

Initial Performance Testing of Srl Gamma Spectroscopy Scintillators and Comparison to Other Improved-Resolution Detectors for Typical Health Physics Applications

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Abstract

Europium-doped Strontium Iodide (Srl) scintillation gamma detectors have only recently become commercially available. Srl has very high light output ranging from 80,000-100,000 ph/MeV and energy resolution as good as 2.9% at 662 keV and 14.8% at 32 keV. The high density of 4.6 g/cc in combination with a high effective Atomic number and the absence of internal activity make Srl a very attractive sensor material for spectroscopic radiation monitoring devices. This new entrant into the field of commercially available gamma spectroscopy detectors offers improved energy resolution over NaI, without the elevated background of LaBr. While CZT and HPGe detectors have better resolution, the small available size of CZT and the cooling requirement of HPGe limit their usefulness in portable applications. Of these scintillators, Srl2 has the lowest detection limits when compared to other equal-size scintillation detectors, especially at low energies where U and Pu are of interest.

Introduction

Strontium Iodide was originally explored as a potential enhanced-resolution radiation detector by R. Hofstadter in 1968. He expected promising results however the doping methods and crystal purity available at that time did not give satisfactory results. Strontium Iodide was largely ignored until ~2008 when scientists at Lawrence Livermore National Laboratory resurrected that effort and headed the research into enhanced-resolution scintillation detectors. That effort has now led to the availability of large-size good quality commercially available scintillators made from Europium-doped Strontium Iodide (Srl2:Eu), hereafter abbreviated Srl.

Table 1 shows some of the characteristics of various gamma sensors. The features that make Srl attractive for gamma spectral measurements include:

- High light yield for better statistics
- Favorable wavelength for better matching to Photomultiplier Tubes
- High Effective Atomic Number for better stopping of gamma radiation
- Absence of internal radioactivity which limits the usefulness of scintillators containing La, due to the naturally radioactive La-138 it contains.

Table 1 Properties of various gamma detectors

Detector Material Properties				
Material	Density	Z-Effective	Light Yield	Peak Wavelength
	g/cc		photons/MeV	nM
NaI(Tl)	3.67	32	38,000	415
Srl(Eu)	4.59	48.2	80,000	435
LaBr(Ce)	5.08	40.5	70,000	380
CeBr	5.18	40.8	68,000	371
CLYC	3.31	25.3	20,000	370
CZT	5.78	42.4	-	-
Ge	5.36	32	-	-

Another new scintillator coming out of the LLNL research efforts is Cs2LiYCl6 – thankfully commonly abbreviated as CLYC. It is also an enhanced resolution detector with also the ability to detect neutrons.

A cursory evaluation of the detector material properties would also seem to favor LaBr and CeBr scintillators. Both have been commercially available for about 10y. LaBr has demonstrated very good energy resolution – in the 3-3.5% range, while CeBr has generally been in the 4-4.5% range. But the major drawback of LaBr is the internal radioactivity from the naturally occurring La-138. The graph in Figure 1 shows two similar size detectors in a shielded cave. The LaBr scintillator has a background nearly 10x higher than the Srl. This is due to La-138. The higher energy peaks are due to residual radium impurities that are apparently quite difficult to remove. CeBr, NaI, and CLYC will all have background levels similar to that shown for Srl.

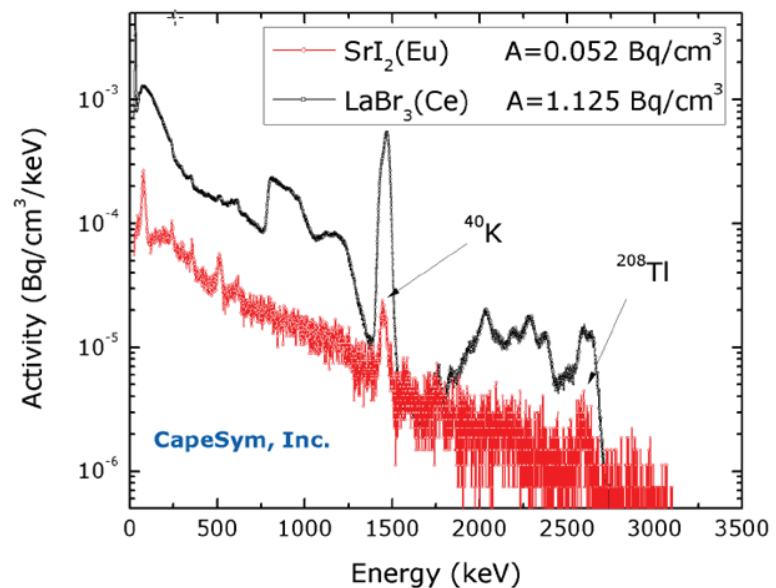


Figure 1 Shielded background for Srl and LaBr detectors of similar size

Theoretical Calculations of Comparative Efficiency

To obtain an early prediction of the potential performance of Srl, efficiency calculations were performed using the Mirion Technologies [Canberra] ISOCS software. Hypothetical perfect “radioactive sources” that were 51mm diameter by 51mm in length at 100mm from a reference detector were modeled. Each “radioactive source” had the material composition and density of the tested scintillator. The modeled “radioactive sources” were: Srl, NaI, LaBr, CeBr, CLYC, CZT, and Ge, all common and available radiation detector materials. The ISOCS computed efficiency vs. energy is the product of the emission fraction from the “radioactive source”, a geometrical factor, and the full energy peak absorption fraction of the detector. But since the detector is the same in all of these cases, ratio of the NaI emission fraction to the Srl emission fraction is equivalent to the ratio of the Srl efficiency to the NaI efficiency for that particular geometry. This is essentially the same concept as the Reciprocity Principle in radiation dosimetry where the source and the detector can be interchanged without changing the absorbed dose.

The following graphs in Figure 2 show the expected gamma peak efficiency of all of these “perfect” hypothetical detectors – as compared to the performance of NaI. The energy span is from 10 keV to 7000 keV. All of these comparisons assume the same size detector. The left graph shows all of the data points for all of the materials. The right graph is an expanded view of the scintillators to better visualize the low energy performance.

Features that can be observed in these plots include:

- The much higher efficiency compared to NaI at 10-40 keV, due to the low Z-effective of NaI. CLYC is the exception as it has a low Z-effective.
- The higher efficiency of all of the scintillators [except CLYC] at all energies, due to the low density of NaI.
- Srl has the best efficiency in the energy range of 50-250 keV, due to the high Z-effective. The low Z-effective of CLYC and Ge is why their relative efficiency is low in the 50-300 keV range.

- LaBr and CeBr have the best efficiency at >250 keV due to their high density.
- CZT shows the highest relative efficiency due to the highest density and high Z-effective and would make an excellent spectroscopy detector if it were ever available in large sizes.

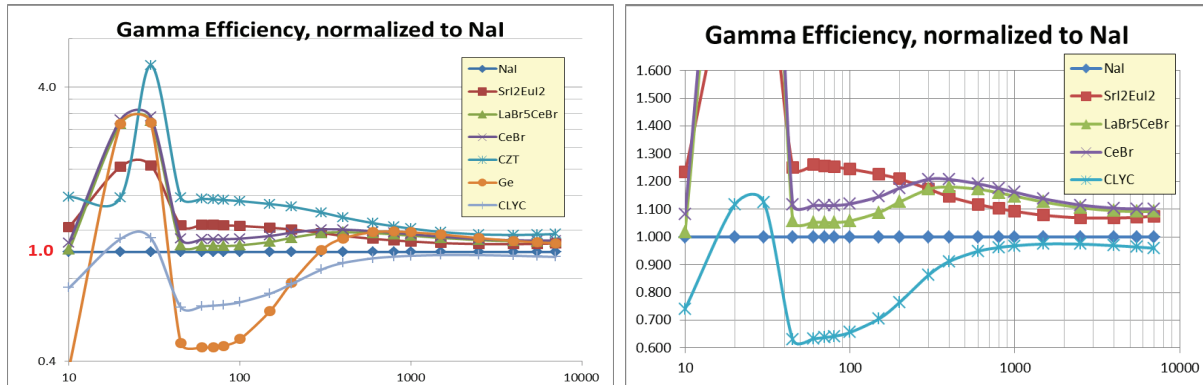


Figure 2 Gamma efficiency for equal size detectors, all normalized to NaI efficiency. The graph on the right is a vertically expanded scale of the graph on the left, and only for the enhanced resolution scintillators discussed in this document.

Equipment Tested

Two different sized Srl detectors were obtained, one is 38mm diameter and 38mm long, the other is 46mm diameter and 26mm long. Figure 3 shows these two detectors, along with a 3x3 NaI LED-stabilized detector and a 1cc CZT detector-MCA. The 38x38mm Srl detector is coupled to at 10-stage PMT which mostly explains why it is longer than the 46x46mm detector which is coupled to an 8-stage PMT.

The Mirion Osprey MCA is shown on the 38x38mm detector and was used in all of the scintillator tests.

Both of the Srl scintillators have essentially the same volume and therefore are approximately the same price. However the aspect ratio [ratio of diameter to thickness] of the 46x26mm detector gives it an efficiency advantage for most all geometries. This is the advantage long seen with the high aspect ratio Canberra BEGe detectors as compared the more traditional co-axial Ge detectors.



Figure 3 Detectors and MCA used in this document.

An ISOCS characterization was performed for both SRI detectors which allows us to quickly do efficiency calculations. Below in Figure 4 on the left is an energy-efficiency calibration comparison for a typical

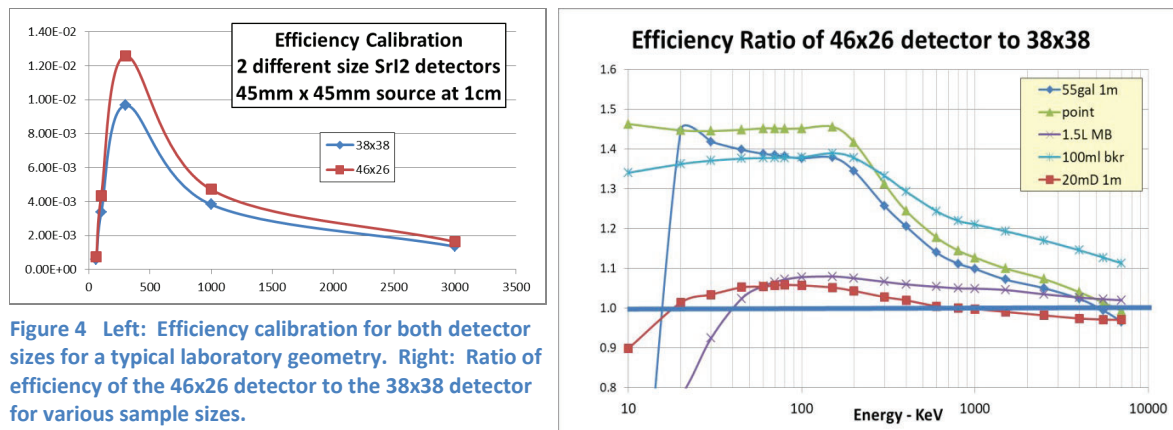


Figure 4 Left: Efficiency calibration for both detector sizes for a typical laboratory geometry. Right: Ratio of efficiency of the 46x26 detector to the 38x38 detector for various sample sizes.

laboratory sample geometry. The 46x26mm detector has up to 25% higher efficiency.

The graphic on the right of Figure 4 explores this relationship for a wide range of other geometries. For all geometries where the source is predominately in front of the detector the efficiency for the high aspect-ratio 46x26 detector is always better up to very high energies. This true for both laboratory-type samples [100ml beaker, point-like sources] and common InSitu sources [200 liter drums]. Here, the 46% larger front surface area of the 46x26 detector more than makes up for the 32% shorter length. For other geometries where the source is both in front and at the sides [Marinelli Beakers, very large diameter sources] the 46x26 detector is still better at most all energies for most all gamma sources.

Tests and Results

We first explored the properties of SRI and CLYC in 2010-12 when executing a DHS contract on the development of a hand-held direction sensing spectrometer. The small crystals [25x25mm] available at that time had respectable resolution, typically 3.5-4% FWHM. But they all had high energy tailing, as shown in the top graphic of Figure 5. This is one of the better crystals from that project. While the low energy peaks were quite symmetric, the high-energy side of the Cs-137 peak is wider than the low energy side. Larger detectors and higher energies show even more high-energy tailing. This makes it difficult for the peak fitting software to analyze the spectra, and

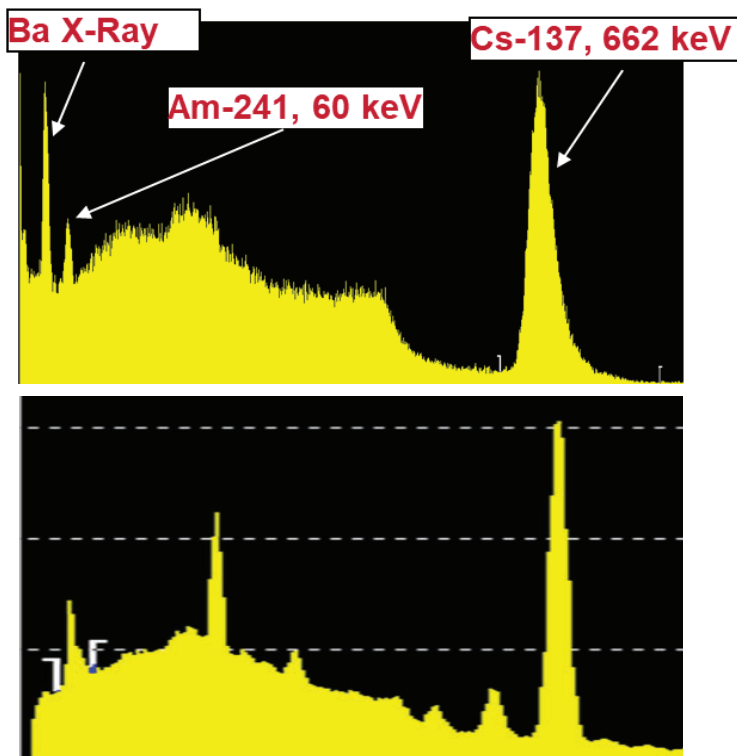


Figure 5 Top: 25x25mm SRI from 2011 DHS project. Bottom: detector from this study. Both have linear vertical counts scale and the same horizontal energy scale.

can mask nearby higher energy peaks. Since older Ge detectors frequently had low-energy tailing due to charge loss, there are features in the peak fitting software to handle that; but no such features for high-energy tailing.

Fortunately today, this problem has mostly disappeared due to advances in material purity, better doping, better quality crystals, and better packaging. The bottom graphic in Figure 5 shows one of the detectors tested here – on the same energy scale. A thorium source is used in addition to Cs-137 which generates the peaks between 60 keV and 662 keV. Notice that the Cs-137 peak is narrower [better resolution] and no observable high energy tailing. At high energies [e.g. 2614 keV] there still is some noticeable high energy tailing but fortunately there are fewer peaks up there, and the net peak area fitting process is still accurate.

The prime performance parameter is energy resolution – FWHM. Obtaining the best energy resolution requires setting the many parameters in the MCA signal processing chain at their optimum values. Due to the rather long decay time constant of the Srl scintillation light pulse, the MCA must integrate the signal long enough to capture most all this light – the more light captured, the higher the peak amplitude and the better the resolution. The Osprey Digital Signal Processing amplifier has two parameters that primarily affect this – Flat Top [FT] time and Rise Time [RT]. The FT time has a maximum limit of 3 uSec, and the RT has a maximum time of 5 uSec. It was quickly determined that these were not long enough for Srl. So a special version of the Osprey firmware was created that allows the Osprey RT to be increased to 50 uSec.

The test rig that was used is shown in Figure 6. A Cs-137 source is at the right with the radiation label. The detector is pointed to a case of Tungsten welding rods that have 2% Thorium. The Thorium and



Figure 6 Test geometry used for these initial set of tests. Thoriated tungsten welding rods under detector and Cs-137 source.

daughters have many nice gammas all the way up to 2614 keV. And since the thorium is intimately mixed with the tungsten, the captured gammas cause the emission of the thorium X-ray which is at 60 keV – the same energy as the important nuclide of Am-241. This test rig allows the examination of 7 energies that are all rather cleanly separated from near-by gammas. The Cs-137 source is there since that is commonly how performance of scintillators is referenced. The construction of the Cs-137 source is such that the 32 keV Ba X-rays do not escape.

For these tests the FT was set at the maximum of 3 uSec, and the RT was varied. At each new RT the gain was adjusted to keep the keV/channel value constant.

The graph in Figure 7 shows how the FWHM in keV changes as the RT is changed. As the RT is increased the FWHM decreases. The improvement in FWHM is much more significant at high energies than at low energies.

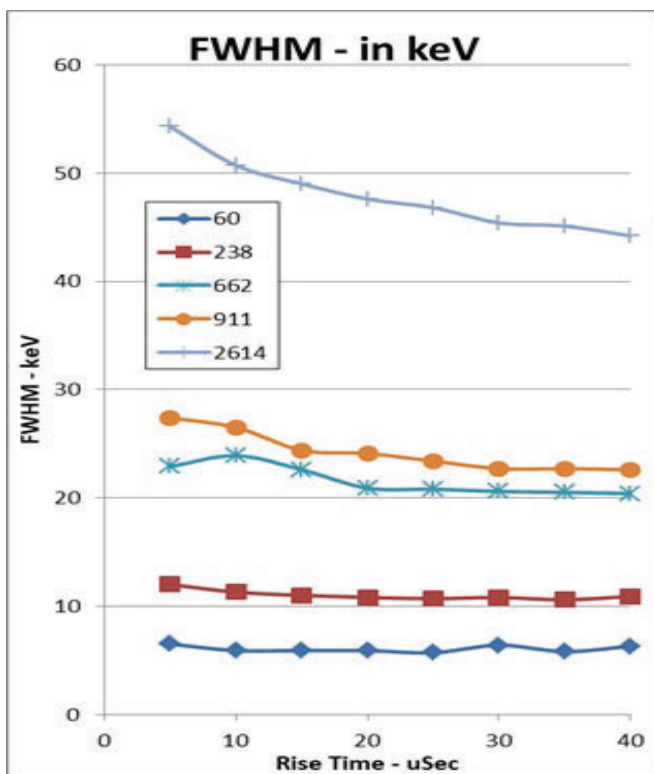


Figure 7 FWHM for 5 energies from 60 to 2614 keV as the Rise Time is varied.

One might be tempted to operate at the highest RT setting. However as the signal integrating time is increased, the number of signals that can be processed per second decreases. Figure 8 shows that even with no change in total count rate, the % DeadTime is doubled when going from 5 uSec RT to 25 uSec RT.

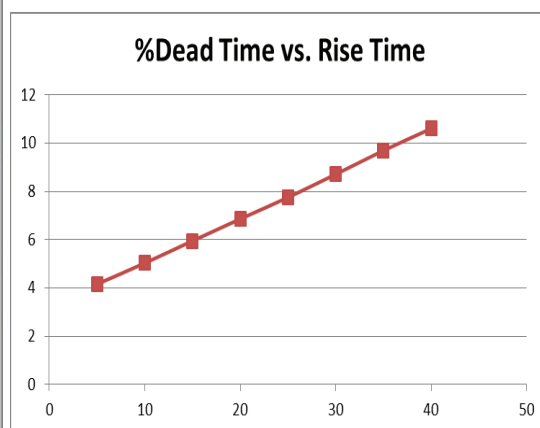


Figure 8 Percent dead time [vertical axis] as the Rise Time is changed [horizontal axis].

Another test that was performed to examine the impact of increasing count rates in the detector. With the detector still sitting on top of the Thorium welding rods, the Cs-137 source was moved stepwise closer to the detector. The total spectrum counts per second was recorded along with the 662 keV Cs-137 peak centroid and %FWHM. This is shown in Figure 9. The count rate was changed from around 100 to around 15,000. The gain was quite stable, only changing a few channels [less than 3 keV] which is insignificant given the approximately 20 keV FWHM. There was a slight increase of 2-3 keV in the FWHM value, not a very large value either. But the % deadtime did increase from 6% at 132 cps to 41% at about 15,000 cps.

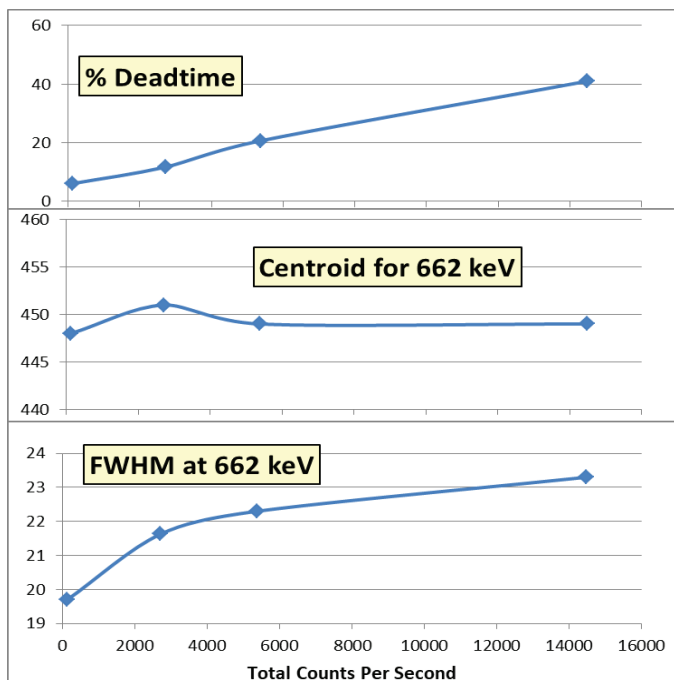


Figure 9 Change in %DeadTime, peak centroid, and FWHM as the total CPS was increased.

With modern digital signal processing MCAs like the Osprey, high % dead times are not

necessarily a problem, but it would seem prudent to carefully examine the accuracy of the live time before pushing Srl detectors much beyond the 20 - 25,000 cps range.

This does point out one of the shortcomings of Srl - the long decay time. Other scintillators, notably LaBr have very short decay times, and are much better suited for high count rates, up to where limited by the performance of the PMT. For similar reasons, Srl scintillators are now as well suited for measurement situations where accurate signal timing is required.

FWHM performance of Srl and other scintillators

The following graphs in Figure 10 show the measured FWHM from these Srl tests and comparisons with the other scintillators. In comparison to NaI, all of these enhanced resolution scintillators are better, with Srl being the best, just slightly better than LaBr. The graphic on the left is the high energy range – up to 1400 keV, while the graphic on the right is an expanded view of the same data in the 50-300 keV range.

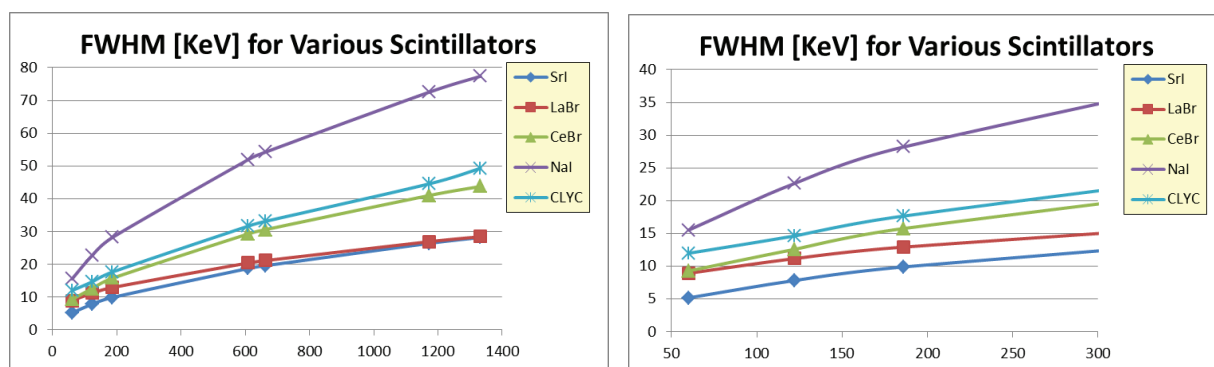


Figure 10 FWHM in keV for various scintillators as a function of gamma energy. The graph on the right is a expanded view of the graphs on the left.

The low energy range up to ~300 keV has some important nuclides that are commonly required to be measured. These include Am241, U-235, U-238, Pu-239, and most all nuclear medicine radionuclides. The expanded range chart on the right of Figure 10 clearly shows the advantage of Srl to separated peaks in this crowded low energy area. This performance can be even improved with better PMTs and better matching of signal processing electronics. The graphic in Figure 11 shows the same detectors with these improved features, as measured by the Srl supplier, CapeSym. The upper line is the same data as in Figure 10.

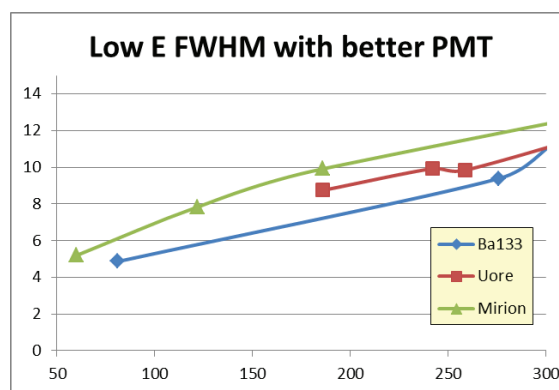


Figure 11 FWHM for low energy nuclides when measured with higher performance PMTs

Temperature Effects on Srl Scintillators

While there is a small dependence on light output from all scintillators as a function of temperature that variation is overwhelmed by the temperature dependence of the PMT. Figure 12 shows data from CapeSym on the variation of centroid position [gain] and FWHM as the detector and similar PMT are going through a temperature cycle test.

The detector and PMT were in an environmental chamber. The temperature was changed from -25 degC to +55 degC. The right-hand scale shows the FWHM change with the values expressed as Percent at 662 keV. While the FWHM does increase slightly as the temperature is increased, it only changes very little around the nominal room temperature value of 3.2% [21 keV]. These changes are well within the ability of the Genie gamma spectral analysis software to handle.

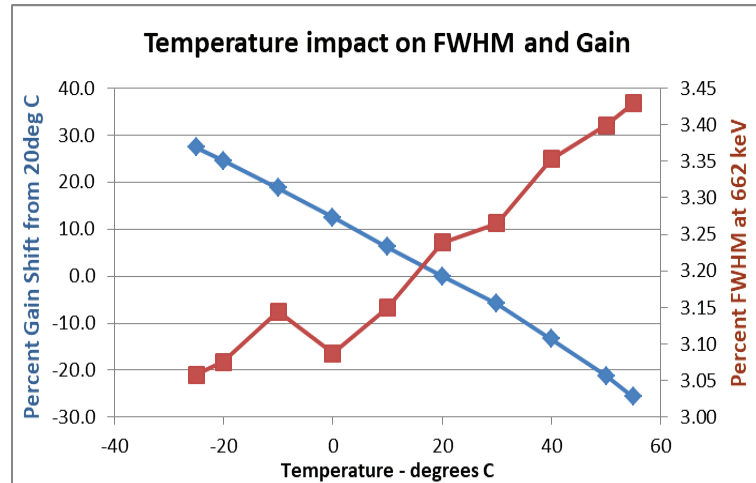


Figure 12 Temperature sensitivity of FWHM and Gain.

However the gain changes very significantly going from 25% higher at -25 degC to 30% lower at +55 degC. The typical parameters in the Genie gamma spectral analysis software will typically correctly assign the nuclide identity to a peak as long as it is within 1 FWHM of the expected position. For these enhanced resolution scintillators the reduced FWHM requires more gain stability than the common NaI detector. The “within 1 FWHM” requirement means for proper nuclide identification performance, the temperature must be kept within +/- 4.5 degC [8 degF] of the energy calibration temperature.

Alternately, and preferably, an automatic gain stabilization method should be implemented, such as the Canberra LED stabilization process that we have already implemented on NaI, LaBr, and CeBr scintillators.

Minimum Detectable Activity estimations.

This last section attempts to predict the performance of Srl to measure low activity samples and objects. Since there are an infinitely wide variety of sample detector geometries to choose from and since NaI is the most common scintillator, it was decided to simplify this section by computing these MDAs relative to the MDA of NaI.

Factors affecting MDA are:

- Detection Efficiency: the higher the detection efficiency the lower the MDA. The MDA is inversely proportional the detection efficiency.
- FWHM: the lower the FWHM the lower the background under the peak and the lower the MDA. The MDA is proportional to the square root of the FWHM
- Background: the lower the background under the peak and the lower the MDA. For equal size detectors [without any internal contamination], the background is approximately proportional to the mass of the detector. The MDA is proportional to the square root of the mass of the detector.

Combining the relationships above, the MDA is proportional to:

$$\frac{\sqrt{(\text{detector mass}) \times (\text{FWHM})}}{\text{peak efficiency}}$$

Using this relationship a value for each of the enhanced resolution detectors was calculated and ratio to the computed value for NaI was determined.

For this exercise we “assumed” that somehow the natural radioactivity of LaBr could be eliminated.

Table 2 shows the numerical data, and the Figure 13 graphic shows it visually.

For all energies these enhanced resolution scintillators have lower MDAs than NaI, with the exception of CLYC at low energies.

For all energies, Srl has the lowest MDA. Srl is slightly better than the [non-existent non-radioactive] LaBr at medium to high energies and significantly better than all of the scintillators at the important low-medium energy range.

Conclusion

Srl has been tested and shown to have the best energy resolution of the group of enhanced-resolution detectors.

This energy resolution combined with the low internal radioactivity background make it best currently available enhanced resolution detector for low MDAs and optimum nuclide identification.

A high-aspect ratio size detector shape has been shown to have up to 25% higher efficiency and therefore 25% lower MDA than a conventional shape where the diameter is equal to the height of the detector.

Limitations have been found in the current parameters available for adjustment in the Osprey digital MCA and software fixes have been prototyped and tested.

Table 2 Computed Minimum Detectable Activities of various enhanced resolution scintillators as compared to NaI.

Energy keV	Relevant Nuclides	Srl	*LaBr* clean	LaBr today	CLYC	CeBr	NaI
60	Am-241	0.51	0.85	2.55	1.32	0.79	1
122	Co-57	0.53	0.77	2.31	1.13	0.78	1
186	U-235, Ra-226	0.55	0.71	2.13	1.01	0.76	1
609	Radium	0.6	0.63	1.89	0.78	0.75	1
662	Cs-137	0.6	0.63	1.89	0.78	0.75	1
1173	Co-60, U-238	0.62	0.63	1.89	0.77	0.77	1
1332	Co-60	0.62	0.63	1.89	0.78	0.78	1

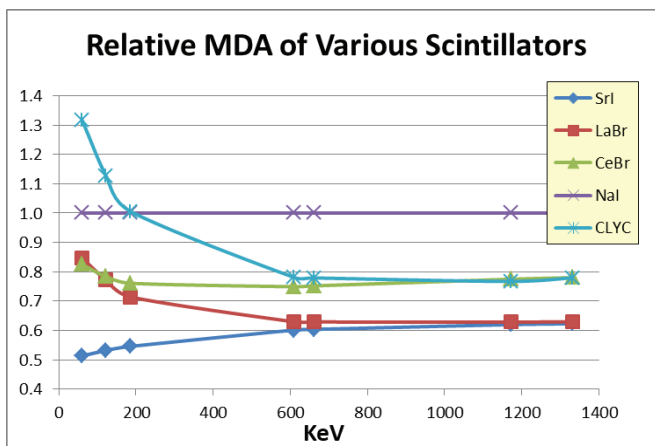


Figure 13 The relative MDA [NaI = 1] of various enhanced resolution scintillators. Note: the LaBr in this graph assumes no internal radioactivity.

Limitations have also been identified in the temperature response from the scintillator-PMT combination. This probably not an issue for laboratory sample counting applications, but would require some means to limit temperature fluctuations or to stabilize the gain for field applications.

This combination of computations and test results has shown that Srl scintillation detectors will have the lowest MDA of these enhanced resolution detectors, if all detectors are the same size.

Srl scintillators should be especially useful in low energy measurements of low activity samples.