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ABSTRACT

Development of the Europium-doped Strontium Iodide scintillator, $SrI_2(Eu^{2+})$, has progressed significantly in recent years. $SrI_2(Eu^{2+})$ has excellent material properties for gamma ray spectroscopy: high light yield (>80,000 ph/MeV), excellent light yield proportionality, and high effective atomic number (Z = 49) for high photoelectric cross-section. High quality 1.5" and 2" diameter boules are now available due to rapid advances in $SrI_2(Eu)$ crystal growth. In these large $SrI_2(Eu)$ crystals, optical self-absorption by Eu^{2+} degrades the energy resolution as measured by analog electronics, but we mitigate this effect through on-the-fly correction of the scintillation pulses by digital readout electronics. Using this digital correction technique we have demonstrated energy resolution of 2.9% FWHM at 662 keV for a 4 in^3 $SrI_2(Eu)$ crystal, over 2.6 inches long. Based on this digital readout technology, we have developed a detector prototype with greatly improved radioisotope identification capability compared to Sodium Iodide, NaI(Tl). The higher resolution of $SrI_2(Eu)$ yields a factor of 2 to 5 improvement in radioisotope identification (RIID) error rate compared to NaI(Tl).

Keywords: scintillators, gamma ray detection, energy resolution, radioisotope identification

1. INTRODUCTION

To complement radiation detection equipment widely deployed at national borders and other points of entry, handheld and wearable personal radiation detectors are needed by soldiers and first-responders. For these devices, gamma scintillators are needed with high resolution, large volume, and low cost. NaI(Tl) is the standard scintillator material for this application: no intrinsic radioactivity, as low as 6% resolution at 662 keV (typically 7%), up to 4000 cm^3 crystal volumes, and typically $5 per cm^3. $LaBr_3(Ce)$ improves on NaI(Tl) in performance, but its intrinsic radioactivity is problematic for low-background measurements, and it cannot match NaI(Tl) in crystal volume or material cost: 3% resolution, 400 cm^3 boules, and $150 per cm^3 for common sizes (i.e. 1.5" x 1.5" cylinder). The Europium-doped Strontium Iodide scintillator, $SrI_2(Eu)$, has the potential to combine the best traits of NaI(Tl) and $LaBr_3(Ce)$, and it has excellent material properties for mobile gamma ray spectroscopy, as described in Table 1 [1,2].

Recent gamma nonproportionality measurements have also shown close agreement with models based on electron nonproportionality data, both in gamma light yield and energy resolution, further supporting the high intrinsic performance of $SrI_2(Eu)$. With high quality 1.5" and 2" diameter boules now available, see Figure 1, we have begun developing a prototype $SrI_2(Eu)$ handheld radiation detector to demonstrate the capabilities of $SrI_2(Eu)$ in a modern platform. In long $SrI_2(Eu)$ crystals, optical self-absorption by Eu^{2+} degrades the energy resolution as measured by analog electronics, but we mitigate this effect through on-the-fly correction of the scintillation pulses using Bridgeport Instruments digital readout electronics. Using this digital correction, we have demonstrated energy resolution of

2.9% FWHM at 662 keV for a 4 in^3 $SrI_2(Eu)$ crystal, over 2.3 in long. Using measured data from $SrI_2(Eu)$ and NaI(Tl), we built full spectral response models of these detectors to simulate their response to a wide range of sources, including background and complex radioisotope mixtures.

Table 1. Properties of $SrI_2(Eu)$ compared to $LaBr_3(Ce)$

Property	$LaBr_3(Ce)$	$SrI_2(Eu)$
Melting point	783°C	538°C
Handling	Easily cleaves	Resists cracking
Density	5.1 g/cm^3	4.6 g/cm^3
Effective atomic number	47	49
Light yield	60,000 Ph/MeV	85,000 Ph/MeV
Proportionality contribution to resolution at 662 keV	~2.0%	~2.0%
Decay time	30 nsec	1.5 µsec
Radioactivity	^{138}La self- activity	None
Hygroscopic / air sensitive?	Very	Very
Gamma absorption (2x3", 662 keV)	22%	24%
Resolution at 662 keV (calculated best achievable)	2.5%	2.3%

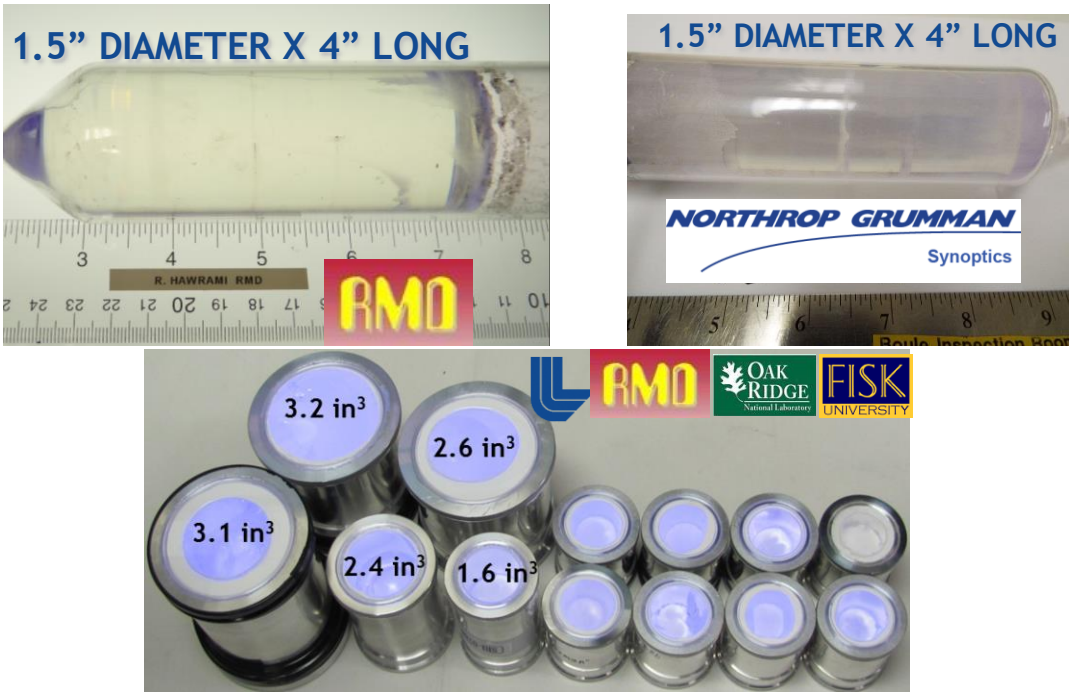

Figure 1. (Top left) RMD grown boule, **(Top right)** Northrop Grumman grown boule, **(Bottom)** array of LLNL packaged crystals from RMD, Fisk University, and Oak Ridge National Laboratory.

2. EQUIPMENT AND METHODS

$SrI_2(Eu)$ crystals are now being grown routinely around the world, at RMD, Oak Ridge National Laboratory, Fisk University, Northrop Grumman, Hellma (Germany), and Union Materials (Japan), among others [3-5]. Crystals from

Union Materials and some crystals from RMD are sold encapsulated by the company. When bare crystals are received, they are polished and encapsulated in aluminum cans at LLNL [6]. All measurements and modeling presented here are performed at LLNL. Electron nonproportionality measurements are performed in the Scintillator Light Yield Nonproportionality Characterization Instrument (SLYNCI). Gamma nonproportionality measurements are performed with an Ortec 113 preamplifier, a Tennelec TC243 spectroscopy amplifier (12 μs shaping time), and an Amptek 8000D MCA. All measurements are performed with the SrI_2(Eu) package optically coupled to a Hamamatsu R6231-100SEL super bialkali PMT (35% quantum efficiency at 350 nm). Gamma nonproportionality measurements utilize 10 sealed radioactive sources and an Amersham variable x-ray source, in which ^{241}Am alpha particles impinge on a thin metal film to generate characteristic x-rays. The Bridgeport Instruments electronics used for the handheld detector prototype will be described in the next section. Detector modeling and radionuclide simulations are performed with GADRAS (GAmma Detector Response and Analysis Software) [7]. Radioisotope identification comparisons are performed using RNAK software (Radionuclide Analysis Kit), developed by Karl Nelson, LLNL [8].

3. RESULTS AND DISCUSSION

3.1 Nonproportionality characterization of SrI_2(Eu)

We are working to determine the maximum potential performance of SrI_2(Eu) and better understand the physics behind its scintillation mechanism. We use the SLYNCI to compute the electron nonproportionality based on Compton coincidence measurements. Using these measurements, we can determine the contributions from the many underlying effects comprising to scintillation nonproportionality using our well validated nonproportionality model [9,10]. From this model, we can calculate the estimated gamma nonproportionality response, different than the electron nonproportionality. We measured sources with gamma and x-ray energies spanning 4 keV to 2.6 MeV. Obtaining accurate measurements of energy resolution and peak centroid is hindered primarily by overlapping gamma photopeaks; at energies below 90 keV, this problem is exacerbated by poorer photon statistics (poorer resolution) and closely spaced x-ray energies, with peaks coming from both absorbed x-rays and strontium and iodine x-ray escapes (due to shallow x-ray penetration). Despite these challenges, the measured gamma nonproportionality data fits well to the performance predicted by our nonproportionality model, with only a slight overestimation of the rising Onsager component [11], as shown in Figure 2. The measured resolution also fits very well to modeled performance, with slightly poorer resolution at low energies likely due to optical effects in these near-surface interactions.

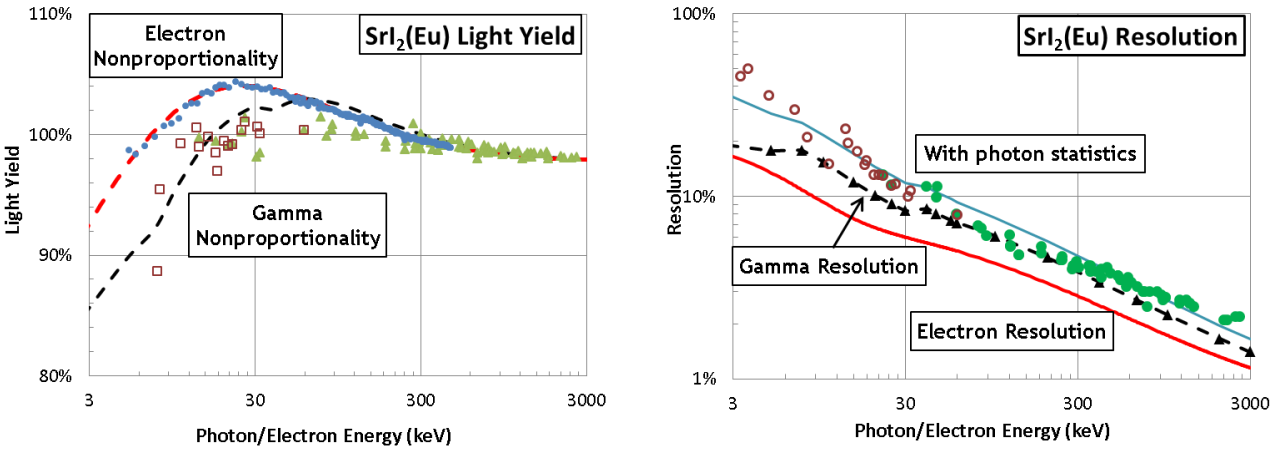

Figure 2. Electron and gamma nonproportionality with modeled performance shown as dashed lines and measured data shown with markers. Ideal light yield nonproportionality would be a flat line at 100% light yield. In gamma non-proportionality measurements, two different crystals were used: the first set of measurements (solid, green markers) used a 7 cm^3 tapered cylinder of SrI_2(3%Eu) packaged at LLNL, the second set of measurements (open, red markers) used an RMD packaged 12 cm^3 SrI_2(3%Eu) right cylinder with a thinner aluminum shell and less reflector material to yield a stronger response at low energies. **(Left)** light yield, **(Right)** energy resolution.

3.2 $SrI_2(Eu)$ handheld prototype development

With crack-free SrI_2 boules regularly available, we are developing a prototype $SrI_2(Eu)$ handheld instrument to demonstrate the potential performance of $SrI_2(Eu)$. To realize the full potential of $SrI_2(Eu)$ crystals, we need to account for "light-trapping", a type of optical scatter that increases effective decay time. This effect is caused by overlap between the absorbance and radioluminescence of Eu^{2+}, leading to successive red shifting of scintillation photons in the re-absorption and re-emission process as they traverse the crystal [13]. In crystals longer than approximately 1 inch, "light-trapping" causes the amount and uniformity of light collected by the PMT to vary for each gamma interaction of a given energy depending on the location of the interaction, as shown in Figure 3.

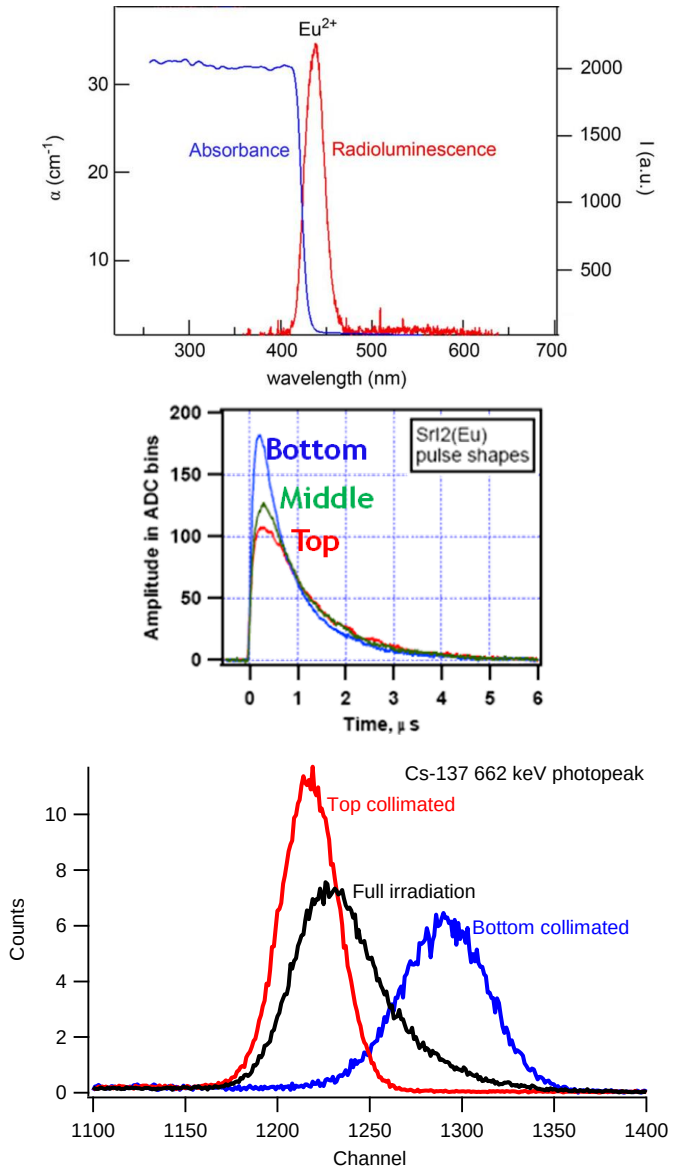

Figure 3. (Top) Absorption and beta-excited radioluminescence spectra of $SrI_2(Eu)$ with small overlap in bands leading to "light-trapping". **(Middle)** Scintillation pulses showing decreased magnitude and longer decay time for interactions further from the PMT. **(Bottom)** Cs-137 spectra showing the 662 keV photopeak. Top collimated measurements produce the lowest but most uniform light yield. Bottom collimated produces the highest but most non-uniform light yield, since some photons go directly to the PMT and others traverse the full length of the crystal before reaching the PMT. Full irradiation combines these components leading to degraded resolution and a non-Gaussian tailing of the photopeak to higher energy.

To account for "light-trapping" and realize the full potential of $SrI_2(Eu)$ detectors, we use digital readout electronics from Bridgeport Instruments. Using an on-the-fly correction factor applied through the onboard FPGA, we can correct for the scintillation pulse lengthening caused by "light-trapping". We have tested this technique using many crystals up to 2" long, in addition to a 1.5" diameter x 2.3" long crystal from Northrop Grumman. Our algorithm on the FPGA is optimized to yield the best possible resolution at 662 keV, but this correction improves energy resolution at all gamma energies, as shown in Figure 4. The spectra in Figure 5 show in more detail the resolution improvement going from 12 µs analog shaping time to 30 µs digital shaping time with pulse shape correction.

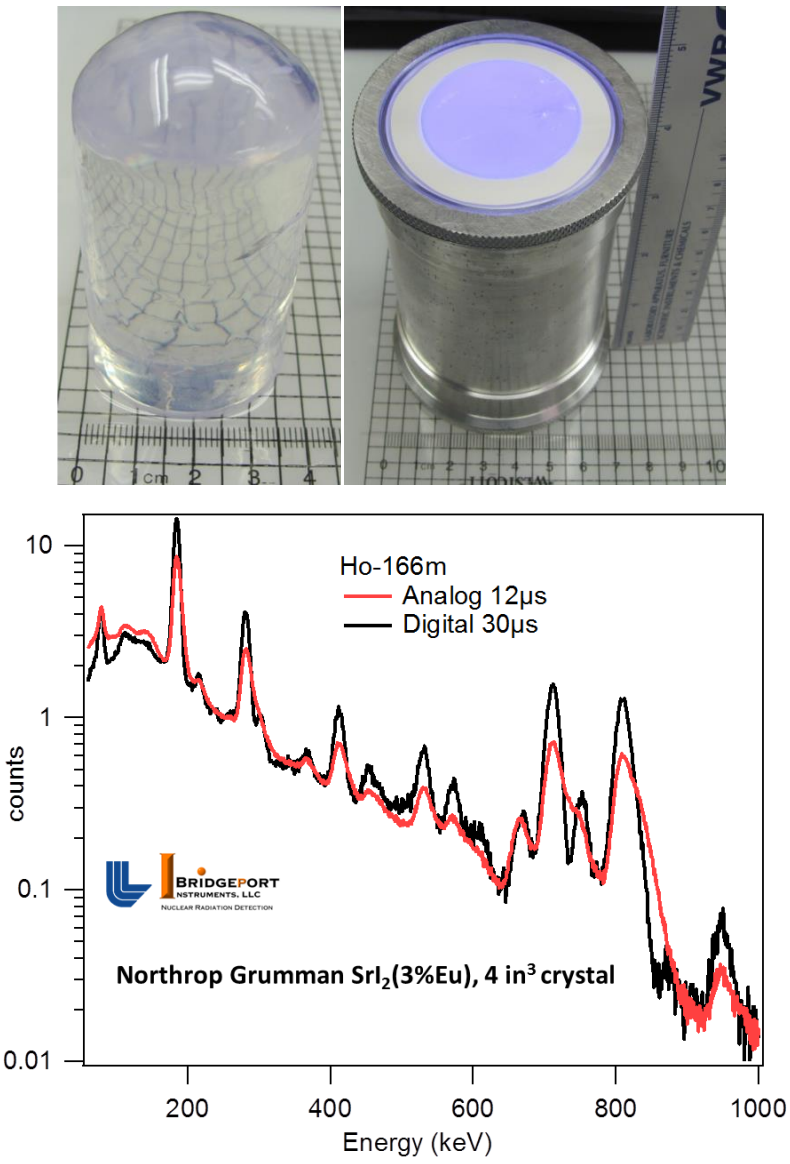

Figure 4. (Top) Northrop Grumman grown $SrI_2(Eu)$ polished and packaged at LLNL. 4 in^3 volume, 1.5" diameter x 2.3" length. **(Bottom)** Ho-166m spectra showing the improvement in resolution going from analog electronics with 12 µs shaping time to digital electronics with 30 µs shaping time and digital pulse shape correction.

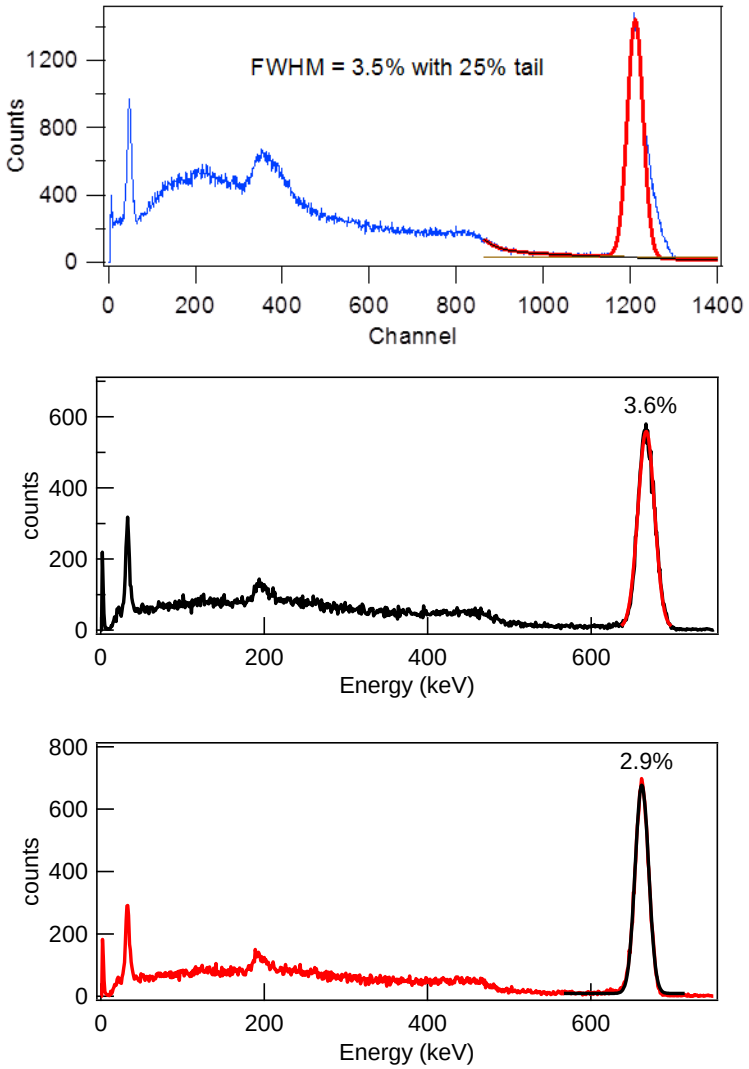

Figure 5. (Top) Analog 12 µs shaping time yields 3.5% FWHM energy resolution at 662 keV with 25% of the photopeak counts above 662 keV falling outside of the Gaussian peak fit. **(Middle)** Digital 30 µs shaping time captures the full scintillation pulse, removing the high energy tail but only gives 3.6% FWHM energy resolution at 662 keV due to "light-trapping". **(Bottom)** Digital 30 µs shaping time with pulse shape correction improves the resolution to 2.9% FWHM with no high energy tailing, comparable to the best performing 1" crystals.

The prototype handheld detector offering this performance is shown in Figure 6. The system can accommodate an 8 in^3 crystal, and currently contains a 1.5" x 1.5" LLNL-packaged $SrI_2(Eu)$ crystal. The crystal is held in place by hard foam and optically cemented to a Hamamatsu 6231-100SEL PMT. The PMT is read out through a Bridgeport Instruments digital PMT base (12 bit ADC, 20 MHz digitization, 1024 point waveform capture via FPGA, and less than 0.5 W operating power) and adjoining wireless system (embedded mini-computer, communication via WiFi 802.11b/g/n, and embedded battery for more than 8 hour operation). The system can be connected to any WiFi capable device and operated from a simple PHP interface. We are also developing custom software to take advantage of the modern interaction capabilities of an Android tablet interface. With feedback from field users of handheld detectors, we are integrating all of the desired capabilities and features into our interface. The main features include: spectrum display with zoom capability, instantaneous count rate meter, count rate versus time, and waterfall plots (full spectral heat map of gamma energy intensity versus time). We currently have a library of radionuclide gamma energies that can be overlaid on real-time data, and we plan to implement full spectral radioisotope ID.

Figure 6. Prototype handheld $SrI_2(Eu)$ detector system built in a collaborative effort between LLNL and Bridgeport Instruments, using Bridgeport Instruments digital readout electronics and an Android tablet display.

3.3 Detector modeling and radioisotope identification performance

To determine the potential improvement of a $SrI_2(Eu)$-based detector over existing NaI(Tl) handheld detectors, we utilize the detector and source modeling capabilities of GADRAS. To build an accurate model of the full spectral response of a detector, spectra that cover the full range of energies of the model are needed. The $SrI_2(Eu)$ spectra in Figure 7 are more than sufficient to model the response of this detector over the typical RIID range of 30 keV to 3000 keV. The sources with only one or two lines in a given energy range facilitate accurate reproduction of scattering and resolution parameters. Sources with many gamma lines over a range of energies provide excellent data for energy and nonproportionality calibration. Six spectra at a time are used to tune the model while the other spectra are used to verify the accuracy of the response model.

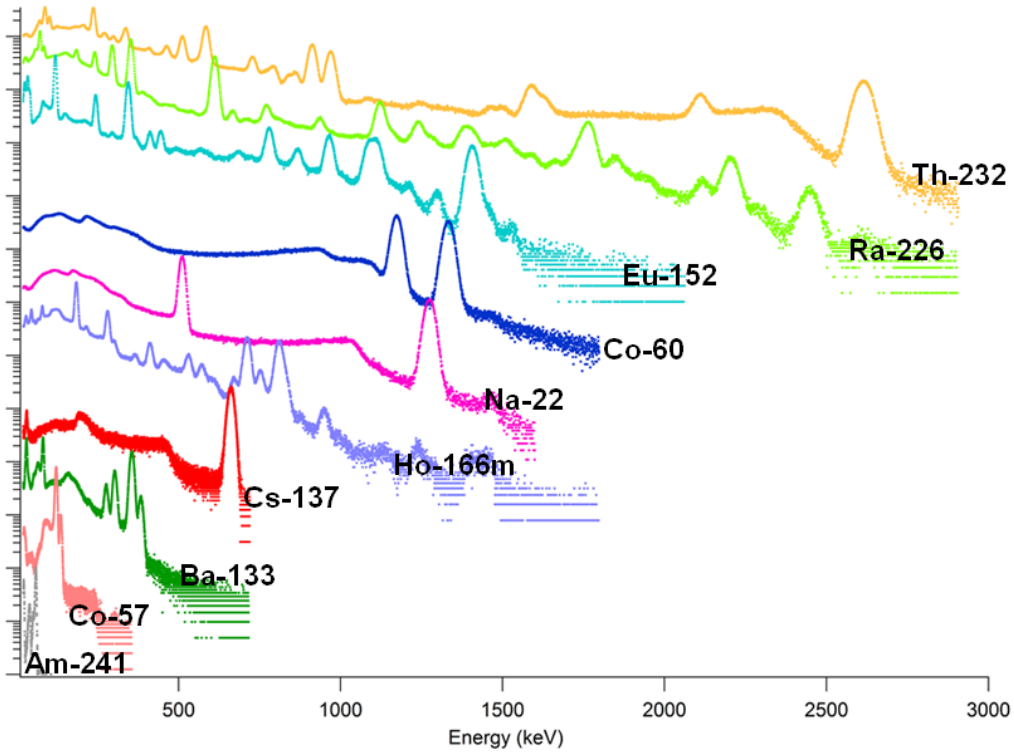

Figure 7. $SrI_2(Eu)$ spectra for 10 radioisotopes covering the range of energies from 30 keV to 3000 keV, sufficient to model the full spectral response of this detector.

The NaI(Tl) detector model was based on the GR-135+ handheld detector, containing a 1.5" x 2.2" NaI(Tl) crystal (3.9 in^3) and giving 6.8% FWHM energy resolution at 662 keV. The model of this detector does not reflect the performance of any GR-135+ software or algorithms, only the crystal itself. The $SrI_2(Eu)$ model was based on the readily available crystal size, 1" x 1" (0.8 in^3) and readily achievable resolution of 3.4% FWHM at 662 keV. This crystal was specifically chosen to have energy resolution a factor of 2 better than the NaI(Tl) detector. The currently available 4 in^3 $SrI_2(Eu)$ detectors with 2.9% FWHM energy resolution at 662 keV would perform significantly better and will be the subject of future comparative studies.

The parameters of the radioisotope identification study are described in Table 2. There were 21 common nuclides used. Each spectrum could contain 0 to 4 isotopes combined randomly with the number of isotopes determined by the mixture frequency: zero isotopes means background only, a mixture of 2 isotopes is most common with the mixture likelihood decreasing toward 0 or 4 isotope mixtures. The spectra contain 500-2500 counts and 400 background counts.

Table 2. Parameters of the radioisotope identification study

Nuclides	^{241}Am, ^{133}Ba, ^{207}Bi, ^{57}Co, ^{60}Co, ^{137}Cs, ^{152}Eu, ^{67}Ga, ^{166}Ho, ^{131}I, ^{111}In, ^{192}Ir, ^{138}La, ^{176}Lu, ^{52}Mn, ^{99}Mo, ^{22}Na, ^{99m}Tc, ^{201}Tl, ^{88}Y, ^{65}Zr
Mixture Frequency	0 = 5%, 1 = 25%, 2 = 40%, 3 = 25%, 4 = 5%
Spectrum counts	500-2500
Background counts	400

Thousands of spectra were analyzed with the Radionuclide Analysis Kit software. The plots in Figure 8 show the radioisotope identification performance for the modeled NaI(Tl) and $SrI_2(Eu)$. The abscissa shows the number of radioisotopes in a sample and the ordinate shows the identification error rate. The plot breaks down the total error rate into incorrect inclusions (false positives), incorrect exclusions (false negatives), and multiple exclusions. In both detectors, false negatives are by far the dominant error type, so the total error rate closely tracks this. False positives are much less common, and in most cases multiple false negative is the least common. With the challenging 4 isotope mixture problem, the improved resolution of $SrI_2(Eu)$, a factor of 2 better than NaI(Tl), provides better than a factor of 2 improvement in total error rate, with multiple exclusions reduced by a greater factor. With the more common 2 isotope mixture problem, $SrI_2(Eu)$ decreases the error rate of NaI(Tl) by a factor of 5.

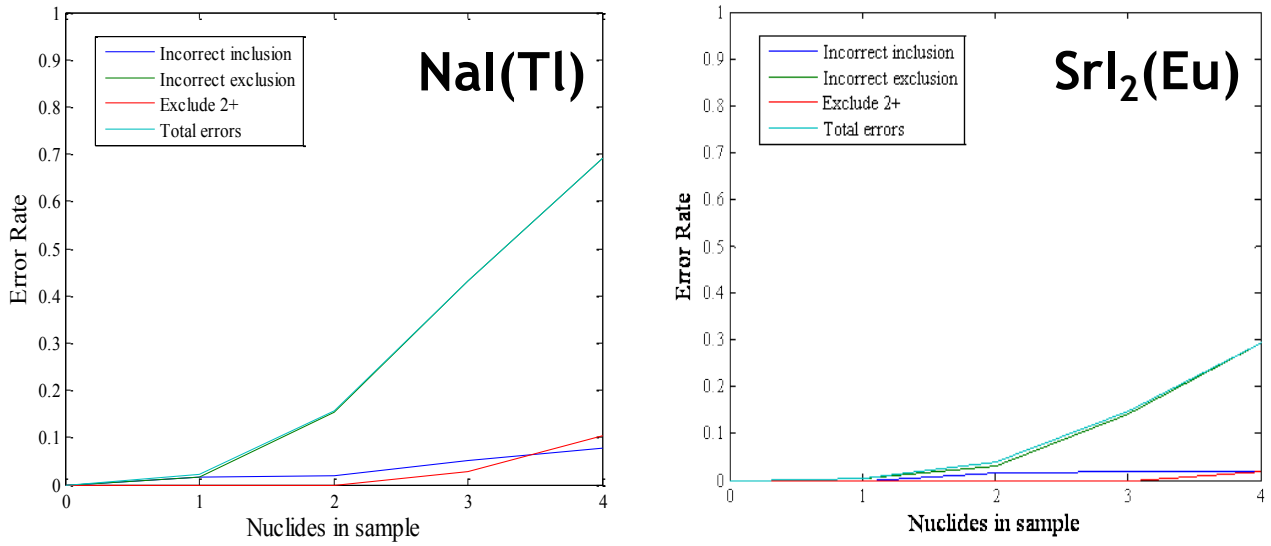

Figure 8. Radioisotope identification performance for (Left) NaI(Tl) and (Right) $SrI_2(Eu)$. The plots show the identification error rate as a function of the number of isotopes in a mixture. Incorrect exclusions (false negatives) are by far the most common, incorrect inclusions (false positives) are much less common, and in most cases multiple exclusions are the least common.

4. SUMMARY

Europium-doped strontium iodide offers excellent resolution with no intrinsic background, and it has the potential to be produced for a target price of $100 per cm^3 through the increased yield now being obtained by crystal growers [14]. Further crystal scale-up is in progress and there are several commercial vendors for SrI_2(Eu). Future work will include radioisotope identification performance comparisons between the now available 1.5" diameter SrI_2(Eu) crystals with $LaBr_3$(Ce), implementation of RIID software on the SrI_2(Eu) detector prototype, and further analysis and optimization of the SrI_2(Eu) detector prototype.

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