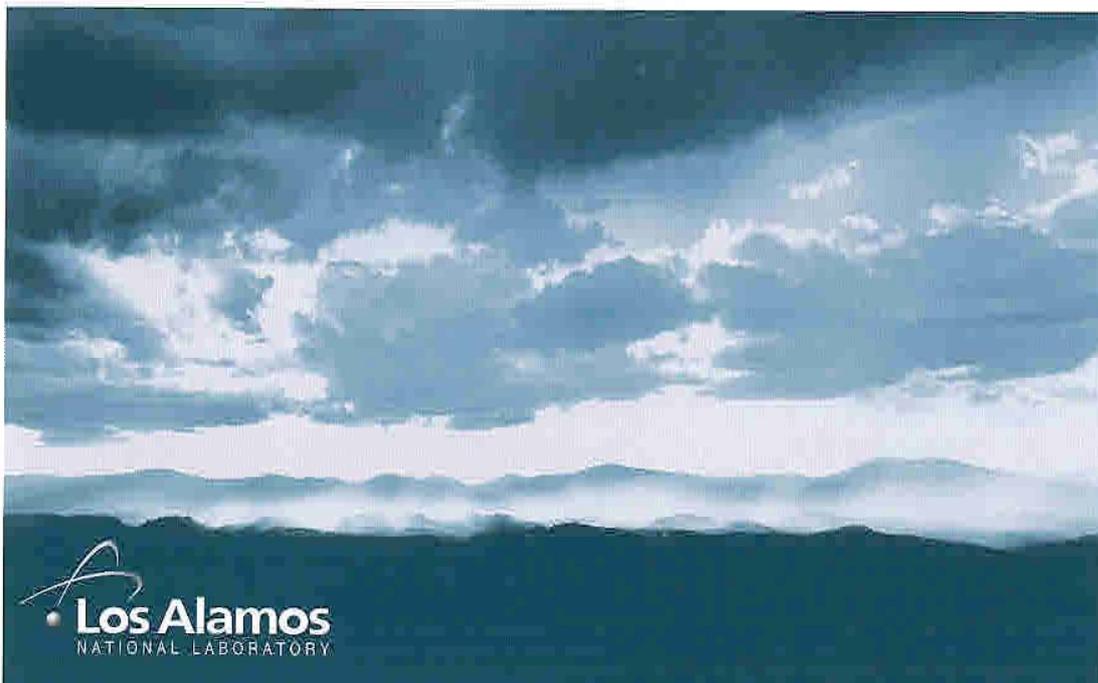


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**Application of Lanthanum Halide Scintillators and Low-Resolution Dense Plastics for  
Modern MC&A Needs.**

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## **Application of Lanthanum Halide Scintillators and Low-Resolution Dense Plastics for Modern MC&A Needs.**

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### **Abstract**

Recent developments in lanthanum halide scintillators and low-resolution dense plastics give breadth to gamma-ray methods of nuclear material detection suitable for modern MC&A needs. Demanding goals for modernization of MC&A cover both portable and continuous on-line measurement applications that are quantitative for inventory/verification, and that serve those quantitative measurement needs plant-wide. 1) Improved performance (sensitivity and resolution) is important for portable applications in which a single detector must measure many types of materials. 2) Budget is a major issue for continuous inventory measurements with hundreds or even thousands of detectors placed throughout a facility. Experimentally proven resolution of under 4% for 662 keV  $^{137}\text{Cs}$  gamma rays measured with large cerium-doped  $\text{LaCl}_3$  (lanthanum chloride) crystals sets a new performance standard for versatile, efficient portable applications comparable in price to  $\text{NaI}(\text{Tl})$ , which has been dominant for decades. While the relatively high cost of crystals remains an obstacle for the application of very large numbers of lanthanum halide scintillators as distributed networked detectors, scintillators made from high-density plastic offer a different type of solution for these gamma-ray measurements. Compared to lanthanum halide crystals they are inexpensive and can be larger in size. Despite lower resolution than  $\text{NaI}(\text{Tl})$ , a quantitative interpretation of the photopeak response of the low-cost dense plastic detectors can be tailored to the unique mechanical and spectral properties of different materials at each of hundreds of fixed on-line locations in a plant. This paper describes the properties and presents experimental results for the two new spectrometer types that, together, bracket  $\text{NaI}(\text{Tl})$  detectors in both performance and cost, fulfilling modern demands for portable and continuous on-line accountability of uranium and plutonium.

### **Introduction**

Decades of experience in the manufacturing of materials and the implementation of crystals in gamma spectroscopy gives low-resolution thallium-activated sodium iodide ( $\text{NaI}$ ) scintillators advantages of ready availability, moderate cost, and well-understood and -supported technology over most spectrometer detectors that operate at room temperature. Future safeguards needs include performance better than that of  $\text{NaI}$  in portable applications and costs lower than those of inorganic scintillators in applications to wide-area quantitative monitoring of nuclear materials using networks of distributed sensors. Visions for modernized MC&A prompt investigations of two alternatives to  $\text{NaI}$ .

Portable gamma spectroscopy will persist in its verification role in modern MC&A. The breadth of spectroscopic issues that arise in portable measurements, from those of variable backgrounds to a range of gamma interferences that represent increasingly complex missions of modern facilities, demands the very best performance combined with the highest degree of portability for access to almost any location. The cost of large ( $2.2\text{ cm}^2$  by 1.5 cm thick) intermediate-resolution

cadmium zinc telluride (CZT) detectors, in use as alternatives to NaI for the more spectroscopically demanding measurements of holdup at several DOE facilities,<sup>1</sup> remains very high, and the availability of CZT is still unacceptably limited after a decade of commercial development. However, a new intermediate-resolution 10%-cerium-activated lanthanum-chloride (LaCl<sub>3</sub>) scintillator has resolution approaching that of the large CZT detector at 662 keV. The larger size of the LaCl<sub>3</sub> crystals (5.1 cm<sup>2</sup> by 2.5 cm thick), and their ready availability and moderate cost (an assembled detector is less than one third the cost of the large CZT detector) of this new scintillator material drive this investigation of the performance of LaCl<sub>3</sub> as a sodium-iodide alternative, superior to that of CZT.

Models of modernized MC&A in the DOE Complex<sup>2</sup> include frequent material-balance closures approaching real-time accountability. This requires continuous quantitative measurements of inventory including both stored and in-process materials. The concept involves hundreds or thousands of distributed sensors managed by networks that implement algorithms for both quantitative analysis at each detection point and evaluation of inventory. The cost of the hardware and needs for rugged, proven technology drive the choice of sensors. We have verified the spectroscopic capability of a lead-loaded plastic scintillator with these needs in mind. Compelling results of these tests raise expectations for this material in applications to continuous quantitative measurements of SNM using hundreds or thousands of networked sensors. Benefits of high-Z-loaded plastics include those of plastic scintillators in general – large size, good gamma sensitivity, low cost, and ruggedness – as well as spectroscopy that enables the use of plastic in quantitative NDA.

We report the results of experimental investigations of two scintillators as alternatives to NaI. The investigations are motivated as follows for near- to mid-term safeguards applications.

1. The intermediate-resolution LaCl<sub>3</sub> detector is the possible future technology for portable gamma spectroscopy with performance superior to that of NaI and comparable to CZT, but with advantages of larger size, lower cost and greater availability compared to CZT.
2. Plastic scintillators with high-Z-materials loadings for low-resolution spectroscopy may be the key to affordability of hundreds or thousands of large, low-cost distributed sensors networked to achieve timely inventory of in-process or stored nuclear materials.

## Measurements

### *1. Lanthanum Halide Scintillators*

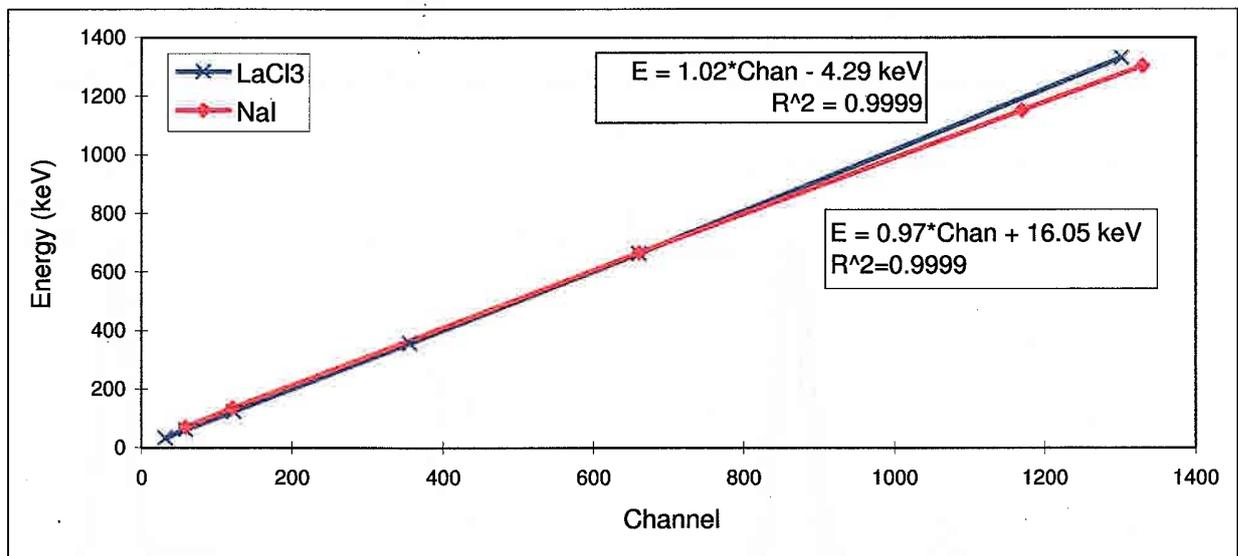
Measurements with NaI and LaCl<sub>3</sub> crystals identical in size with comparable packaging are conducted inside a dark box using the same 7.5-cm diameter Burle S83049F PMT. The PMT is built with a glass window, which prevents obtaining the manufacturer's tested resolution of 3.4% at 662 keV. Each scintillator in turn was optically coupled to the face of PMT using GE Viscasil 600 M.

Table 1 illustrates the characteristics of the two scintillators, which have similar density and Z, but differ in their emitted photon spectra and light outputs. The LaCl<sub>3</sub> emits photons close to the UV region for which a PMT with a quartz window is optimal. The energy calibration was measured with multiple gamma ray sources ranging from 60 to 1330 keV. Figure 2 shows its linearity for both scintillators. Lanthanum has a naturally radioactive isotope <sup>138</sup>La. Its 30% beta-decay branch produces the 788.7 keV gamma ray. Its 70% electron-capture decay produces

the 1435.8 keV gamma as well as barium x-rays. Figure 3 shows the natural background spectra measured with LaCl<sub>3</sub> and NaI. The prominence of the 1435.8 keV gamma peak in the LaCl<sub>3</sub> spectrum is compounded by <sup>40</sup>K 1464 keV peak from room background. The considerably weaker 788.7 keV gamma peak, which sits on top of the Compton continuum, does not appear in the NaI spectrum. Other peaks appearing in both the LaCl<sub>3</sub> and NaI spectra are from room background. The inset of Figure 3 shows the prominent barium K<sub>α1</sub> x-ray at 32.2 keV from the intrinsic LaCl<sub>3</sub> background.

**Table 1. Characteristics of LaCl<sub>3</sub> and NaI Scintillators.**

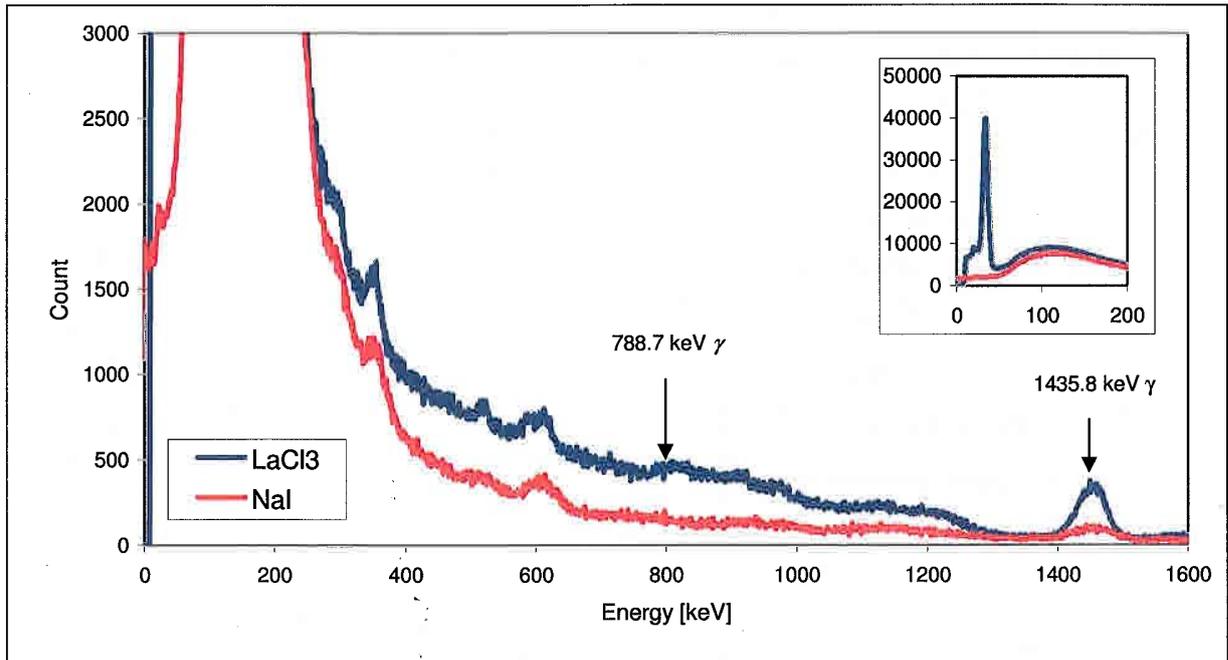
Detector	NaI	LaCl <sub>3</sub>
Dimension	1"x1"	1"x1"
Average Z	32	28
Density (g/cm <sup>3</sup> )	3.7	3.9
Resolution @ 662 keV	6.5%	3.8%
Emitted Photon Peak (nm)	415	350, 430 <sup>4</sup>
Decay time (nS)	230	20 (70%) 213 (30%) <sup>4</sup>
Light Output (photons/MeV)	38000 <sup>5</sup>	50000 <sup>4</sup>



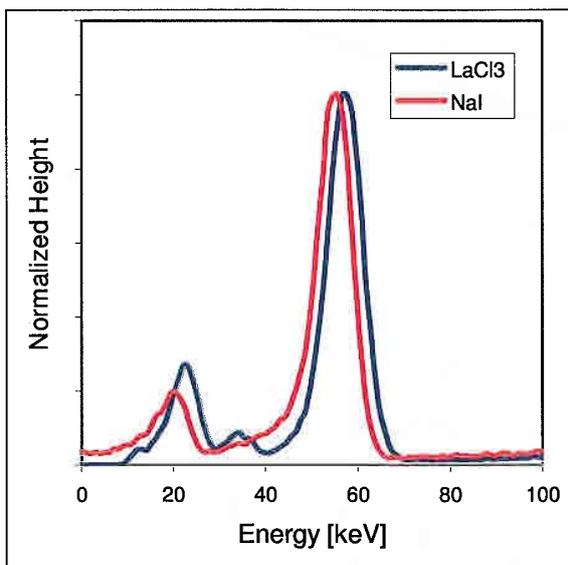
**Figure 2. Energy calibration of LaCl<sub>3</sub> and NaI scintillators.**

Sources of <sup>241</sup>Am, <sup>57</sup>Co, <sup>137</sup>Cs, and <sup>60</sup>Co were used to measure energy resolution with both scintillators. The peak regions for these spectra are illustrated in Figures 4, 5, 6, and 7, respectively. Source position 1 in Fig. 10 illustrates the approximate location of the sources with

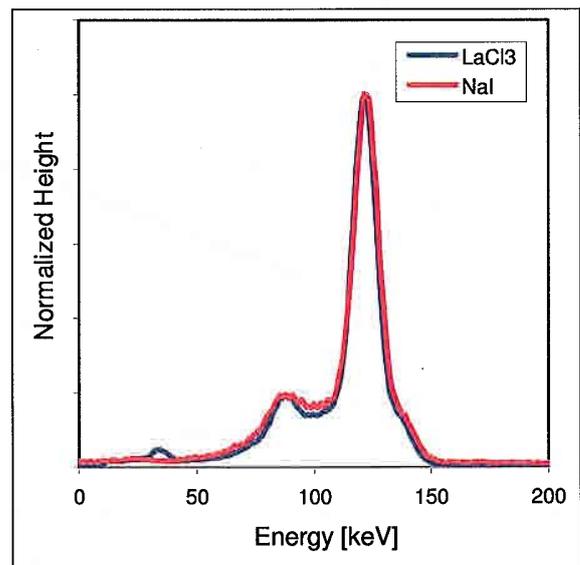
respect to the PMT in this series of measurement, and the nominal source-to detector distance is 10cm for source position 1. The significantly better performance of  $\text{LaCl}_3$  at 662 keV is diminished at lower energies. There is no significant difference in resolution between  $\text{LaCl}_3$  and  $\text{NaI}$  at 122 keV and lower energies, as shown in Figure 4 and 5. Each low-energy peak in Figures 4 and 5 shows a  $K_{\alpha 1}$  x-ray escape peak from iodine in  $\text{NaI}$  and from lanthanum in  $\text{LaCl}_3$  at 29 and 33 keV, respectively, below the main gamma peak.



**Figure 3. Background Spectra of  $\text{LaCl}_3$  and  $\text{NaI}$  measured in 10-hour counts.**

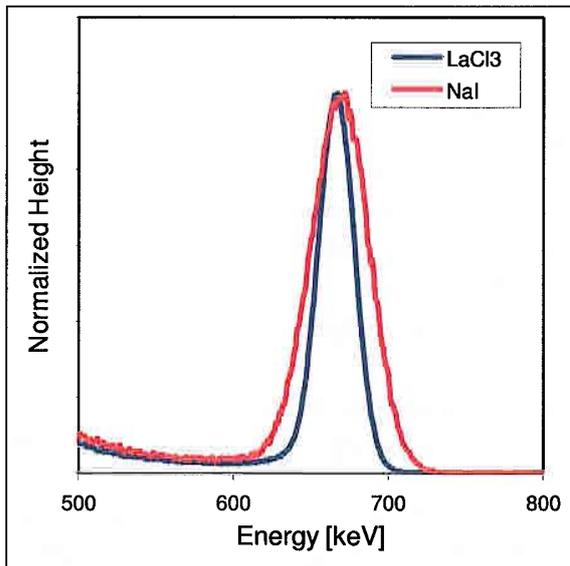


**Figure 4. The  $\text{Am-241}$  60 keV peak.**

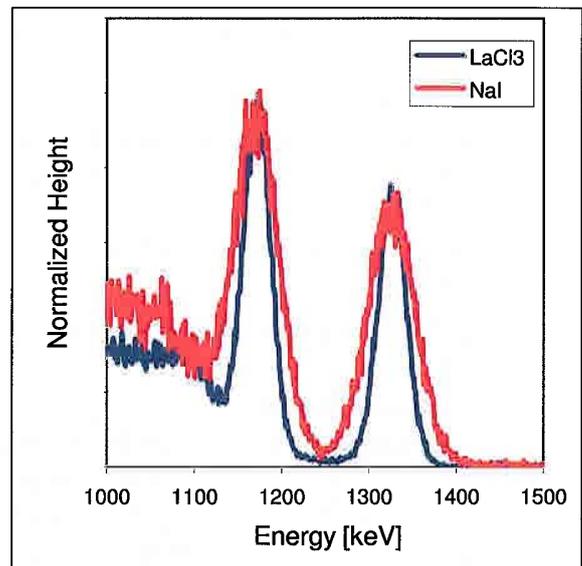


**Figure 5. The  $\text{Co-57}$  122 keV peak.**

The best resolution obtained for the 662 keV  $^{137}\text{Cs}$  gamma peak with the current experimental setup is 3.8% (FWHM) with  $\text{LaCl}_3$ . The corresponding result for the NaI scintillator is 6.5%.



*Figure 6. The Cs-137 662 keV peak.*



*Figure 7. The Co-60 1173 and 1332 keV peaks.*

## 2. Lead-Loaded Plastic Scintillators

The absence of energy resolution in plastic scintillators is the result of the very low photoelectric interaction probability of gamma rays in low-Z, low-density plastic. Most gamma rays interact in plastic by Compton scattering in which a varying fraction of the full gamma-ray energy is deposited. The resulting spectrum from a plastic scintillator is an energy continuum with no peak. The loading of plastic scintillators with higher-Z materials achieves a dramatic increase in the theoretical photoelectric interaction probability.

Table 2 illustrates this with theoretical photoelectric interaction probabilities of the typical plastic scintillator compared to those with 5% and 10% lead loadings by weight. The results are calculated for 5-cm-thick plastic scintillator used commonly in portal monitors.

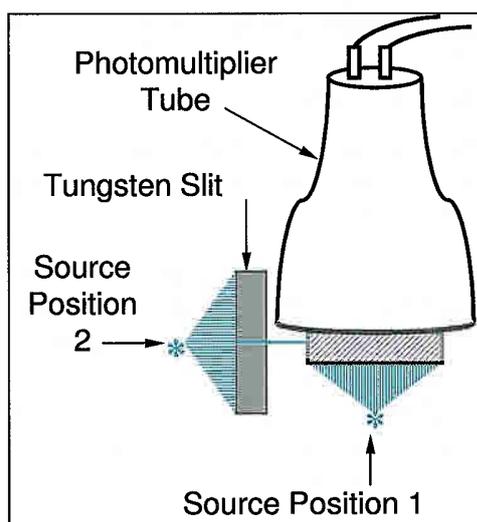
Recent experimental data provide compelling evidence of the spectroscopic capability of lead-loaded plastic scintillators, corroborating the theoretical results in Table 2. The gamma-ray spectra of  $^{109}\text{Cd}$ ,  $^{57}\text{Co}$  and  $^{235}\text{U}$  were measured using a small disk (5-cm-diameter by 1.27-cm-thick) of 5%-lead-loaded commercial plastic scintillator coupled to a three-inch diameter photomultiplier tube. Figure 8 illustrates the experimental setup with two source positions. Measurements were performed with the same Burle PMT within the same dark box utilized for the  $\text{LaCl}_3$  and NaI measurements.

Evidence of spectroscopic capability is most apparent with the sources in Position 2, in which some gamma-ray path lengths in the crystal were as long as 5 cm. Figure 9 shows the three gamma-ray spectra measured with the lead-loaded plastic. The energy calibration, gamma-ray-peak energy vs. channel number, plotted in the inset is linear. The energy resolution at 122 keV (the  $^{57}\text{Co}$  gamma ray in channel 270 of Fig. 9) is 23%, about twice that of NaI at this energy.

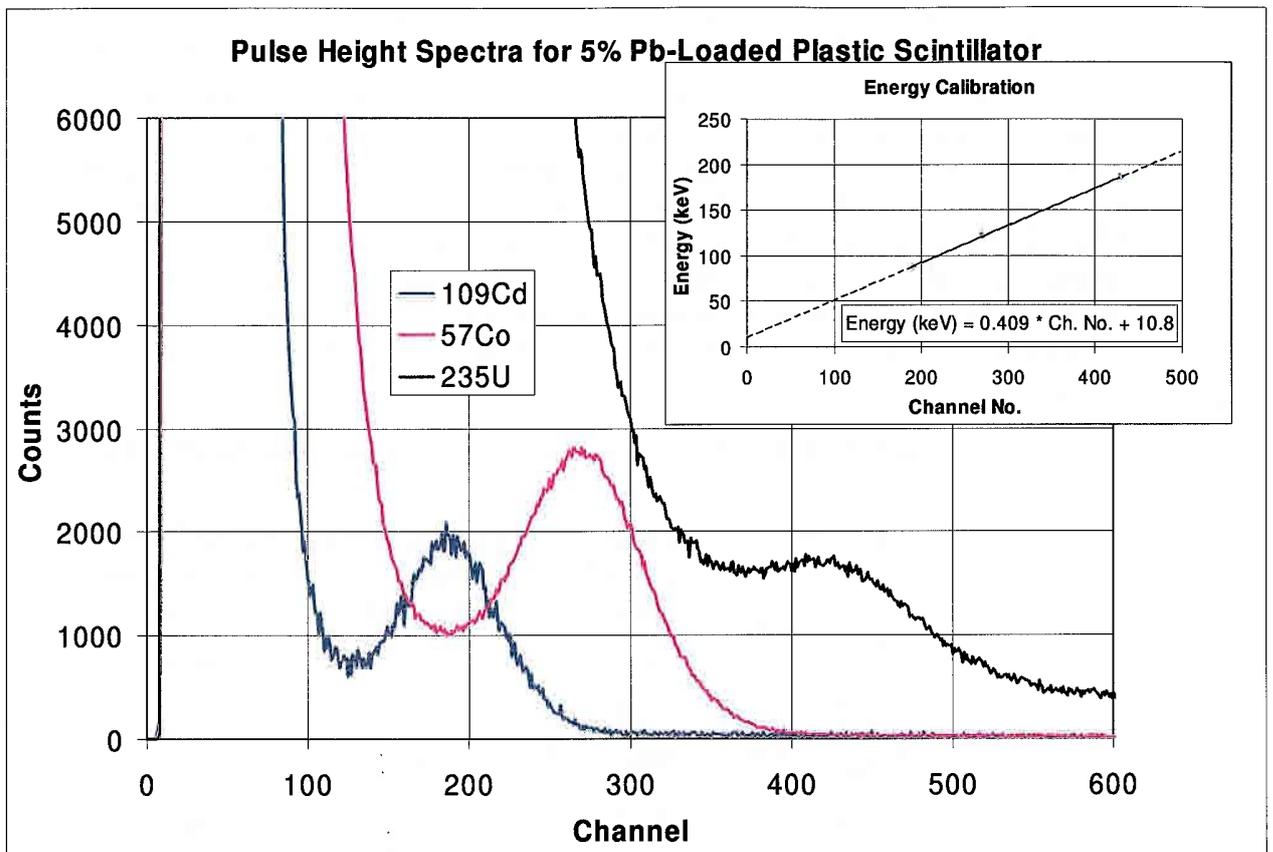
The 186-keV gamma-ray peak is only partly resolved from its Compton continuum in the  $^{235}\text{U}$  spectrum shown in Figure 9. A higher lead loading should significantly enhance the definition of this peak relative to the continuum, as indicated by the data in Table 2. The data in this table also suggest that spectroscopy at higher gamma-ray energies such as 414-keV ( $^{239}\text{Pu}$ ) may not be practical for any of the high-Z-loaded plastics. However, applications to  $^{235}\text{U}$  in continuous measurements using distributed networked scintillators should be practical because the limitations of low energy resolution in such applications are offset by the fixed location of each sensor. Unlike portable measurements in which the best possible resolution must combine with a robust analysis algorithm in order to be valid in a wide array of signal intensities, discrete spectral interferences and background contributions, the algorithms used for each fixed sensor can be tailored to the much smaller range of variations at each fixed location.

**Table 2. Photoelectric Interaction Probability (%) / Photoelectric-to-total Probability Ratio (%)**

Gamma-Ray Energy (keV)	Plastic Scintillator Type		
	No Pb loading	5% Pb loading	10% Pb loading
100	0.44% / 0.76%	73.02% / 82.15%	92.69% / 95.49%
150	0.11% / 0.21%	36.61% / 51.07%	59.78% / 72.48%
200	0.05% / 0.09%	19.12% / 31.15%	34.56% / 49.75%
250	0.03% / 0.07%	11.17% / 20.17%	21.06% / 34.39%
300	0.00% / 0.00%	7.06% / 13.78%	13.63% / 24.51%
400	0.00% / 0.00%	3.47% / 7.57%	6.82% / 14.02%
500	0.00% / 0.00%	2.04% / 4.84%	4.04% / 9.12%



**Figure 8. Hardware setup for measurements of gamma spectra with lead-loaded plastic scintillator (diagonally shaded) coupled to a photomultiplier tube. The 1.3-cm-thick tungsten collimator is a 5-cm by 2-mm horizontal slit.**



**Figure 9.** Pulse-height spectra are plotted for three gamma-ray sources in Source Position 2 (see Fig. 8) measured with the lead-loaded plastic scintillator. Energies of the three major photopeaks (88, 122 and 186 keV) of  $^{109}\text{Cd}$ ,  $^{57}\text{Co}$  and  $^{235}\text{U}$  are plotted vs. peak channel number in the inset illustrating the linearity of the pulse amplitude with energy. The spectra, acquired for fixed integral counts, were normalized (by factors of 4 and 0.4 for  $^{109}\text{Cd}$  and  $^{57}\text{Co}$ , respectively) to enable comparison of the three photopeaks on a common scale.

## Discussion

### 1. Lanthanum Halide Scintillators

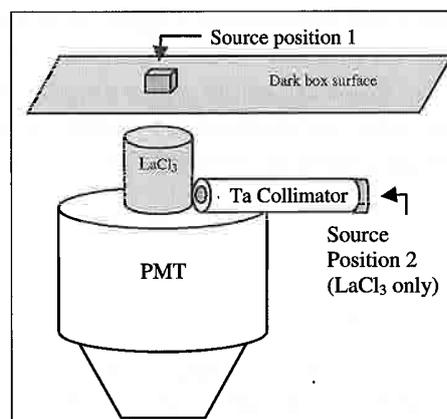
The resolution of  $\text{LaCl}_3$  at high gamma energy is among the best obtained with room-temperature gamma-ray detectors; however, in the lower energy region the measured resolution is diminished relative to that of NaI scintillators. Given that scintillation light from the lowest energy gammas must transmit through the 2.5-cm length of the crystal when the source is in position 1 (Fig. 10), a series of measurements was performed to test the possible effects on measured resolution of scintillation light attenuation by the  $\text{LaCl}_3$ . A 7.5-cm long cylindrical tantalum collimator with a 0.5-cm diameter hole is placed alongside the PMT such that the crystal window and the collimator surface rest on the same plane as shown in Fig. 10. The collimated gamma rays of all energies interact 0.9 cm from the PMT window and photocathode. Using the same gamma sources described above, spectra are acquired with each source at

position 2 in Fig. 10. The resolution measured in this setup was nearly unchanged as shown in Table 3, indicating that photon attenuation is not a problem for the 2.5-cm-thick  $\text{LaCl}_3$  crystal.

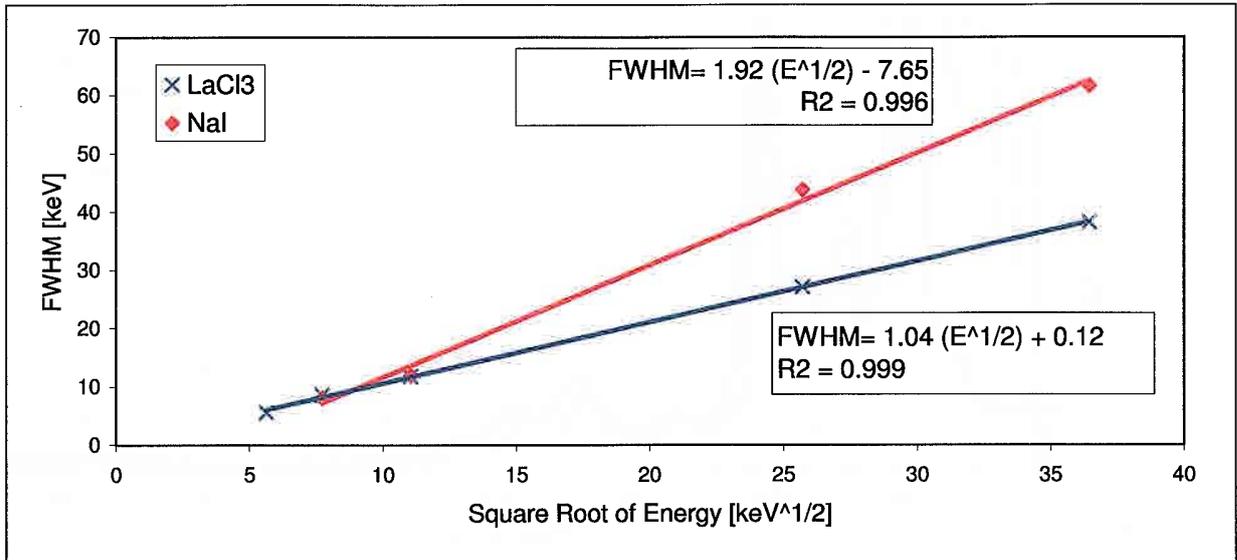
While the 10-hour long background measurements were performed to ascertain a spectrum of room background plus the intrinsic the intrinsic radiation of  $\text{LaCl}_3$ , the resolution of 32.2 keV Ba x-ray is also obtained from this spectrum. Table 3 shows that the intrinsic x-ray, whose mean free path in the scintillator is 0.5 mm, has much better resolution than the same x-ray peak in the  $^{137}\text{Cs}$  spectrum measured with the external source. This information indicates that photon transport in  $\text{LaCl}_3$  is more effective when the photons interact throughout the bulk volume of the crystal, as in intrinsic radiation, compared to interactions concentrated at the surface. The corresponding measurement with  $\text{NaI}$ , which has no intrinsic radiation source, is not possible.

**Table 3.  $\text{LaCl}_3$  Resolution (FWHM) at Various Gamma Energies.**

Energy [keV]	FWHM [keV] Position 1	FWHM [keV] Position 2	FWHM [keV] Background
32.2	N/A	8.3+-0.2	6.4+-0.1
60	8.6	8.1	N/A
122	11.8	11.2	N/A
662	27.0	25.1	N/A
1330	38.5	N/A	N/A

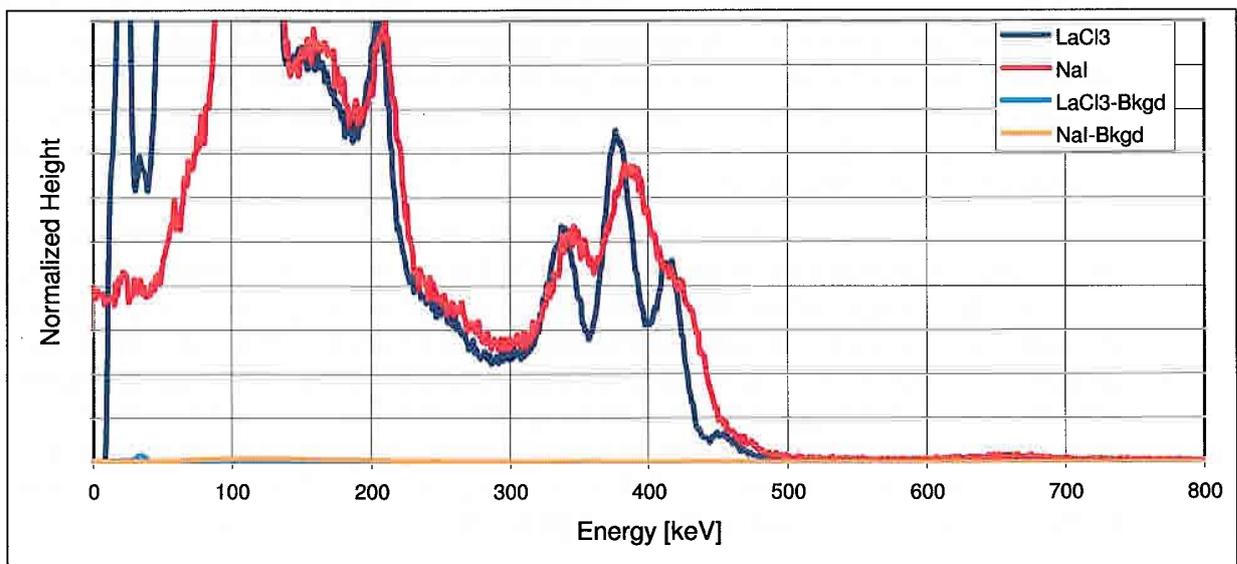


**Figure 10. Source positions for measurements with  $\text{LaCl}_3$  and  $\text{NaI}$ .**

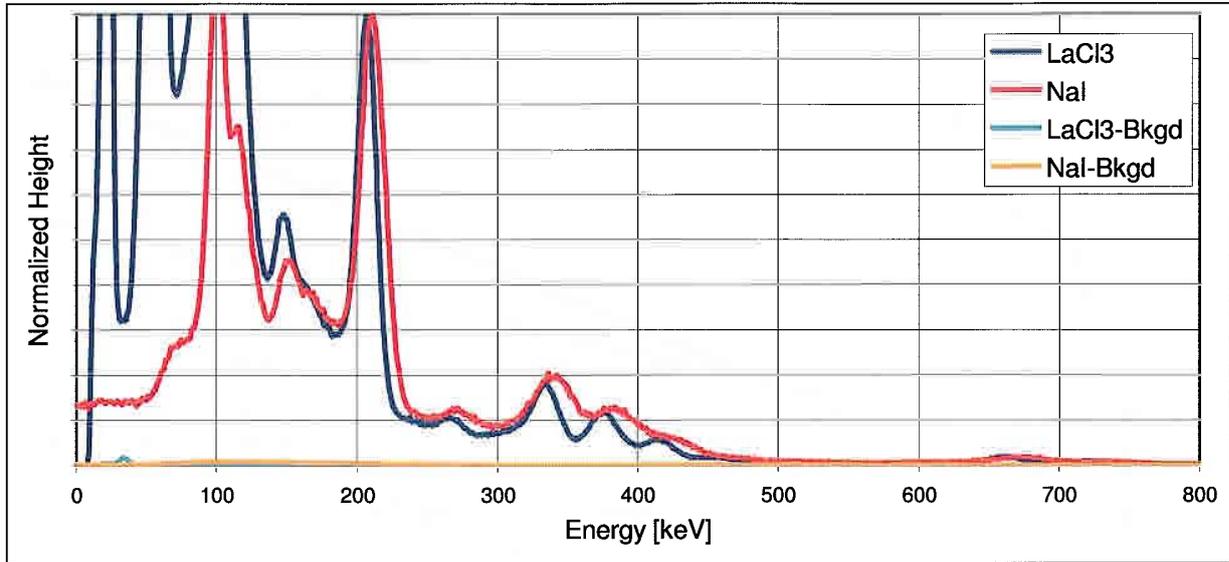


**Figure 11.** A linear relation between *FWHM* and the square root of gamma energy is shown in the straight-line fits to the data for *LaCl<sub>3</sub>* and *NaI*.

Figures 12 and 13 are plutonium spectra measured with 5-g samples of plutonium as oxide. Table 5 lists the isotopics of the two samples. *LaCl<sub>3</sub>* spectra of both the low and high burn-up plutonium in Figure 12 show peaks in the energy region from 300 and 450 keV,<sup>7</sup> while the *NaI* spectra have relatively little structure in the same energy region. The peak at 413.7 keV from <sup>239</sup>Pu is of particular interest in portable measurements of plutonium. Resolving it from the <sup>241</sup>Am peak at 376.6 keV is straightforward for *LaCl<sub>3</sub>*, but very difficult with *NaI*, particularly with aged or higher-burnup material. The background spectra for *LaCl<sub>3</sub>* and *NaI*, appropriately, normalized in time and amplitude, are also plotted in Figures 12 and 13 to illustrate their negligible contributions to the plutonium spectra.



**Figure 12.** Spectra of low-burnup plutonium and background radiation. The plutonium spectrum measured with the *NaI* scintillator is filtered with 1mm of tin.



**Figure 13. Spectra of high-burnup plutonium and background radiation. The plutonium spectrum measured with the NaI scintillator is filtered with 1mm of tin.**

**Table 5. Mass per cent isotopics of plutonium samples**

	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu	<sup>241</sup> Am
Low Burnup	0.0117	93.4123	6.3131	0.2235	0.0395	0.1047
High Burnup	0.8458	73.3191	18.2945	5.4634	2.0772	1.1705

## 2. Lead-Loaded Plastic Scintillators

There is no previous work published on the implementation or optimization of high-Z-loaded plastic scintillators for low-resolution gamma spectroscopy. The likely applications of these scintillators described above in networks of distributed sensors for continuous quantitative measurements toward timely inventory of special nuclear material warrants additional work to understand the limitations and optimize the performance of the scintillators in this and other possible safeguards applications.

Reduced light output and increased opacity to scintillation light are two possible challenges to the theoretical capability illustrated by the data in Table 2 for gamma spectroscopy with higher lead loadings or larger sensor elements composed of lead-loaded plastic. Coarse segmentation of the scintillator elements, alternative photodetectors with higher quantum efficiency, and addition of wavelength shifters toward an improved combination of attenuation length and quantum efficiency are planned approaches toward an optimized design of prototype detector. Some photodetector options may contribute to more compact, robust and lower cost mechanical designs. Investigations on loadings of alternative high-Z materials are also planned, and applications to portal monitoring have been proposed.

## Conclusion

Two scintillator alternatives to NaI show promise in applications to nuclear safeguards. This preliminary work suggests that the proposed applications will contribute to concepts of modernized MC&A for the DOE facilities.

Measurements of gamma-ray spectra with LaCl<sub>3</sub> indicate that these intermediate-resolution scintillators are very good candidates for portable quantitative measurements of <sup>239</sup>Pu with

- performance significantly improved over that of NaI and likely equivalent to that of CZT.
- substantial advantages of size, availability and cost compared to CZT.

Additional work is needed to determine whether the benefits of LaCl<sub>3</sub> performance will extend to portable measurements of <sup>235</sup>U as well when optimized photodetectors and analog electronics are implemented.

Measurements of gamma-ray spectra with lead-loaded plastics indicate that these low-resolution scintillators are very good candidates for timely measurements of <sup>235</sup>U inventory in large arrays of networked sensors. Two characteristics of these scintillators are strongly linked to the candidate application:

- Sensors can be large for high detection sensitivity.
- Detectors can be very low in cost so that very large arrays are affordable.

Continuing work will identify scintillator/photodetector combinations in which photoelectric response and optical properties are optimized.

## References

1. Smith, S.E., K.A. Thompson, J. Malcom, and P.A. Russo. 2004. "Holdup Measurement System 4 (HMS4) - Automation & Improved Accuracy," BWXT Y-12 report Y/DK-2190, to be published in *Proc.45th Annual Meeting of the INMM*, CD ROM, Northbrook IL: INMM.
2. Ensslin, N., V. Longmire, R. Stevens, M. Smith, C. Rudy, and P. Russo. 2004. "The SO-20.3 MC&A Modernization Plan Measurement Needs Summary", Los Alamos National Laboratory report LA-UR-04-4189, to be published in *Proc.45th Annual Meeting of the INMM*, Northbrook IL: INMM.
3. Burle, "Specification Sheet for S83049F Photomultipliers"
4. Shah, K.S., J. Glodo, J.M. Klugerman, L. Cirignano, W.W. Moses, S.E. Derenzo, and M.J. Weber. 2003. "LaCl<sub>3</sub>:Ce scintillator for  $\gamma$ -ray detection", *Nuclear Instruments and Methods in Physics Research A*505, 76-81.
5. Knoll, G. 2000. *Radiation Detection and Measurement*. New York: John Wiley & Sons, Inc., pp. 243.
6. van Loef, E.V.D., P. Dorenbos, C.W.E. van Eijk, K. Kramer, and H.U. Gudel. 2000. "High-energy-resolution scintillator: Ce<sup>3+</sup> activated LaCl<sub>3</sub>" *Applied Physics Letters* 77, 1467-1468.
7. Reilly, D., N. Ensslin, H. Smith, Jr., and S. Kreiner. 1991. *Passive Nondestructive Assay of Nuclear Materials*. Washington D.C.: USNRC, pp. 221-243.