energy spectra of 662 keV $\gamma$-rays from a $^{137}$Cs source, as measured with LaCl$_3$ and NaI(Tl) crystals at 3 $\mu$s shaping time. Note a very good energy resolution of LaCl$_3$ of 4.2% in comparison to 6.5% observed with NaI(Tl).

The non-proportionality curves for LaCl$_3$ and NaI(Tl) are shown in Fig. 2. The non-proportionality is defined here as the photoelectron (PHE) number yield measured at specific $\gamma$-ray energy relative to the PHE number yield at 662 keV $\gamma$-peak. LaCl$_3$ is clearly superior to NaI(Tl) in terms of non-proportionality.

2.2. LaBr$_3$:Ce Crystal

LaBr$_3$ crystals are characterized by a high energy resolution down to 2.8% for 662 keV gamma-rays from $^{137}$Cs source, as measured with 3′′ × 3′′ LaBr$_3$ crystal. A high light output above 60000 ph/MeV, very good proportionality of the light yield versus gamma ray energy and a high speed of the light pulse with the decay time constant of 18 ns must be pointed out [5,6]. Figure 3 presents a comparison of the energy spectra of 662 keV $\gamma$-rays from a $^{137}$Cs source measured with 3′′ × 3′′ LaBr$_3$ and NaI(Tl) crystals.

![Figure 3: A comparison of the energy spectra of 662 keV $\gamma$-rays from a $^{137}$Cs source measured with 3′′ × 3′′ LaBr$_3$ and NaI(Tl) crystals. Courtesy of Saint-Gobain Crystals.](image)

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![Figure 4: A comparison of the non-proportionality curves measured for the LaBr$_3$ and NaI(Tl) (25 mm × 31 mm) crystals, respectively.](image)

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662 keV gamma rays from a $^{137}$Cs source measured with $3'' \times 3''$ LaBr$_3$ and NaI(Tl) crystals. Note a superior energy resolution of the new crystal and its better full energy peak efficiency.

Figure 4 presents a comparison of non-proportionality characteristics of LaBr$_3$ and NaI(Tl) crystals. A much better proportionality of LaBr$_3$ allows a simpler energy calibration of gamma spectrometers and, moreover, it is reflected in the superior energy resolution.

A high temperature stability of the light output of LaBr$_3$ crystal corresponding to about 0.01%/°C was recently measured in the temperature range from $-30^\circ$C to $60^\circ$C [7] (see Fig. 5).

A high light output and a fast light pulse of LaBr$_3$ crystal allowed getting an excellent time resolution. Figure 6 presents time spectra measured with $1'' \times 1''$ LaBr$_3$ coupled to the XP20D0 PMT in relation to the BaF$_2$ crystal [8].

No doubt that the LaBr$_3$ crystal is the scintillator of a choice for all equipment used in the border monitoring, when gamma spectrometry is needed. It includes also the neutron activation methods addressed to detect explosives in the luggage and big trucks.

3. Scintillators with a High Sensitivity – Comparison to BGO and NaI(Tl) Scintillators

New high-Z scintillator detectors are needed to significantly improve current border monitoring instrumentation utilizing plastic detectors, as well as the development of highly efficient hand-held instrumentation. BGO, widely used high-Z scintillator, suffers because of a low light yield and in consequence a poor energy resolution. CdWO$_4$ (CWO) [9,10] and CaWO$_4$ (CaWO) [11,12] are candidate to be used for this purpose. An application of CWO and CaWO in the gamma spectrometry is limited because of a long main decay time constant of the light pulses, which does not allow measurements at high counting rates. Both crystals belong to the group of “very slow” scintillators with a long decay time constant of the light pulse of 14 µs for CWO [9] and 9µs for CaWO [11,12]. However, in the border monitoring equipment, one is looking for traces of gamma rays emitted by well-shielded radioactive sources. Thus, high rate capabilities of the detectors are less important.

Figure 7 shows the energy spectra of 662 keV γ-rays from a $^{137}$Cs source measured with a small CWO crystal of $10 \times 10 \times 3$ mm$^3$ and with a 25 mm

![Figure 6: Time spectra of $^{60}$Co and $^{22}$Na γ-rays detected in the LaBr$_3$ crystal coupled to the XP20D0, measured in relation to the BaF$_2$ detector.](image-url)
Figure 7: Energy spectra of 662 keV $\gamma$-rays from a $^{137}$Cs source, as recorded with the $10 \times 10 \times 3 \text{ mm}^3$ CWO (bottom panel) and with the $\varnothing 25 \text{ mm} \times 30 \text{ mm}$ NaI(Tl) (upper panel). An escape peak of $K$ x-rays of Tungstate is seen at the 662 keV peak in the CWO spectrum.

in diameter and 30 mm high NaI(Tl).

Note the high energy resolution of $6.6 \pm 0.2\%$ for the 662 keV peak obtained with the CWO, which is comparable to that of NaI(Tl). Note also the comparable photofractions of 26% and 23% in both spectra for the CWO and the NaI(Tl) crystals, respectively, while the volume of the NaI(Tl) crystal is 50 times larger.

Figure 8 presents the non-proportionality characteristics of the small CWO in comparison to that of a BGO crystal, according to Ref. [10]. The non-proportionality is defined here as the ratio of the photoelectron yield measured for photopeaks at a specific $\gamma$-ray energy relative to the yield at 662 keV $\gamma$-peak [13]. Note a comparable shape of the measured curve for CWO to that reported in Ref. [14] for BGO.

The study of the small CWO crystals showed a high light output of $27300 \pm 2700 \text{ ph/MeV}$ and an energy resolution of $6.6 \pm 0.2\%$ for the 662 keV $\gamma$-rays from a $^{137}$Cs source, both placing the CWO crystal within bright scintillators with a good energy resolution.

In the case of CaWO, its high light output of $4800 \pm 200 \text{ phe/MeV}$ and a good energy resolution of $6.6\pm0.2\%$ for 662 keV gamma rays from a $^{137}$Cs source have to be noted, as measured for the small samples coupled to the XP3212 photomultiplier.

There is no doubt that due to the high energy resolution, the CWO and CaWO crystals can replace the BGO in several applications.

REFERENCES