

Project Number: **90109**

Project Title: **Advanced Gamma-Ray Detection Concepts Combined with Real-Time Compton Suppression for Nondestructive, Gamma-Ray Characterization of Remote Handled Waste**

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Research Objective

Nondestructive gamma ray characterization of remote-handled waste is significantly complicated by the presence of Compton scattering in the detector and waste matrix produced by intense cesium gamma rays. This research seeks to understand the photophysics of a new type of inorganic scintillation gamma ray detector, optimize the combination of this gamma ray detector with a Compton guard detector, develop new Monte Carlo solution algorithms for modeling Compton scattering in the waste, and to model the real time intensity of cesium produced Compton scattering. A successful research program will provide the fundamental information needed to design and develop advanced Compton spectrometers for assay of remote handled waste and new higher sensitivity spectrometers for environmental measurements.

This report summarizes the work after year 2 of the project.

Research Progress and Implications

Photophysics of Cerium Doped Lanthanum Halides

P. Dorenbos and G. Bizarri

Non-proportionality, as a function of photon energy, in scintillator light yield can be one of the important reasons for degradation in energy resolution of inorganic scintillators. The proportionality of a ϕ 2.5 x 2.5 cm³ LaBr₃:0.5%Ce⁺³ maintained at room temperature is evaluated. The light yield shows a very good proportionality at energies between 20 keV and 2 MeV and decreases less than 5% over this range. This behavior is about the same as was observed for smaller crystal sizes. Cerium concentration and crystal size do not seem to be relevant parameters in non-proportionality response of the LaBr₃ material.

The energy resolution for this crystal was determined from peaks in the pulse height spectra. The data, between 6 keV and 2 MeV, displayed an energy resolution that decreased with the square root of the gamma ray energy. These results were similar to the ones that were reported in the literature for low Cerium concentration in crystals.

These results confirm again that the upgrade from the small crystal size to a large one leads to much deterioration in scintillator performance.

The temperature dependence (70 K to 700 K) of fluorescence lifetime under gamma-ray excitation has been studied on unpackaged crystals. The decay curves for crystals maintained above 200 K have single exponential time dependence. Below 200 K, the time profiles are not single exponential and have a long decay component extending into the microsecond region. For the fast component, the lifetime increases from 15 ns to 24 ns from 80 K to 700 K. For the slow component, the lifetime decreases with increased temperature and disappears above 200 K. The decreasing lifetime is typical for self-

trapped excitons, and the decrease of STE luminescence with increase of temperature supports this mechanism.

Waste and Detector Model Development

University of Florida
Ali Hahgihat

The impact of different physical processes and detector parameters on the computed detector response as determined as a pulse-height spectrum were investigated. This investigation showed the inability of MCNP5 code to model the visible light photons due to limitations in its physical models. This hindered the ability to model accurately the leakage/absorption and the spectrum of the visible light emitted by the crystal. However, it was encouraging to note that the code was able to predict spectra having features similar to those determined for an actual scintillator detector.

The energy broadening of the modeled pulse height spectrum was studied in order to represent the inherent statistical fluctuations present in the experimental data. This was necessary for constructing an effective detector response library. This work demonstrated that it was necessary to analyze experimental data to obtain appropriate values for broadening parameters.

Some analyses were also performed to estimate the detector performance and sensitivity as a function of various characteristics such as crystal size, detector window thickness, and source position. Based on this analysis, we estimated the peak-to-total ratios for different detector configurations to identify the most suitable experimental setup for validating our modeling results.

Compton Detector Development

Idaho National Laboratory
M. McIlwain, J. K. Hartwell, W. Scates

We have revisited Compton Suppression Spectrometer (CSS) parameter optimization using MCNP transport calculations. What is novel about these results is that we have used an inorganic scintillator ($\text{LaCl}_3:10\% \text{ Ce}$) as the Primary Detector (PD) instead of the conventional germanium detector. Our basic premise for this substitution is that by employing a scintillator that does not require cooling will greatly reduce the amount of high Z material between the PD and Suppression Detector (SD). The resulting suppression factors will be greatly enhanced. Our optimization studies substantiate this premise.

Because there are often a great number of assumptions and approximations made to model a CSS detector, we validated the MCNP transport model by comparing the model predictions of a ^{137}Cs source measured by two different types of CSS detectors with actual measurements obtained using two laboratory constructed units. The MCNP predicted spectra compare reasonably well with the experimental spectra. Further, the resulting suppression factors prepared from the model and experimental spectra are

within in 20% of each other for our energy range of interest, of 100 to 300 keV. This agreement is sufficiently strong to suggest that we have a reasonable MCNP model for the CSS. The agreement between model and experiment results further supports the use of the model to predict optimum CSS parameters. Further, this agreement with experimental data supports using MCNP transport calculations for optimizing other detector configurations beyond those studied in this work.

The parameter optimization studies suggest that for a $\text{LaCl}_3\text{:Ce}$ scintillator PD enclosed by cylindrical NaI(Tl) SD the optimum radial thickness for the SD is approximately 100 mm. The optimum position of the PD within the NaI was centrally located along the axis of symmetry about 58 mm from the front surface of the SD and with about 50 to 60 mm of SD material behind the PD. CSS detectors based on scintillators require some path for the PD light to be detected. The orientation of this path was found not to be strongly sensitive to the angle between the axis of symmetry and light path. The conventional 90° orientation significantly reduces the resulting suppression factor. This analysis indicates that a 0° orientation relative to the axis of symmetry represents the optimum geometry.

Finally, to support our premise that reducing the amount of material between the PD and SD would have a significant impact on the Compton suppression, the thickness of the aluminum metal associated with the crystal aluminum enclosure was varied. Over the range of values examined, very little impact to the suppression factor was observed until the metal thickness decreased below 0.1 mm. Between 0 and 0.1 mm there was a dramatic improvement in the resulting suppression factor. These results indicate that we need to find some way to remove the metal can that surrounds the $\text{LaCl}_3\text{:Ce}$ crystal. We are currently attempting to validate this conclusion experimentally. Another option might be to use a semiconductor detector like CZT that requires only optical isolation from the SD and a path for a few wires to power the device and access the signal.

Relevant Publication and Presentations

1. G. Bizarri, J.T.M. de Haas, P. Dorenbos, C.W.E. van Eijk, (2005) "Scintillation properties of ϕ 1 x 1 inch³ $\text{LaBr}_3\text{:5% Ce}^{3+}$ crystals, accepted for publication in IEEE Transactions in Nuclear Sciences.
2. J. K. Hartwell, R. J. Gehrke, M. E. McIlwain, (2005) "Performance comparison of four compact room-temperature detectors – two cadmium zinc telluride (CZT) semiconductor detectors, a $\text{LaCl}_3\text{(Ce)}$ scintillator, and an NaI(Tl) scintillator", accepted for publication in IEEE Transactions in Nuclear Sciences.
3. R. Aryaeinejad, J. K Hartwell, W.W. Scates (2005) "High-Resolution Compton-Suppressed CZT and LaCl_3 Detectors for Fission Products Identifications", accepted for publication in IEEE Transactions in Nuclear Sciences.
4. W. W. Scates, J. K. Hartwell, R. Aryaeinejad, M. E. McIlwain, (2005)" Optimization studies of a Compton suppression spectrometer using experimentally validated Monte Carlo simulations", accepted for publication in Nuclear Instrumentation and Methods – Section A.