Scintillation Products
Whitepaper

Performance Summary: BrilLanCe® Scintillators $LaCl_3$:Ce and $LaBr_3$:Ce

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INTRODUCTION –

Since the discovery of $LaCl_3$:Ce and $LaBr_3$:Ce as scintillators by Delft and Bern Universities[1,2], several groups have furthered understanding of their properties; and Saint-Gobain Crystals has made considerable progress in their commercial availability as BrilLanCe® 350 and BrilLanCe® 380 crystals. Specifically, in this work, BrilLanCe 350 is $LaCl_3$(10%Ce) and BrilLanCe 380 is $LaBr_3$(5%Ce).

In this summary, we report the performance of BrilLanCe 350 and BrilLanCe 380 detectors and extend results to the recently available 3" diameter, 3" long ("3x3") crystals, drawing mainly from results published or presented elsewhere[3,4,5]. We are not attempting a comprehensive review and remind the reader that a partial compilation of the general literature is available on our website, www.detectors.saint-gobain.com. A table of scintillator properties is found at the end of this summary, where we note in particular that BrilLanCe 380 crystals emit some 60% more light than NaI(Tl) for energies near 1 MeV and have much faster decay times and better timing properties.

Herein we cover energy resolution and relative efficiency as a function of gamma-ray energy emphasizing a comparison of BrilLanCe 380 and NaI(Tl) detectors for the 3"x3" size. We also take a look at performance versus temperature, performance versus count rate, coincidence resolving time, and intrinsic background.

PERFORMANCE –

Energy Resolution versus Energy

The energy resolution for BrilLanCe 380 crystals is determined both by their high light output and by their excellent energy linearity.

For the results reported in this subsection, 3" diameter, 3" long BrilLanCe 380 and NaI(Tl) detectors were compared. They were both in integrated packages, i.e., coupled directly to a 3" diameter photomultiplier – a Photonis XP5300B for the BrilLanCe 380 detector and an ETI 9305 for the NaI(Tl). The source was "end-on", i.e., on axis with the detector. When the source was changed from one isotope to another, the distance was adjusted to achieve reasonable counting rates of a few thousand per second, and that same distance was used for both detectors so that efficiencies could be compared directly.

We begin the comparison with the response of the two detectors to ^{137}Cs (662keV) and Figure 1, where the 3"x3" detector spectra are compared. Both the source's gamma ray at 662 keV and its barium K_α X-ray at 32 keV are shown. Spectra are normalized to 662 keV on the energy scale.

The figure also reports the areas under the 662 keV photopeak, set as 100% for NaI(Tl) and giving 118% as the relative efficiency for the BrilLanCe 380 unit primarily due to its higher density. For NaI(Tl) the peak near 32 keV is slightly higher on the energy scale than the one for the BrilLanCe 380 detector because NaI(Tl) is non-linear, producing a bit more light per keV at lower energies than at higher ones.

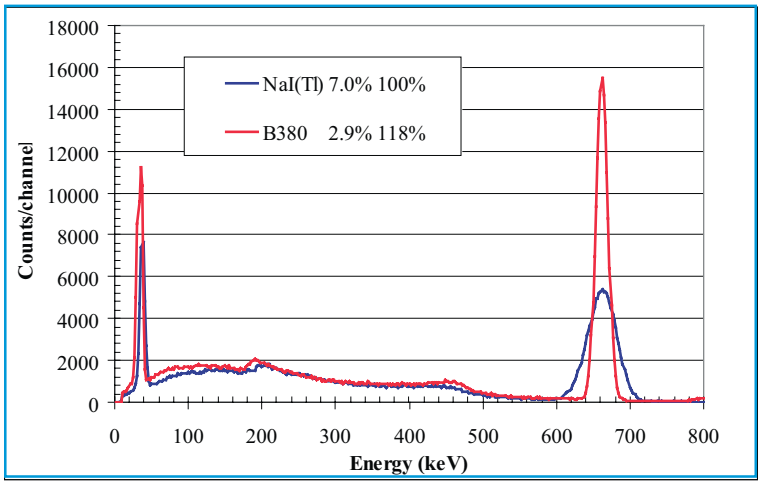


Figure 1
Comparison of 3"x3" spectra for ^{137}Cs (662 keV) BrilLanCe 380 detector (red) and NaI(Tl) (blue)

Figure 2 shows the response of the two detectors for ^{60}Co where the well-known lines at 1332 and 1173 keV are seen. At 1332 keV, the BrilLanCe 380 unit gives 2.1% energy resolution versus 5.4% for the NaI(Tl) unit. The BrilLanCe 380 detector is 43% more efficient. In addition, a line is seen in the 35 keV region in the BrilLanCe 380 curve. This is due to emission of Ba X-rays from ^{138}La background which is discussed in detail below.

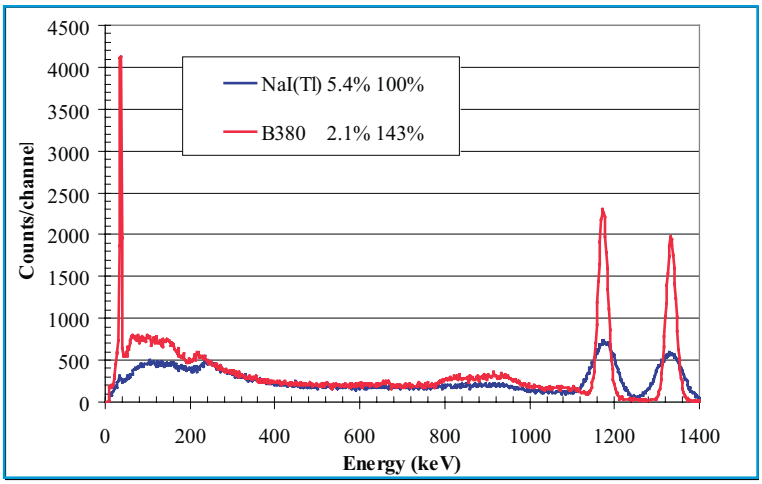


Figure 2
Comparison of 3"x3" spectra for ^{60}Co
BrilLanCe 380 detector (red) and NaI(Tl) (blue)

At 2615 keV (^{208}Tl in the thorium decay chain), the BrilLanCe 380 detector achieves 1.6% energy resolution versus NaI(Tl)'s 4.5% and is 65% more efficient as seen in Figure 3. Again, spectra are normalized at the highest energy line, 2615 keV, and energy offsets are seen between the two materials at lower energies due to the differences in linearity. The improvement in spectral resolution with the BrilLanCe 380 package is particularly apparent in this multiple energy spectrum. The line seen just below 1500 keV in the BrilLanCe 380 spectrum is again due to background and will be discussed further.

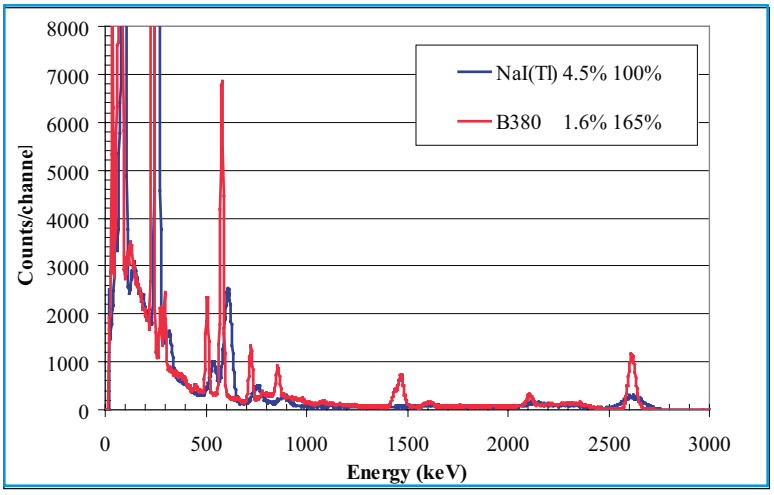


Figure 3
Comparison of 3"x3" spectra for the
Thorium decay chain.
BrilLanCe 380 detector (red) and NaI(Tl) (blue)

The advantages of BriLanCe 380 detectors continue to low energies as seen in Figure 4, which shows the response to ^{57}Co. The BrilLanCe 380 detector clearly resolves the 136 keV line from the 122 keV line while NaI(Tl) does not. The background from Ba X-ray lines in the 35 keV region is also seen in the BrilLanCe 380 spectrum.

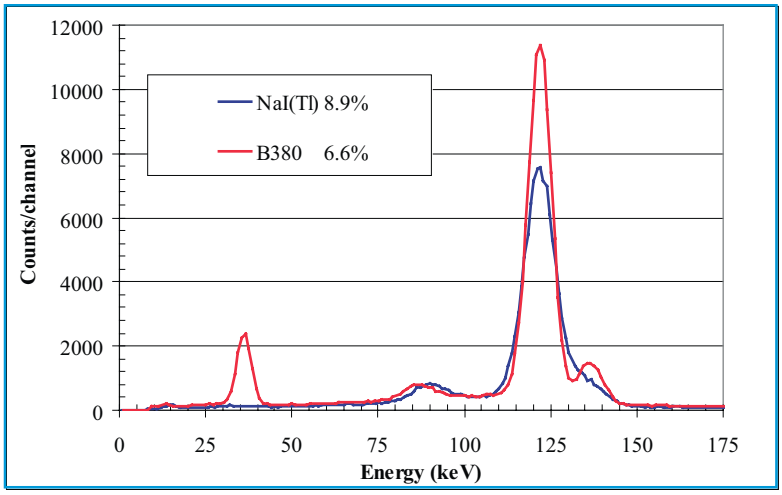


Figure 4
Comparison of 3"x3" spectra for ^{57}Co
BrilLanCe 380 detector (red) and NaI(Tl) (blue)

To complete the survey, Figure 5 shows a ^{133}Ba spectrum for the two detectors. The BrilLanCe 380 detector shows substantially better separation of the lines near 350 keV. Both detectors show prominent lines just above 30 keV due to Cs K_α X-rays emitted by the source.

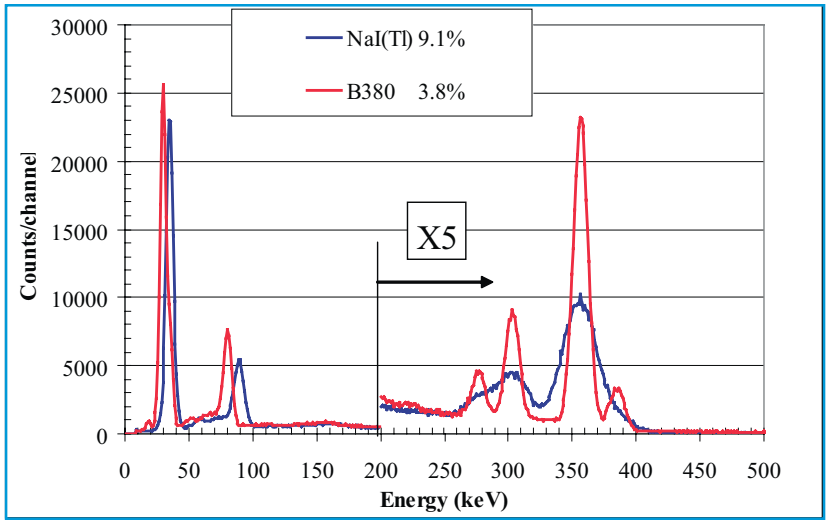


Figure 5
Comparison of 3"x3" spectra for ^{133}Ba
BrilLanCe 380 detector (red) and NaI(Tl) (blue)

Table 1 summarizes the results from this section with energy resolution and relative efficiency tabulated for several of the energies presented. The advantages of BrilLanCe 380 detectors over NaI(Tl) are seen at all energies.

Table 1 - Summary
3"x3" Detector Response vs. Energy
Resolution and Relative Efficiency

Energy (keV)	Resolution BrilLanCe 380	Resolution NaI(Tl)	Ratio Peak Counts
122	6.6%	8.9%	1.05
356	3.8%	9.1%	1.06
662	2.9%	7.0%	1.18
1332	2.1%	5.4%	1.43
2615	1.6%	4.5%	1.65

The well-behaved nature of the energy resolution versus energy is displayed in Figure 6 with this data overlaid on points extracted from earlier work and smaller detectors. The energy resolution faithfully follows the square root of energy as expected statistically for linear detectors. Although it is not demonstrated, one can conclude from the data in Table 2 and Figure 6 that NaI(Tl) does not track this scheme.

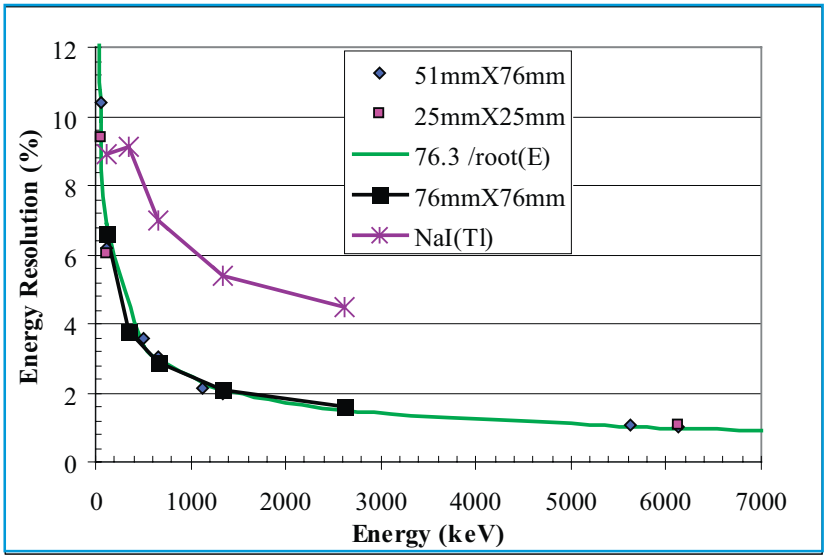


Figure 6
Energy resolution as a function of energy

Response versus Temperature

The remarkable properties of BrilLanCe 380 crystals are preserved as temperature increases as shown in Figure 7, and light output is dramatically higher at high temperatures than it is for all other crystals tested. The data for Figure 7 was taken early in the program[1] and only showed 30% more light for the BrilLanCe 380 system, but subsequent data confirms 60% more light in similar rugged, high temperature packages useful in oil well logging. Investigators at Delft University have shown high light output is preserved to as high as 600 K.

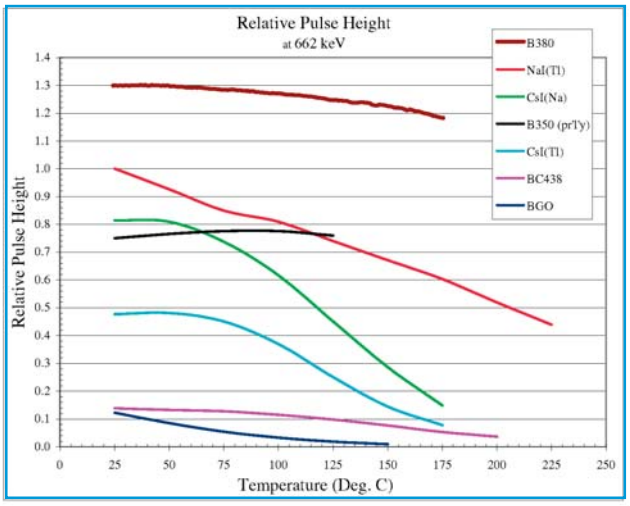


Figure 7
Response versus Temperature

Response versus Rate

Given the ten-fold difference in decay time between NaI(Tl) and BrilLanCe crystals, performance to high rate is expected and is displayed in Figure 8. These measurements used 1"x1" crystals and the same 8575 photomultiplier for each detector followed by a timing filter amplifier and constant fraction discriminator. Rate was adjusted by changing source strength and position.[3] This demonstration verifies the expected difference between materials, but detailed results are highly dependent on the electronics configuration chosen, of course.

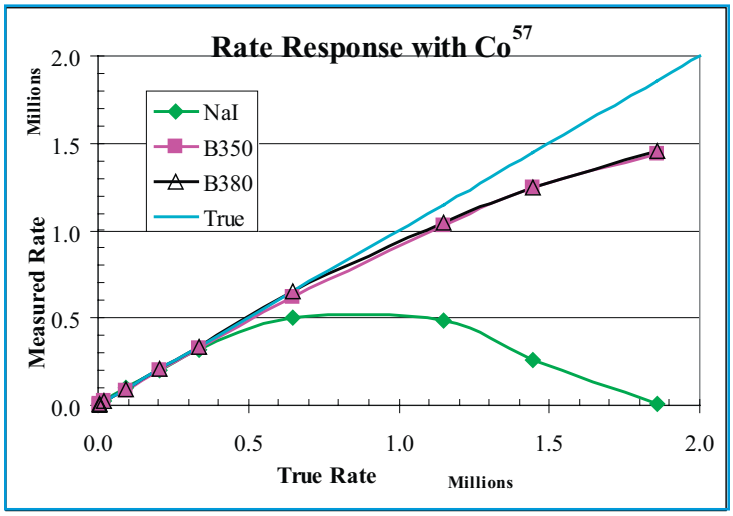


Figure 8
Response versus Rate

Coincidence Resolving Time

As also expected from decay time properties and high light output, the BrilLanCe crystals have excellent timing properties as the figure of merit column suggests in Properties Table at the end of this summary. The figure of merit (F.O.M.) column there, square root of decay time divided by light output gives a first indication of expected timing performance. These crystals are fast enough that timing results also depend on light propagation times and thereby on crystal size as we will see. Other groups have also shown that decay time is only a rudimentary indicator

of rise time which is the more telling parameter, but more difficult to measure, and very dependent on the choice of photomultipier.

Representative coincidence resolving times (CRT) are shown in Figure 9 for various sizes of BrilLanCe 350 and BrilLanCe 380 detectors. The data was taken using two Photonis XP20Y0 photomultipliers (PMTs). The PMT serving as the STOP channel was coupled to the crystal to be measured and the other PMT was coupled to a dedicated START crystal, a 4x4x5 mm BrilLanCe 380 crystal of standard (5%) doping. This particular crystal had been previously measured with a "single channel" CRT value of 115 ps. "Single channel" value means that this is what would be measured against an infinitely fast channel. CRT data were taken using a ^{22}Na source. The system was gated such that only events which resulted in a 511 keV photopeak in both channels were counted.

The figure shows that BrilLanCe 380 detectors have somewhat better timing than BrilLanCe 350. We also see the dependence on crystal size, slowing as crystals and light transit times become larger. The 76 mm point is a special case because the Photonis XP20Y0 is a 51 mm diameter tube, and thus does not completely cover the crystal. When measuring this larger diameter crystal, a reflective Teflon® annulus was placed to cover the area of the window which was not covered by the PMT. The effect of this geometry on CRT has not been quantified but probably increases its value measurably.

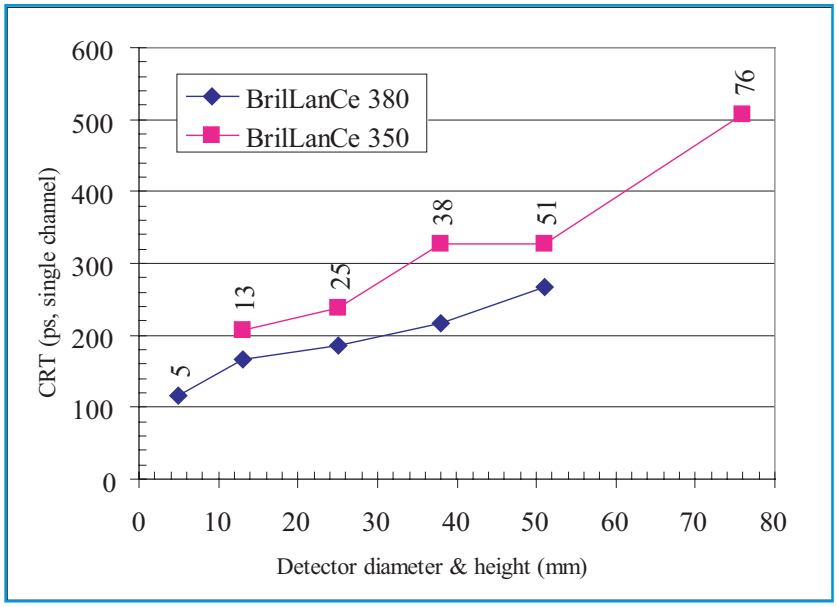


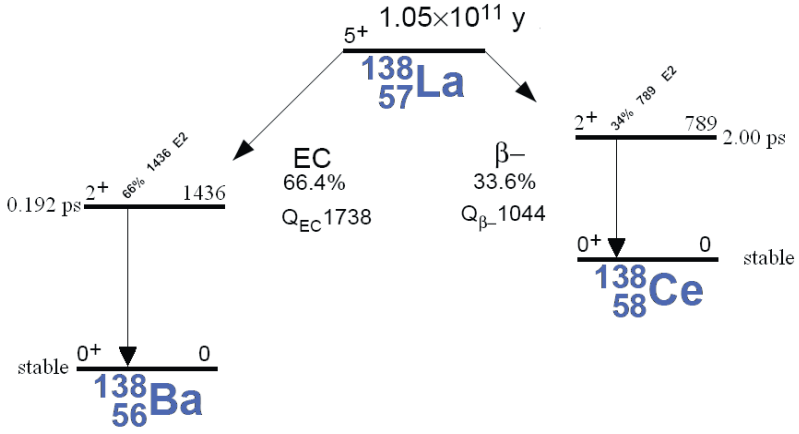
Figure 9
Coincidence Resolving Time (CRT) of BrilLanCe detectors as a function of the crystals' longest dimension.

^{138}La and ^{227}Ac Background

^{138}La is a naturally occurring radioisotope of La with 0.09% abundance and has the decay scheme shown in Table 2.[1] In 66.4% of its decays, ^{138}La undergoes electron capture (EC) to produce excited ^{138}Ba, which in turn decays by emission of a 1436 keV gamma. A necessary byproduct of electron capture is refilling of the electron shell which results in emission of coincident barium X-rays in the 35 keV region. The remaining decays, 33.6%, proceed by beta emission to ^{138}Ce, which decays by emitting a 789 keV gamma in coincidence with the beta having an end point energy of 255 keV.

The background spectrum is easily measured by self-counting in a low background chamber. Figure 10 shows such a spectrum for a 1.5" x 1.5" detector (38x38 mm) counted for about 3 days (278278 sec). Reviewing the self-counting spectrum from left to right, we see first a beta continuum at low energies for ^{138}La decays to ^{138}Ce in which the 789 keV gamma has escaped the detector altogether. This beta-only spectrum continues to its end point of 255 keV. From about 255 to 750 keV the spectrum displays the Compton continua from the 789 and 1436 keV gamma rays. The 789 keV line is next as we proceed to higher energies, but since it is in coincidence with the beta, it is smeared to high energy in a gamma plus beta continuum ending a little above 1 MeV. Finally, we see the 1436 gamma, but displaced to high energy in most events from 32 keV to 37.4 keV to total at least 1468 keV because of the coincident Ba X-rays which are also captured. The hump to the low energy side of the 1468 keV line looks at first glance to result from K_α X-ray escape but the detailed spectra are not consistent with the expected behavior. This feature is not fully explained for the moment. The Ba X-ray line in the 35 keV region can be seen alone when the 1436 keV gamma escapes, but is not visible in Figure 10 because the discriminator was set above this level. We plan supplementary experiments to confirm the X-ray energy absorbed in each decay which should be at least 32 keV and as much as 37.4 keV. The sum line at 1468 keV might be used as a calibration peak, and while potentially interfering with detection of ^{40}K at 1461 keV, has a constant and measurable activity that can be subtracted within statistical limits to determine ^{40}K.

Table 2
^{138}La decay scheme.
(from 8th edition, Table of the Isotopes)



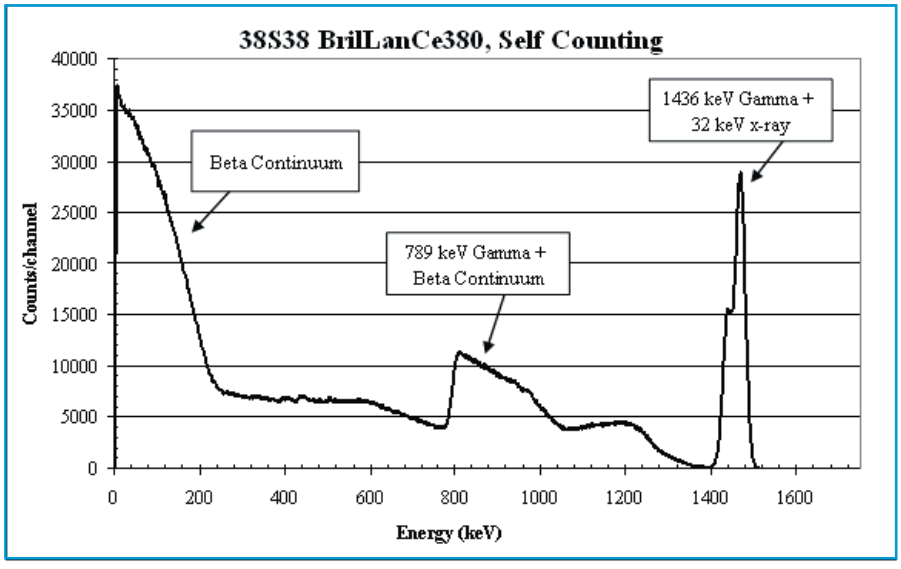


Figure 10
Self-counting background spectrum for a BrilLanCe 380 detector

Figure 11 shows background data extended to energies above 1750 keV. The presence of low level alpha contaminants is revealed. These have been shown to result from ^{227}Ac contamination.[1] Detectors produced early in our program contained higher levels of these alpha emitters and subsequent process refinements reduced them to the point that ^{138}La now produces the dominant background features.

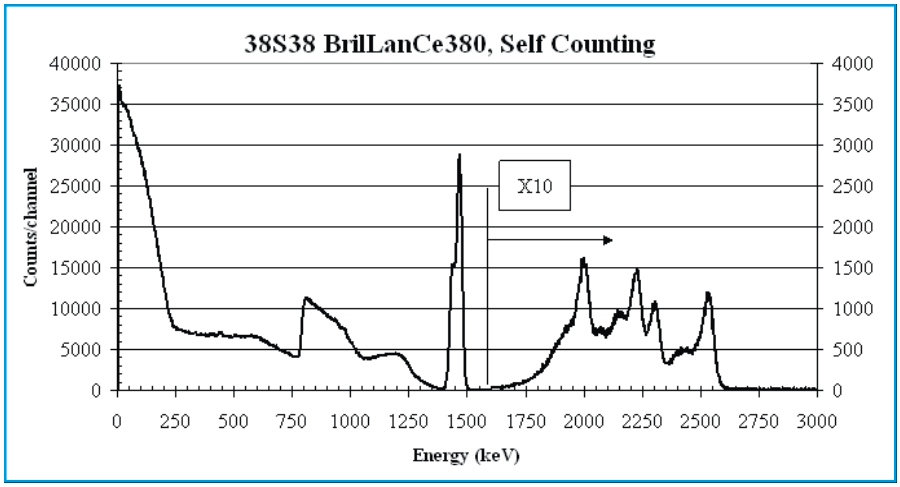


Figure 11
Self-counting background count to 3000 keV

Overall background is summarized in Table 3 for a detector about 38 x38 mm:

Table 3 - Background Count Rates/cc
From 1.5" x 1.5" Detector

0.226 cps/cc	0-255 keV beta continuum
0.065 cps/cc	790 keV – 1000 keV gamma + beta
0.068 cps/cc	1468 gamma peaks
0.034 cps/cc	Alphas above 1600 keV

TABLE OF SCINTILLATOR PROPERTIES –

Scintillator	Light Yield (photons/keV)	1/e Decay time t(ns)	F. O. M. √(t/LY)	Wavelength of maximum emission λm (nm)	Refractive index at λm	Density (g/cm^3)	Thickness (cm) for 50% attenuation (662keV)
NaI(Tl)	38	250	2.6	415	1.85	3.67	2.5
BrilLanCe® 350	49	28	0.8	350	~1.9	3.79	2.4
BrilLanCe® 380	63	16	0.5	380	~1.9	5.29	1.8
BaF2	1.8	0.7	0.6	~210	1.54	4.88	1.9
PreLude® 420	32	41	1.1	420	1.81	7.1	1.1
BGO	9	300	5.8	480	2.15	7.13	1.0

FOOTNOTES –

[1] Compiled and Edited by C. M. Rozsa, Peter R. Menge, and M. R. Mayhugh. Originally prepared for distribution by Saint-Gobain Crystals at IEEE/MIC San Diego CA, November 2006.

[2] E.V.D. van Loef , P. Dorenbos, C.W.E van Eijk, H.U. Gudel, K.W. Kraemer, *Applied Physics letters*, **77**, 04 Sept 2000, pp1467-1469

[3] E.V.D. van Loef , P. Dorenbos, C.W.E van Eijk, H.U. Gudel, K.W. Kraemer, *Applied Physics Letters.*, **79**, 1573-1575 (2001)

[4] Peter R. Menge, G. Gautier, A. Iltis, C. Rozsa, V. Solovyev *to be published in Proceedings of 2006 Symposium on Radiation Measurements and Applications, Ann Arbor MI (2006)*)

[5] A. Iltis , M. R. Mayhugh, P. R. Menge, C. Rozsa, O. Selles, V. Solovyev, III Workshop on Advanced Transition Radiation Detectors Proceedings, Ostuni, Italy Sept 7-10, 2005, to be published in Nucl. Instr. and Meth. A.

[6] C. M. Rozsa, M. R. Mayhugh, P. R. Menge Presentation to the 51st Annual Health Physics Society Meeting, Providence Rhode Island, June 27, 2006. Available on our website. Search Rozsa or Health

[7] Temperature data for NaI(Tl), CsI(Na), CsI(Tl), Plastic Scintillator (BC438), and BGO are from: "Characteristics of Scintillators for Well Logging to 225 °C" by C.M. Rozsa, *et al*. prepared for the IEEE Nuclear Science Symposium, San Francisco, October 1989. The PMT is held near room temperature. The full text is available at www.detectors.saint-gobain.com. The temperature response for BrilLanCe 380 ($LaBr_3$:Ce) and BrilLanCe 350 ($LaCl_3$:Ce) were measured in mid 2005 in high temperature packages, again with the PMT held isothermally near room temperature. To overlay the temperature curves from these two eras, the relative pulse heights for BrilLanCe 380 ($LaBr_3$:Ce) and BrilLanCe 350 ($LaCl_3$:Ce) were measured by comparing 1" diameter x 1" long crystals in low temperature packages to NaI(Tl) of the same size also in a low temperature package. This result placed the curves at 130% and 75% of NaI(Tl) at room temperature, as shown. As mentioned in the text, later data shows BrilLanCe 380 (LaBr3:Ce) to be over 160% of NaI(Tl) at room temperature for more recent data all taken in high temperature Ti-sapphire packages. Other groups report similar room temperature light output, 160% NaI(Tl) or greater.

[8] G. Bizarri, J. T. M. de Haas, P. Dorenbos, and C. W. E. van Eijk, *Phys. Stat. Sol. (a)* **203**, No. 5, R41– R43 (2006)

[9] For details search High Count Rate or Note 519 for the work by Vladimir Solovyev at www.detectors.saint-gobain.com

[10] Table of Isotopes, Eight Edition. Richard B. Firestone, Virgina S. Shirley, Ed. John Wiley & Sons (1996)

[11] B.D. Milbrath, R.C Runkle, T.W. Hossbach, W.R. Kaye, E.A. Lepel, B.S. McDonald, L.E. Smith. Nuclear Instruments and Methods in Physics Research, Section A, 547, 2-3, pp 504-510, August 1, 2005

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