

The Comparison of Large Scintillators for High Energy Gamma-Rays Detection

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Abstract—The quest for finding the suitable detector for application in neutron activation analysis triggered us to investigate the properties of three 3"×3" cylindrical scintillation detectors. The emphasis was put on the detectors' properties in the multi-MeV energy region as well as their response to neutron radiation. In this work we compare energy resolution efficiency and timing properties of BGO, LaBr₃ and NaI(Tl) scintillation detectors coupled to Photonis spectrometric photomultiplier tubes.

Index Terms—Neutron activation analysis, scintillation detectors.

I. INTRODUCTION

THIS work was motivated by the research project aimed at designing a mobile system taking the advantage of the neutron activation analysis (NAA) [1],[2],[3]. NAA is a sensitive, analytical technique, which is used for qualitative as well as for quantitative analysis of numerous materials. Nuclei interacting with neutrons can be excited to energies unavailable to any other, cheaply available technique. Thus, the efficient application of NAA requires a gamma detector with sufficiently high absorption efficiency for the gamma energies approximately between 1 MeV and 10 MeV [4],[5]. For this reason size matters, but with this particular parameter fixed, we undertook the effort to compare the other scintillator properties, such as the energy and timing resolutions, the stability and the detection efficiency. In this work we focused at three, commercially available cylindrical scintillators, each 3"×3" large, see Fig. 1 and Table I. LaBr₃ was chosen because of its superior energy resolution, BGO is well known of its efficiency, while NaI(Tl) serves as the reference point for comparison. Plastic scintillators, even though fast and cheap, were excluded due to their poor efficiency and susceptibility to neutron radiation. It would be probably an interesting idea to take a closer look at BaF₂, unfortunately, there was no sample large enough in our possession.

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Table I
APPLIED DETECTORS

Type	Manufacturer	Size	Shape	PMT
LaBr ₃	Saint Gobain	3" × 3"	cylindrical	XP5700B
BGO	Novosibirsk	3" × 3"	cylindrical	XP5300B
NaI(Tl)	Amcrys-H	3" × 3"	cylindrical	XP3312



Fig.1. The photograph of the three tested detectors: NaI(Tl) (top), LaBr₃ (right), BGO (left).

TABLE II
BASIC PROPERTIES OF THE STUDIED SCINTILLATORS

Type	Decay time (ns)	Wavelength of peak emission (nm)	Density (g/cm ³)	Light output (ph/MeV)	hygroscopic
LaBr ₃	16	380	5.08	74000 ±7000 ^{a)}	yes
BGO	300	480	7.13	8500 ±350 ^{b)}	no
NaI(Tl)	250	415	3.67	38000 ±3000 ^{c)}	yes

a) see [6], b) see [7], c) see [8].

Various physical properties of crystals, collated for the reference in Table II, result in different properties of complete detectors. High photoelectron numbers measured with tested crystals reflect high quantum efficiency of modern PMTs of order of 35%. Which one of the detectors is supposed to be most suitable for high energy gamma spectrometry, whether it is LaBr₃ with its excellent energy resolution or BGO with the high peak-to-total fraction is the question that we try to answer in this work.

II. EXPERIMENTAL RESULTS

It is clear that the scintillator common properties and the corresponding measurement techniques in particular are well known [6],[9],[10] so the paper focuses only on the properties that are crucial for the application in neutron activation analysis techniques. This is also the reason why we put the interest in performance of the detectors in the energy range above 500 keV.

A. Energy resolution

With the Genie 16d neutron generator in our possession [11], and the detectors large enough to efficiently accumulate desired statistics, we were able to reach energies as high as 10 MeV for the energy resolution consideration.

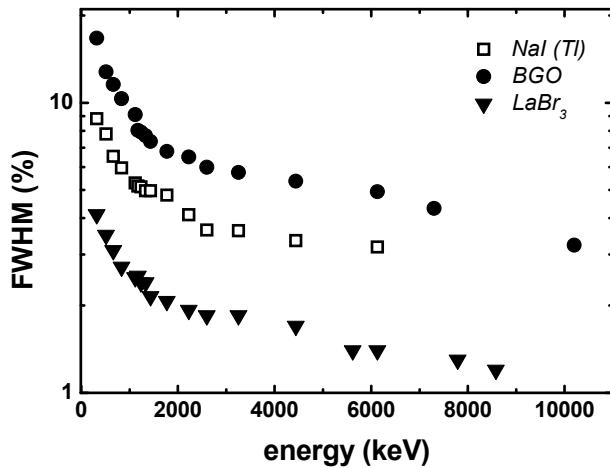


Fig. 2. The energy resolution characteristics of BGO (solid circles), LaBr₃ (solid triangles), and NaI(Tl) (open squares) detectors. The FWHM(E) parameters were derived by fitting energy spectrum peaks with Gaussian function. Error bars are of the size of points and represent statistical errors.

Figure 2 presents relative energy resolution as a function of gamma energy for all tested scintillators. The performance of BGO detector at the energy of 661.7 keV reveals energy resolution of 11.5% which deteriorates rapidly, when the energy of γ -rays decreases. At the gamma energy of 320 keV the energy resolution of BGO is about two times worse than that of NaI(Tl) and over four times worse than that of LaBr₃. This fact is confirmed in our previous paper [4] where the results of the non-proportionality of large BGO and NaI(Tl) were published. Fortunately, in the case of NAA, the

scope of interest is far above this energy, and in fact, above the energy of 4.44 MeV the relative differences between investigated scintillators are not that significant, and where other factors, such as the full absorption efficiency, become more important, as can be seen in the Fig. 2. However, The studied LaBr₃ yields the superior energy resolution of 1.7% at 4.4 MeV gamma rays.

In order to obtain clear high energy peaks we took the advantage of our neutron generator. The 6128 keV peak can be measured by the activation of oxygen, abundant in water. The knockout reaction



yields the nitrogen isotope, which 7.2 s half-life enables easy isolation of the interesting transition in oxygen. The measurement is performed in the pulsed mode of generator and the signal is taken just after the pulse of fast neutrons. The points in Fig. 2, corresponding to 7.3 MeV and 10.2 MeV peaks, are visible only in BGO crystal, as this is a result of epithermal neutrons capture by one of the germanium isotope (⁷³Ge) within the scintillator itself.

Finally, the exceptionally good energy resolution of LaBr₃ allows for identifying more subtle lines such as the transitions in ³⁶Cl after the neutron capture by the stable chlorine isotope in salt. Access to higher energies is possible, however, it requires access to accelerator and experimental capability to perform in-beam measurements [12].

It is worth to remind that the absolute energy resolution is deteriorating with the increase of gamma energy. 3% at 6 MeV means that the peak has almost 200 keV at its FWHM (*Full Width at Half Maximum*), and that this number becomes 300 keV when the energy approaches 10 MeV. The effect of pair creation, adding the first and the second escape peaks, blurs the picture even more. Unless the investigated line is well isolated, as it is in the case of 10.8 MeV neutron capture line of ¹⁴N, the task of distinguishing separate lines becomes extremely difficult, and/or weighted down with measurement and analysis uncertainties.

B. Efficiency

When speaking about efficiency in the context of gamma detection one usually thinks about the probability of registration of gamma rays emitted and gamma rays that hit the detector. Whereas the former aspect is mainly related to the setup geometry, the latter is determined by the intrinsic properties of the applied detector. The absolute efficiency of the detector is important in every application constrained by the measurement time. This parameter gets even more important whenever there are limits to the device physical dimensions, and when the growing price tag prevents from applying just a bigger unit. These are exactly the same requirements that made scintillators so popular in commercial applications and that made the HPGe detectors visible mainly among laboratory equipment. In the neutron activation analysis, which itself is a form of spectrometry, the emphasis

is also put on the total absorption efficiency, i.e. the probability of registering the full energy of gamma rays that interact with the detector. This quantity is related to the peak-to-total fraction of the particular detector, being in turn a function of the Z value of mayor element and a size of the detector.

Since we are interested in a high energy gamma spectrometry, a comparison of the peak-to-total fraction of studied detectors was done for 4.4 MeV gamma rays emitted from a Pu-Be source and detected by the studied detectors, see Fig. 3. The numerical values are collected in Table III, calculated above the threshold of 2.5 MeV.

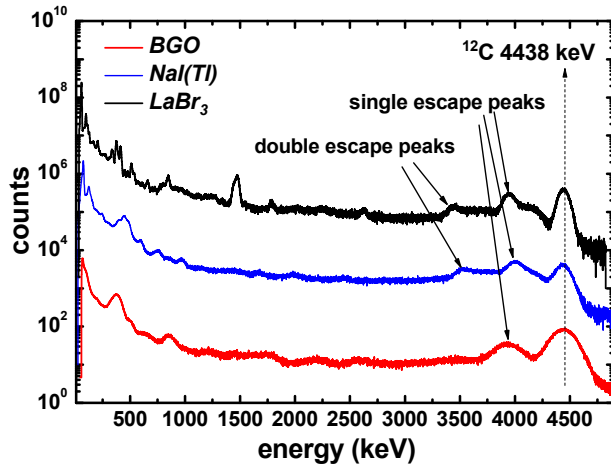


Fig. 3. Energy spectra of 4.4 MeV γ -rays from a Pu-Be source measured with three tested scintillators: BGO (red line), NaI(Tl) (blue line) and LaBr₃ (black line) measured with Pu-Be source. Full energy peak from ¹²C as well as single and double escape peaks can be clearly seen in the figure.

It confirms the largest full energy peak efficiency in the BGO crystal, twice as large, as that of the LaBr₃, but the energy resolution of LaBr₃ is three times better, see Table III.

It is of course no big science to learn, that full absorption efficiency is a direct function of the scintillator density, or more precisely, its effective atomic mass Z (for the same dimension of crystals). Thus further, we did a practical comparison of spectra quality obtained by measuring high energy transitions in oxygen from the beta decay of nitrogen, by BGO and LaBr₃, as in the (1) reaction at the beam line of the neutron generator.

Both the spectra, visible in the Fig. 4, were accumulated in the same experimental conditions. As we were vividly interested in practical aspects of the crystals different absorption capabilities, we compared area of the Gaussian full absorption peak fit to the total number of counts in the selected energy region between 3180 keV and 8000 keV. The outcome, 12% for BGO and 11% for LaBr₃, revealed to be contrary to the measured peak-to-total ratio in a similar region of energies, see Table III. The fact, that over 13 times weaker 7118 keV and its 6607 keV escape peaks are plainly visible in the LaBr₃ detector, and are hardly noticeable in BGO, clearly points out that superior efficiency of BGO cannot compensate

other deficits of this scintillator.

The results of the measurements presented in Fig. 4 shows that in the selection process of the optimal scintillator for application in the NAA, that besides energy resolution and a detection efficiency, the cross sections for reactions with fast and thermal neutrons of mayor elements in the crystal have to be taken into account.

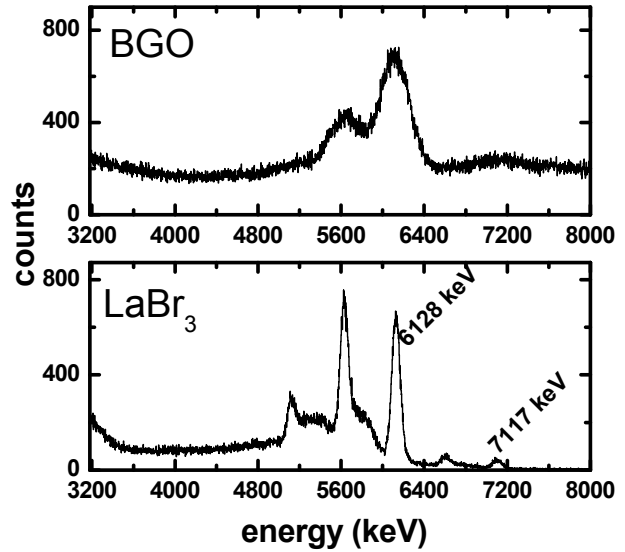


Fig. 4. The comparison of the detectors response to the (mainly) 6128 keV γ -line from oxygen. The intensity of the 7117 keV line, which is clearly visible only by the lanthanum bromide detector, is only 7.25% of the former one.

Table III summarizes the main performances of the studied detectors. The energy resolution of 1.7% for 4.4 MeV gamma line confirms the excellent performance of LaBr₃ crystal in spectrometry of high energy gamma rays. On the other hand, peak-to-total ratio, calculated for 4.4 MeV gammas, favors the BGO.

TABLE III
A COMPARISON OF ENERGY RESOLUTION AND EFFICIENCY PERFORMANCE OF TESTED SCINTILLATORS

Crystal	Energy resolution (%)		Peak-to-total ^{a)} %
	662 keV	4.4 MeV	
NaI(Tl)	6.5	3.3	10±4
BGO	11.5	5.4	34±3
LaBr ₃	3.1	1.7	16±5

a) determined above 2.5 MeV threshold

C. Time resolution

Application of the tagged neutron generator, i.e. the one with the dedicated associated alpha-particle detector, offers the opportunity of creating the device capable to obtain the 3D visualization of the investigated object. Taking the advantage

of any time-of-flight technique, however, requires a detector of sufficiently good time resolution, as the latter has the direct impact on spatial resolution. Since we were testing the complete detectors, i.e. scintillators coupled with the PMTs and unmodified voltage dividers, the achieved results are significantly worse, than those obtained with the dedicated timing PMTs. In order to make our measurements as close as possible to real application we were measuring the n - γ coincidences in a slow-fast set-up with a Pu-Be source. The additional measurements with the ^{60}Co source were performed to provide the reader with numbers that may be compared with the results of the dedicated timing measurements [4],[12], [13],[14].

The measurements were done in the optimal conditions for each detector, using constant fraction timing (CFD) for LaBr_3 , low level leading-edge for $\text{NaI}(\text{Tl})$ and CFD timing with partly integrated light pulse for BGO, following [15] and [16]. In the case of measurements with Pu-Be source, the threshold was at 1 MeV. In all the measurements, a small BaF_2 crystal coupled to the XP2020Q Photonic PMT was used as a reference.

The measured spectra for all tested detectors are presented in Fig. 5. It shows a superior time resolution of the LaBr_3 detector of 380 ± 15 ps for ^{60}Co gamma rays and 660 ps measured with Pu-Be source.

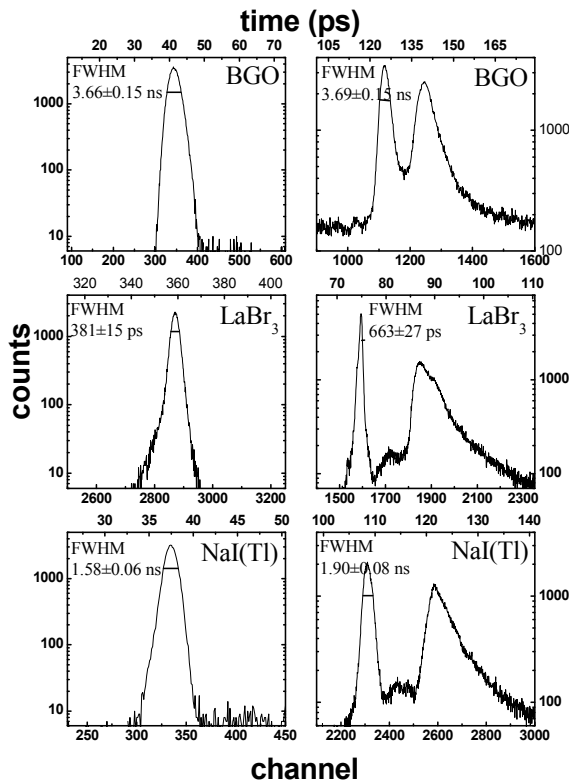


Fig. 5. Examples of time spectra measured with the investigated detectors. Left panels show time spectra of γ - γ coincidences of 1172 keV and 1333 keV gamma rays of ^{60}Co , while right panels depict γ - γ and γ - n coincidences in Pu-Be source. The spectra were measured with, starting from top, BGO coupled with XP5300B PMT, LaBr_3 coupled with XP5700B PMT, and $\text{NaI}(\text{Tl})$ coupled with XP3312 PMT.

TABLE IV
TIME RESOLUTION OF THE TESTED DETECTORS

Scintillator	Timing γ - γ in ^{60}Co (ns)	Timing γ - γ in Pu-Be (ns)
BGO	3.66 ± 0.15	3.69 ± 0.15
LaBr_3	0.381 ± 0.015	0.663 ± 0.027
$\text{NaI}(\text{Tl})$	1.58 ± 0.06	1.90 ± 0.08

Considering the fact that the speed of 14 MeV neutrons is about 5 cm/ns one can easily notice the practical limitation of any 3D, NAA based, scanner that has one of the described detectors applied in its design. Nevertheless, the LaBr_3 detector shows much better performance in this category than its competitors. BGO, mostly due to its long light pulse duration and a low light output, is in a great disadvantage when compared to other detectors.

D. Stability

Almost every time, when a device is moved away from its laboratory, the question about temperature stability arises. The best approach to solve this problem is, of course, to get the advantage of some temperature inert phenomena, compensate the changes or, if it is impossible, stabilize the environment. We would like to point at an interesting result of well known temperature instability of BGO scintillators. Even though our measurements were performed in the air conditioned laboratories yet the effects depicted in the Fig. 6 were still observed.

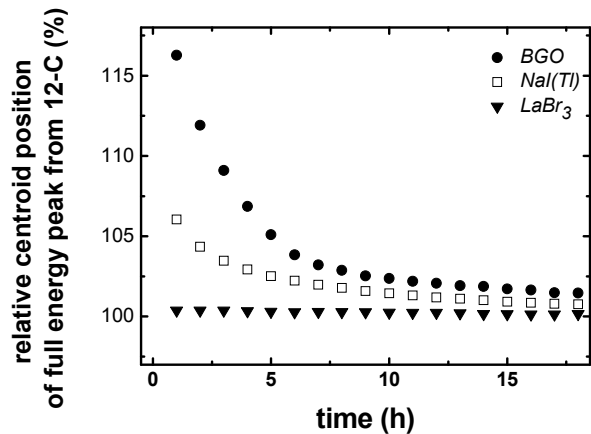


Fig. 6. Turn on effects for investigated detectors. Various crystals show different levels of susceptibility on meager temperature changes.

The temperature of the detector increased by 1.5° while the PMT was warming up. This effect changed the gain of the BGO detector by almost 15%, what suggests that compensation methods are the only solution when considering application of the BGO scintillator in any mobile or outdoor device. Nevertheless, such behavior is definitely not a strong point of BGO and questions its advantages in other categories.

In contrast, the LaBr₃ detector presents excellent thermal stability (Fig. 6.) [16].

III. CONCLUSIONS

The comparative study of large LaBr₃, BGO and NaI(Tl) crystals in a spectrometry of high energy gamma rays showed a superior performance of the LaBr₃. It is due to an excellent energy resolution, below 2% at 4.4 MeV gamma rays, a very good timing of hundreds picoseconds and good long term stability. A significantly lower peak-to-total fraction than that measured with BGO is less important in the application to the neutron activation analysis, because of a spurious background in BGO produced by the interaction of neutrons with the BGO crystal itself.

REFERENCES

[1] D. R. Brown, and T. Gozani, "Cargo inspection system based on pulsed fast neutron analysis", *Nucl. Instr. Meth.*, vol. B99, pp. 753, 1995.

[2] G. Nebbia, S. Pesente, M. Lunardon, G. Viesti, P. LeTourneur, F. Heuveline, M. Mangeard, and C. Tchong, "Performance of a tagged neutron inspection system (TNIS) based on portable sealed generators", *Nucl. Instr. Meth.*, vol. A533, pp. 475, 2004.

[3] B. Perot, M. Moszyński, T. Batsch, M. Gierlik, D. Wolski et al., "Development of the EURITRACK Tagged Neutron Inspection System", *Nucl. Instr. Meth.*, vol. B261, pp. 295, 2007.

[4] M. Gierlik et al. "Comparative Study of Large NaI(Tl) and BGO Scintillators for the EUROpean Illicit TRAfficking Countermeasures Kit Project," *IEEE Trans. Nucl. Sci.*, vol. 53, no. 3, June 2006, pp. 1737-1743.

[5] J. E. McFee, A. A. Faust, H. Robert Andrews, V. Kovaltchouk, E. T. Clifford, and H. Ing, "A comparison of fast inorganic Scintillators for thermal neutron analysis landmine detection," *IEEE Trans. Nucl. Sci.*, vol. 56, no. 3, June 2009, pp. 1584-1592.

[6] M. Moszyński, L. Świdorski, T. Szcześniak, A. Nassalski, A. Syntfeld-Kazuch, W. Czarnacki, G. Pausch, J. Stein, P. Lavoute, F. Lherbert, and F. Kniest, "Study of LaBr crystals coupled to photomultipliers and avalanche photodiodes," *IEEE Trans. Nucl. Sci.*, vol. 55, no. 3, pp. 1774-1780, Jun. 2008.

[7] M. Moszyński, M. Kapusta, M. Mayhugh, S.O Flyckt, "Absolute Light Output of Scintillators", *IEEE Trans. Nucl. Sci.*, vol. 44, no. 3, pp. 1052-1061, June 1997.

[8] I. Holl, E. Lorenz, G. Mageras, "A measurements of light yield of common inorganic scintillators", *IEEE Trans. Nucl. Sci.*, vol. 35, no. 1, pp. 105-109, Febr. 2005.

[9] L. Swiderski, M. Moszynski, W. Czarnacki, A. Syntfeld-Kazuch and M. Gierlik, "Non-Proportionality and Energy Resolution of NaI(Tl) at Wide Temperature Range (40°C to +23°C)", *IEEE Trans. Nucl. Sci.*, vol. 54, no. 4, August 2007, pp. 1372-1377.

[10] M. Moszynski, M. Balcerzyk, W. Czarnacki, M. Kapusta, W. Klamra, A. Syntfeld, and M. Szawłowski, "Intrinsic Energy Resolution and Light Yield Nonproportionality of BGO", *IEEE Trans. Nucl. Sci.*, vol. 51, no. 3, June 2004, pp. 1074-1079

[11] EADS Sodern web page with product specifications, http://www.sodern.fr/site/FO/scripts/siteFO_contenu.php?noeu_id=45&lang=EN

[12] M. Ciemala, D. Balabanski, J. M. Daugas, G. Georiev, J. Gulyás, M. Kmiecik, A. Krasznahorkay, S. Lalkovski, A. Lefebvre-Schuhl, R. Lozeva, A. Maj, and A. Vitez, "Measurements of high-energy γ -rays with LaBr₃:Ce detectors", *Nucl. Instr. Meth.*, vol. A608, pp. 76, 2009.

[13] T. Szcześniak, M. Moszyński, L. Świdorski, A. Nassalski, A. Syntfeld-Kazuch, A. G. Dehaine, and M. Kapusta, "A comparative study of fast photomultipliers for timing experiments and TOF PET", *IEEE Trans. Nucl. Sci.*, vol. 56, no 3, pp. 1017-1023, Jun. 2009.

[14] T. Szcześniak, M. Moszyński, A. Nassalski, P. Lavoute, A. G. Dehaine, "A further study of timing with LSO on XP20D0 for TOF PET", *IEEE Trans. Nucl. Sci.*, vol. 54, no 5, pp. 1464-1473, Oct. 2007.

[15] M. Moszynski, J.H. Bjerregard, J.J. Gaardhoje, B. Herskind, P. Knudsen and G. Sletten" Limitation of the Compton Suppression in Ge-BGO Spectrometers" *Nucl. Instr. Meth.* vol. A280 (1989) pp. 73-82

[16] M. Moszyński, A. Nassalski, A. Syntfeld-Kazuch, T. Szcześniak, W. Czarnacki, D. Wolski, G. Pausch, and J. Stein, "Temperature dependences of LaBr : Ce, LaCl : Ce and NaI(Tl) scintillators," *Nucl. Instrum. Meth.*, vol. A568, pp. 739-751, Jun. 2006.