Non-Destructive Assay Applications Using Temperature-Stabilized Large Volume CeBr Detectors – 14277

D. Nakazawa and F. Bronson
Canberra Industries Inc., 800 Research Parkway, Meriden, CT, 06450, USA.

ABSTRACT

Medium resolution gamma-ray detectors are commonly used in non-destructive assay (NDA) systems and in hand-held probes over a wide range of measurement scenarios and environmental conditions. Cerium Bromide scintillators (CeBr3) have received recent attention as a new detector material due to its absence of internal background, promising resolution, short scintillation decay time, and good stopping power. Large volume crystals (up to 5.1 cm diameter and 5.1 cm length) with temperature stabilization are commercially available and have undergone evaluation and characterization in recent work of the authors. This paper will focus on waste characterization and decommissioning applications using these detectors in comparison with off-the-shelf NaI:Tl and LaBr3:Ce detectors of similar size. Various collimated, shielded, and unshielded detector geometries have been evaluated for soil surveys, hold up measurements, and the non-destructive assay (NDA) containerized material, such as in far-field surveys and high dose rate situations.

INTRODUCTION

Many applications in non-destructive gamma waste assay, decontamination and decommissioning (D&D) of radiological sites, and measuring hold-up within nuclear facilities require spectroscopic detectors for accurate nuclide identification and quantification of material. Hand-held probes or stationary, shielded detectors allow for sensitive and selective assessments of special nuclear material and waste material for disposal. Large volume, medium resolution scintillation crystals, the most common being NaI:Tl, are often employed as cost effective detectors compared to high purity germanium (HPGe) or gamma imaging systems in many of these scenarios. Examples of NDA applications using medium resolution scintillators include uranium enrichment measurements [1-2], remote-handled waste [3], hold-up measurements [4], process monitoring and safeguards [5], and decontamination surveys [6-7]. In some of these references, LaBr3:Ce and LaCl3:Ce have been evaluated and investigated due to its superior resolution (3-3.5% at 662 keV) over NaI:Tl detectors (7-8.5%). However, for low background and shielded geometries, the intrinsic background from La-138 and Ac-227 isotopes within the detector material results in reduced sensitivity [8].

Cerium Bromide (CeBr3) has received recent attention as an improvement upon NaI:Tl scintillators and as a potential replacement for LaBr3:Ce scintillators [9-10]. The energy resolution of temperature-stabilized probes up to 5.1 cm in length and diameter has been measured to be 4.2% at 662 keV. Similar to LaBr3:Ce, CeBr3 has higher density and effective atomic number compared to NaI:Tl which increases CeBr3's overall efficiency. Recent evaluations of the intrinsic background [9-10] have shown varying degrees of internal Ac-227 contamination in CeBr3, which mainly affect gamma-ray energies above 1.4 MeV. Table I below summarizes the main properties of the medium resolution scintillators [11] described in this study, as well as information on Cadmium Zinc Telluride (CZT) and high purity Ge (HPGe) solid state detectors.

Table I. Summary of properties of scintillation crystals within this study. Data is taken from [11]. CZT and HPGe density and resolutions are shown for comparison.

Property	NaI:Tl	LaBr3:Ce	CeBr3	CZT	HPGe
Emission Peak (nm)	415	375	371	N/A	N/A
Density (g/cc)	3.67	5.1	5.2	5.8	5.3
Decay time (ns)	250	30	17	N/A	N/A
Light Yield (photons / MeV)	44,250	70,320	68,000	N/A	N/A
Hygroscopic	Yes	Yes	Yes	N/A	N/A
Internal Background	Low	High	Low	None	None
Non-proportionality	High	Low	Low	N/A	N/A
Resolution at 662 keV (%)	7-8.5%	3-3.5%	4%	0.5-3.0%	0.2%

In this paper, the energy resolution and temperature stability of 5.1 cm diameter CeBr3 probes will be reviewed. Next, development and validation of commercial modeling codes of the probes will be demonstrated. Unshielded and shielded backgrounds of the CeBr3 probes are compared to NaI:Tl and LaBr3:Ce probes of similar size. Finally, soil survey and hold-up geometries are modeled for evaluations of sensitivity in typical NDA applications.

DESCRIPTION

The 5.1 cm x 5.1 cm (OD x Length) CeBr3 and NaI:Tl detectors under evaluation are temperature-stabilized probes similar to that shown in Figure 1 below. The LaBr3:Ce detector is a 3.7 cm. x 3.7 cm probe without temperature stabilization. All scintillation probes were connected to Canberra Osprey Digital Tube Base electronics for supplying high voltage and signal processing. The energy resolution of the three scintillator types was measured, including a smaller 3.7 cm x 3.7 cm CeBr3 detector, with a mixed gamma ray source. Figure 2 displays the resolution as a function of energy of each probe, showing the performance of CeBr3 to be an improvement over NaI:Tl. Temperature stability tests of the CeBr3 probes were previously reported [9], and the 662 keV peak position with respect to 20 degrees C did not exceed ±0.5% when the temperature was varied from -20 to 50 degrees C.

Figure 1. Stabilized 2x2 inch CeBr3 detector. The right, red section contains the scintillator crystal, and the left end is a digital tube base and multi-channel analyzer. The total overall length is approximately 33 cm.

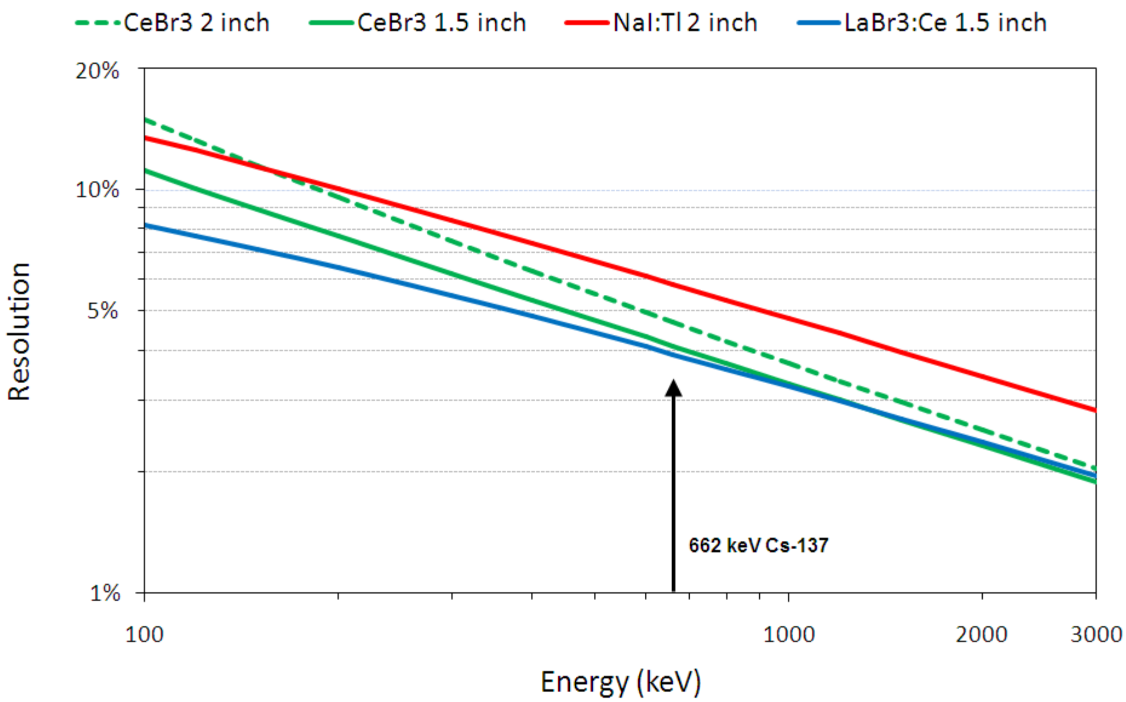


Figure 2. Resolution as a function of energy of the scintillation probes under evaluation in this study. The CeBr3 detectors are the green solid (1.5 inch) and dashed (2 inch) curves, the red curve is for NaI:Tl, and the blue curve is for LaBr3:Ce.

METHODOLOGY

In order to evaluate different geometries and shielding configurations, the efficiency of the CeBr3 probes were validated to MCNP and ISOCS modeling codes [12]. Efficiency measurements with Am-241 and Eu-152 were performed on-axis (0D) and ninety degrees off-axis (90D), both at distances of 30 cm. The methods of characterizing the detector response and verification of the modeled efficiencies using radiation transport codes are described in [12], and a picture of the 0D measurement is shown in Figure 3. ISOCS efficiencies computed for these geometries show good agreement with measured data for the different 2 inch CeBr3 probes tested (Figure 4).

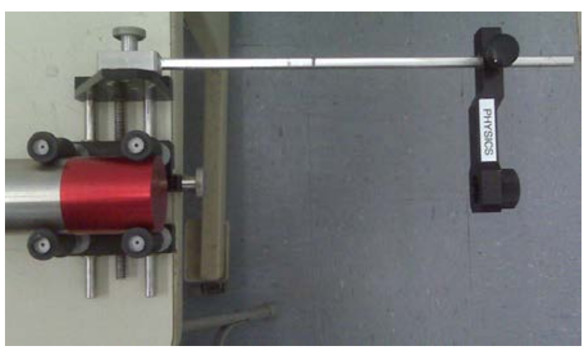


Figure 3. ISOCS characterization measurement setup with a source at 30 cm from the endcap of the CeBr3 probe. The source holder is on the right-hand side where the Am-241 / Eu-152 source is kept.

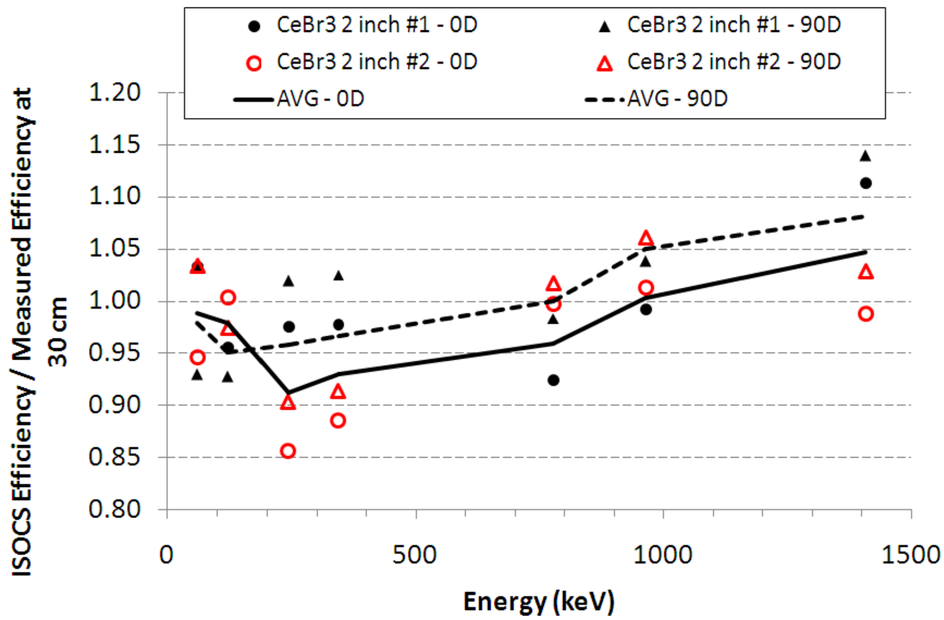


Figure 4. Ratio of ISOCS efficiencies to measured efficiencies for two CeBr3 probes at energies ranging from 60 keV to 1408 keV. The solid and dotted lines correspond to the average efficiency of the two probes. Uncertainties from counting statistics for every peak are less than 1%.

Background measurements were also performed in natural background outside, as well as shielded within 3.8 cm of Pb. Unshielded and shielded spectra are shown in Figure 5 and 6, and Tables II and III display background count rates in several energy regions-of-interest for unshielded and shielded geometries, respectively.

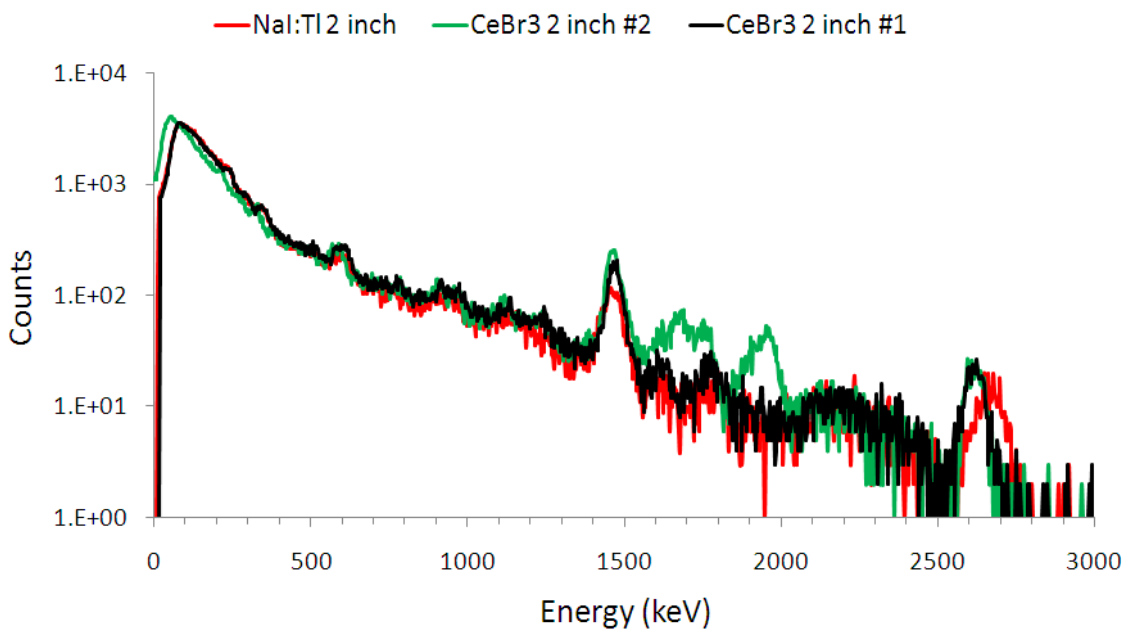


Figure 5. Unshielded background spectra of 2 inch probes for 1800 seconds.

The 1.5 inch and 2 inch CeBr3 probes labeled #2 both show some amount of Ac-227 contamination in the form of alpha counts in the region from 1400 keV to 2000 keV (green spectrum in Figure 5, black spectrum in Figure 6). There also appears to be significantly more counts in the 20-50 keV region for the CeBr3 probes with the Ac-227 contamination. This will result in reduced sensitivity, and careful selection of scintillator stock material will be needed if low background measurements are required in this region [10].

Table II. Background count rates of scintillation probes outside, unshielded.

Energy (keV)	**NaI:Tl 2 inch (cps)**	**CeBr3 2 inch #1 (cps)**	**CeBr3 2 inch #2 (cps)**	**LaBr3:Ce 1.5 inch (cps)**	**CeBr3 1.5 inch #1 (cps)**	**CeBr3 1.5 inch #2 (cps)**
20-50	7.54	7.06	18.03	25.20	3.16	6.91
50-300	98.07	95.47	81.10	79.03	58.34	55.32
300-750	22.44	24.97	20.54	23.41	15.62	11.58
750-1050	4.53	5.69	5.10	8.98	3.22	2.48
1050-1350	2.49	3.19	3.06	4.12	1.88	1.46
1350-1550	1.90	2.62	3.19	3.98	3.12	1.37
1550-2700	1.69	2.28	3.81	3.95	1.24	1.68

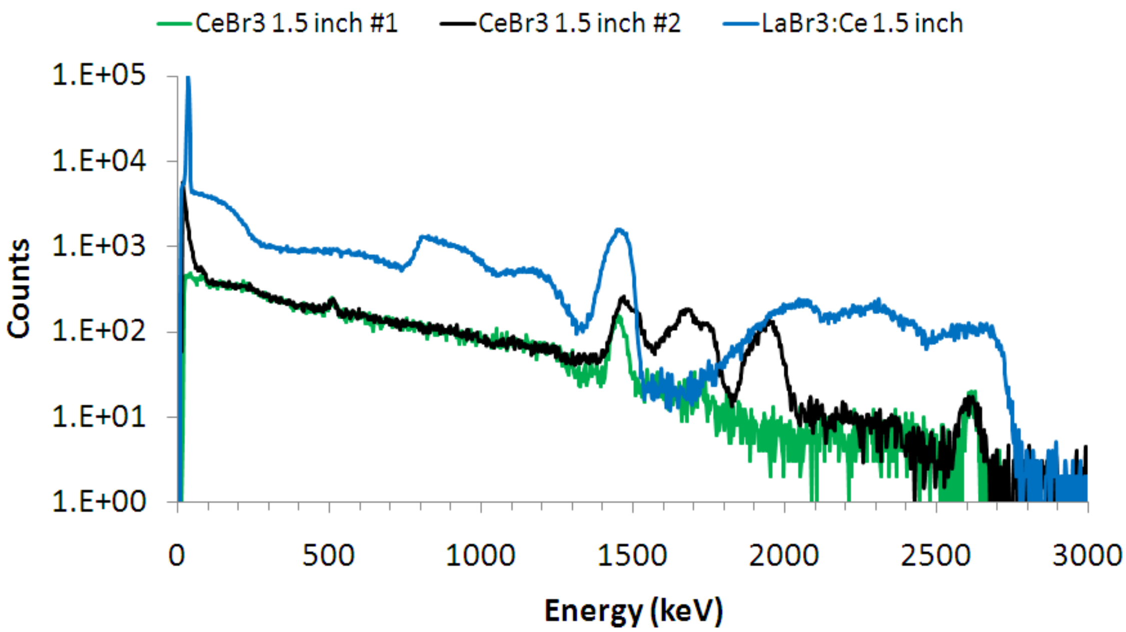


Figure 6. Shielded background spectra of 1.5 inch probes for 1800 seconds.

Table III. Background count rates of scintillation probes within a 3.8 cm Lead Portable Shield

Energy (keV)	NaI:Tl 2 inch (cps)	CeBr3 2 inch #1 (cps)	CeBr3 2 inch #2 (cps)	LaBr3:Ce 1.5 inch (cps)	CeBr3 1.5 inch #1 (cps)	CeBr3 1.5 inch #2 (cps)
20-50	0.33	0.48	0.91	21.43	0.33	1.32
50-300	5.63	2.99	3.35	14.55	1.94	2.14
300-750	1.92	3.04	2.40	8.26	1.74	1.83
750-1050	0.55	1.25	1.47	6.09	0.66	0.68
1050-1350	0.32	0.80	0.94	2.46	0.41	0.42
1350-1550	0.21	0.56	1.33	3.04	0.69	0.55
1550-2700	0.28	0.64	2.60	2.89	0.28	1.04

RESULTS AND DISCUSSION

There are many applications where detectors with improved resolution, increased efficiency, and minimal intrinsic background are desired. Figure 7 below shows an example of soil samples containing Cs-134 and Cs-137. In this case, the detectors had different sizes due to their availability at the time of measurement. However, the two spectra were acquired with the same count time, and it is clear that with the improved resolution of the CeBr3 (green), one is able to resolve the lower energy branches of the Cs-134 better than with NaI:Tl (red).

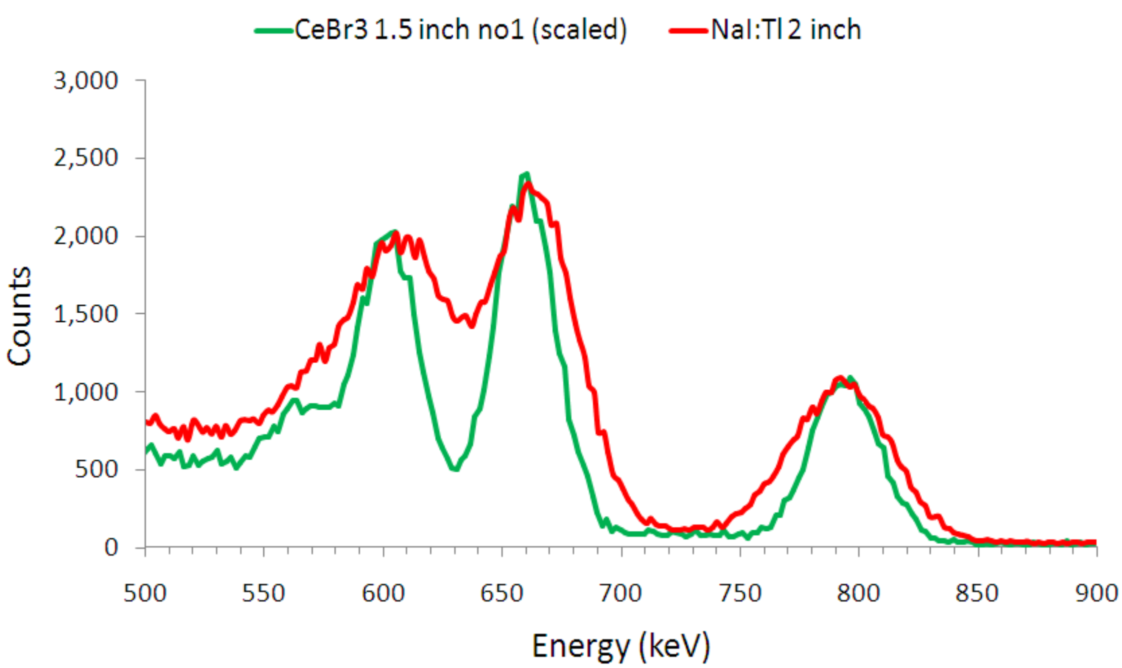


Figure 7. Measured spectra of 1.5 inch CeBr3 and a 2 inch NaI:Tl probes for 326 seconds of soil containing Cs-134 and Cs-137. The smaller CeBr3 probe is scaled to have the same peak counts in the 796 keV Cs-134 peak. The peak fitting of the Cs-134 and Cs-137 peaks is significantly easier with the CeBr3 detector.

The quality of the resolution of the two probes can be compared using a figure-of-merit (FOM) based on the peak positions of the 605 and 662 keV peaks, weighted by the fitted full-width at half maximum at the two positions, similar to the analysis of neutron/gamma discrimination in organic scintillators (Equation 1):

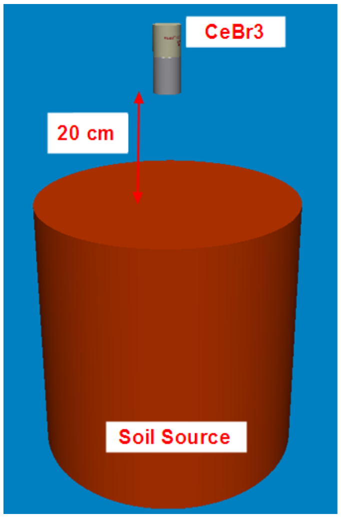
$$FOM = \frac{\Delta E}{FWHM_{E1} + FWHM_{E2}} \quad \text{(Eq. 1).}$$

As shown in Table !V, the figure of merit for the CeBr3 detector is approximately 66% better than that of NaI:Tl.

Table IV. Figure-of-Merit for the resolution of 604 and 662 keV peaks of Cs-134 and Cs-137 for CeBr3 and NaI:Tl detectors.

Detector	Full Width at Half Max. at 604 keV (keV)	Full Width at Half Max. at 662 keV (keV)	FOM
CeBr3	28.4	29.7	1.00
NaI:Tl	44.7	46.0	0.64

The efficiency of the 2 in. scintillation detectors were modeled for a soil measurement using the CeBr3 and NaI:Tl ISOCS characterizations described in the earlier section. An exponential depth profile of the activity of Cs-134 and Cs-137 was simulated, and the distance of the probe to the soil surface was set to 20 cm. The peak activity depth was 5 cm, and the relaxation length at depths greater than 5 cm was also set to 5 cm. Figure 8 displays the activity depth profile (right), and a diagram of the modeled setup (left).



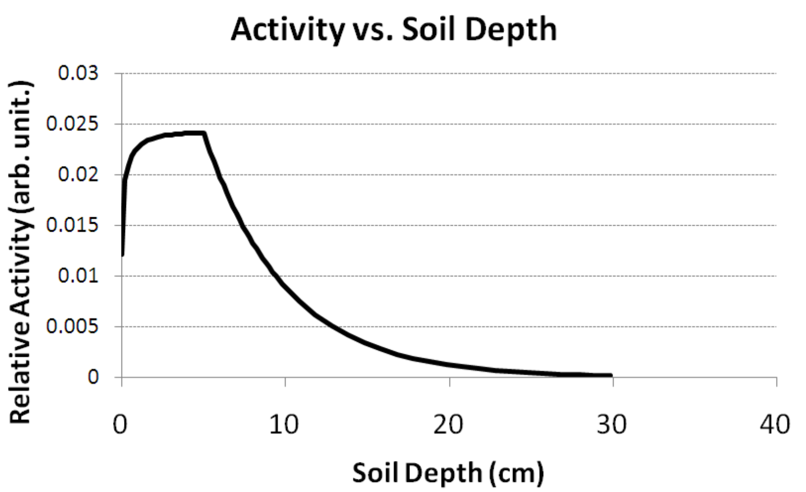


Figure 8. Diagram of the modeled efficiency for soil measurement (left) and soil activity distribution as a function of depth (right).

Table V shows the results of the ISOCS modeling of this geometry using the unshielded background measurements (Figure 5). The minimum detectable activities and counting time within the table are calculated using the Currie formalism at 95% confidence interval, and a background region of ±0.832 FWHM. The total mass of the soil in the ISOCS model is 226 kg, and the count time was determined to reach a minimum detectable activity (MDA) of 3300 Bq. From the results in Table V, the CeBr3 probes reach the minimum detectable concentration

(MDC) of 1.5 Bq per kg a factor of 1.8 times faster compared to NaI:Tl. The increased accuracy of the Cs peak fitting adds the benefits of lower total uncertainty.

Table V. Counting time to reach an MDC of 1.5 Bq / kg within the entire volume of Cs isotopes in soil with the 2 inch scintillators at a distance of 20 cm.

Nuclide	Energy	Unshielded NaI:Tl 2 inch (seconds)	Unshielded CeBr3 2 inch #1 (seconds)	Unshielded CeBr3 2 inch #2 (seconds)
Cs-134	604	99	67	60
Cs-134	796	83	48	44
Cs-137	662	98	57	53

The time-to-MDC comparison was also performed for a shielded geometry (3.8 cm Pb) for the 1.5 inch detectors. An active volume (~1 L) of water filled in a plastic cylinder (12.7 cm OD x 7.3 cm height) was placed directly on top of each detector, and the efficiencies for the two detectors were simulated. Table VI shows the sensitivity comparison of equal-sized CeBr3 and LaBr3:Ce detectors, where the intrinsic background of the LaBr3:Ce detector requires significantly longer count times for shielded geometries. The resulting improvement in counting time (Table VI) ranges from 4-8x when using CeBr3 over LaBr3:Ce.

Table VI. Time to count to reach an MDA of 22.5 Bq within the entire volume of Cs isotopes in 1.5 kg of water with the 1.5 inch scintillators in fully enclosed Pb shield.

Nuclide	Energy	Shielded LaBr3:Ce 1.5 inch (seconds)	Shielded CeBr3 1.5 inch #1 (seconds)	Shielded CeBr3 1.5 inch #2 (seconds)
Cs-134	604	2766	661	703
Cs-134	796	7283	912	939
Cs-137	662	4042	868	909

A hold-up geometry similar to that described in [5] was also modeled to compare the performance between the NaI:Tl and CeBr3 probes. Figure 9 shows the ISOCS model with a 2.54 cm cylindrical Pb collimator and shield, and a pipe containing Uranyl Nitrate solution. The material of the solution was approximated to be water, since this study is mainly for comparison purposes of the efficiency between the two detector types. The distance between the pipe and the detector was set to 10 cm. Figure 10 compares the efficiency as a function of energy, highlighting the increased stopping power and density of CeBr3 compared to NaI:Tl. The improvement in efficiency only starts to become apparent at energies above 300 keV for this geometry, reaching 1.4x higher at 1001 keV.

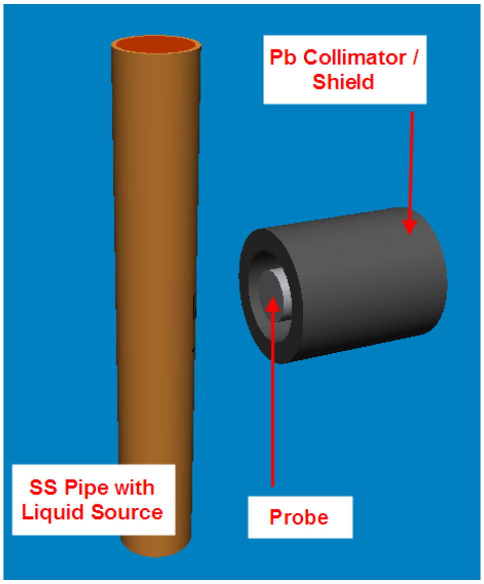


Figure 9. ISOCS model of a 2x2 inch scintillation probe in a 2.54 cm Pb shield and collimator, viewing a 304 SS pipe containing water-based radioactive source.

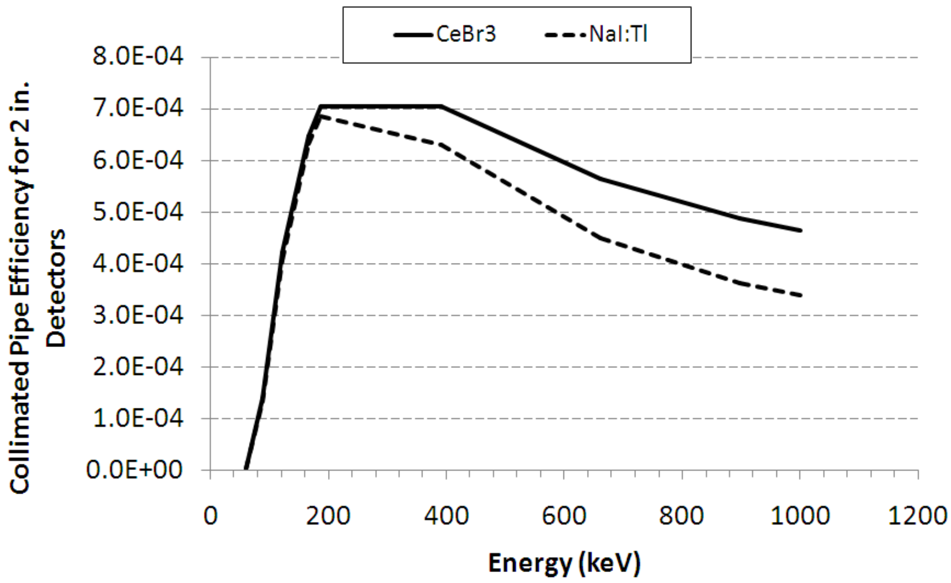


Figure 10. Comparison of pipe efficiencies for 2 inch CeBr3 and NaI:Tl detectors.

CONCLUSIONS

Large volume CeBr3 scintillation probes were characterized and evaluated for their performance in NDA applications. Measurements of the temperature stability, energy resolution, and intrinsic background continue to show that CeBr3 is a promising detector material in hold-up applications, large area surveys, and situations with complex gamma-ray spectra. The CeBr3 detectors have been successfully characterized with commercial mathematically modeled efficiency codes, and their sensitivity has been compared to NaI:Tl in a few different scenarios. The higher stopping power and better resolution of CeBr3 have been shown to both improve the quality of spectra and provide lower minimum detectable activities.

REFERENCES

1. J. K. SPRINKLE, et al. "Low-resolution Gamma-Ray Measurements of Uranium Enrichment". Presented at 3rd Topical Meeting on Industrial Radiation and Radioisotope Measurements and Applications, Raleigh, NC 1996.
2. R. BERNDT, E. FRANKE, and P. MORTREAU. "Survey of state-of-the-art NDA methods Applicable to UF6 cylinders". IAEA task n 07/TAU/04, Joint Research Center Scientific and Technical Report, 2009.
3. J. K. HARTWELL, M. M. MCILWAIN, and J. A. KULISEK. "Design and Testing of a Unique Active Compton-Suppressed LaBr3(Ce) Detector System for Improved Sensitivity Assays of TRU in Remote-Handled TRU Wastes". Presented at IEEE Nuclear Science Symposium, Honolulu, HI, 2007.
4. P. A. RUSSO. "Gamma Ray Measurements of Holdup Plant-Wide: Application Guide for Portable, General Approach", LA-14206, June 2005.
5. S. A. DEWJI, et al. "Development of Integrated Online Monitoring Systems for Detection of Diversion at Natural Uranium Conversion Facilities," Presented at Public Policy and Nuclear Threats Winter Conference, Washington D.C., 2011.
6. V. N. POTAPOV, et al. "Non-Destructive Measurements of the Characteristics of Radioactive Contamination of Near Surface Layers of Concrete and Ground with Collimated Spectrometric Detectors". *Proceedings of the Waste Management Symposia*, Tucson, AZ, 2006.
7. R. J. UNZ, D. M. Rogers, and C. A. Waggoner. "Characterizing Inorganic Scintillation Detectors for Determining Radiation Exposure". *Proceedings of the Waste Management Symposia*, Phoenix, AZ, 2011.
8. B. D. MILBRATH, J. I. MCINTYRE, R. C. RUNKLE, and L. E. Smith. "Contamination Studies of LaCl3:Ce Scintillators". *IEEE Trans. Nucl. Sci.*, **53**, 3031 (2006).
9. D. NAKAZAWA, P. SCHOTANUS, and F. BRONSON. "Characterization and Evaluation of Temperature-Stabilized Large Volume CeBr3 Detectors," Presented at IEEE Nuclear Science Symposium, Seoul, South Korea, 2013.
10. F. G. A. QUARATI, et al. "Scintillation and detection characteristics of high-sensitivity CeBr3 Gamma-Ray spectrometers," *Nucl. Inst. & Meth. In Physics Research A*, **729,** 596 (2013).
11. S. DERENZO, et al., Retrieved November 1, 2013 from http://scintillator.lbl.gov/
12. R. VENKATARAMAN, F. BRONSON, V. ATRASHKEVICH, M. FIELD, and B.M. YOUNG. "Improved detector response characterization method in ISOCS and LabSOCS". Presented at Methods and Applications of Radioanalytical Chemistry (MARC VI) conference, 2003.