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A Compact, Portable, Reduced-Cost, Gamma Ray Spectroscopic System for Nuclear Verification Final Report CRADA No. TSB-1551-98

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A Compact, Portable, Reduced-Cost, Gamma Ray Spectroscopic System For Nuclear Verification

Project Accomplishments Summary
CRADA No. TSB-1551-98
Date Technical Work Ended: November 24, 1999

Date: July 24, 2001 Revision: 4

A. Parties

The project was a relationship between the Lawrence Livermore National Laboratory (LLNL) and Spire Corporation.

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B. Background

Uranium Enrichment Analysis

At the time of the CRADA, there were two primary methods used to determine uranium enrichment. The method selected for use was directly related to the type of radiation detector used to acquire the uranium energy spectrum.

The first method was used in applications requiring highly accurate enrichment determinations and was applied to spectra obtained from high-purity germanium-based (HPGe) detection systems. This analysis method was incorporated in industry standard software packages (including MGA, MGA++, GRPANL, U235) and concentrated the analysis in the 100 keV energy range of the gamma-ray spectrum. This was a very complex energy region with multiple U235, U238 and decay product gamma- and X-ray energies in close proximity. Analyzing energy lines in close proximity eliminated the need to consider the energy dependent efficiency of the detection system resulting in significant increased analysis accuracy. To take advantage of this energy region, the energy resolution of the system was critical. High performance high-purity germanium (HPGe) and cadmium zinc telluride (CZT) detector systems were used in applications using this method.

The second method, referred to as the Meter Method, was an analysis routine used with lower energy resolution systems and for analysis of heavily shielded sources where the 100 keV energy region was not available. This method compared a modeled spectrum with the actual data at the 185.7 keV gamma-ray energy line. A spectrum was acquired from a physical model of the anticipated source-detector geometry (including all shielding and a uranium source of known enrichment). The peak-to-background ratio was determined from this spectrum and used as a basis for assessing the actual measurement. Given similar acquisition parameters, the 185.7 keV peak-to-background ratio is directly proportional to uranium enrichment. There were many sources of uncertainty when using this method including the accuracy of the modeled spectrum with respect to the actual measurement scenario and selection of the peak and background spectral energy points selected for analysis. This method is primarily used in conjunction with sodium iodide-based (NaI) detection systems that typically exhibit characteristically poor energy resolution and spectral instability.

The analysis results of the Meter Method could be improved by using CZT detector technology that provides better energy resolution and spectral stability with less background resulting in more accurate uranium enrichment measurements.

Existing Portable Systems and Their Disadvantages

At the time of the CRADA, there were two types of portable gamma-ray spectroscopic systems that were commercially available for nuclear spectroscopy: low energy resolution systems based on NaI scintillators, and high performance systems based on HPGe detectors. NaI scintillators provide a quick determination of the presence of radioactive materials at remote locations, but poor energy resolution limits accurate identification of the contaminants. HPGe detectors exhibit the highest energy resolution obtainable, though HPGe detectors require liquid nitrogen for proper operation and are therefore not easily used in a portable environment, the focus of this application.

CZT Detector Technology

Room temperature semiconductor detectors are well-suited to complement existing gamma detectors in the above-described applications. CZT detectors have been demonstrated to provide moderate energy resolution (<5% in the 100 to 200 keV) with small detection volumes (<100 mm^3). However, for higher energies (>200 keV), large size detectors were needed to achieve the required energy efficiency. Unfortunately, spectral energy resolution was low in large size crystals due to poor charge carrier mobility. Because of this problem, smaller size (5x5x5 mm) crystals were used, but with longer collection times to compensate for the resulting lower detection efficiency. For 5 mm thick CZT, the stopping efficiency decreased significantly for energies higher than 200 keV.

In CZT detectors electron collection efficiency was quite good and only poor hole collection efficiency degraded the energy resolution. The reason for poor hole collection efficiency was a low mobility-lifetime product, which resulted in drift lengths typically less than the thickness of the detector. Methods to circumvent this limitation included new device electrode and geometry designs, thermoelectric cooling, and electronic signal processing. Device designs included coplanar grids and P-I-N structures, and electronic signal correction included pulse-shape discrimination and charge-loss compensation methods.

Of all the methods, coplanar grid and P-I-N device structures were the most promising methods to improve the energy resolution of CZT detectors.

Coplanar grid designs offer an alternative geometry, based on the Frisch grid, which allow for single-carrier charge collection. The device was operated by applying a potential across the bulk of the device from the back plane to the grid, while maintaining a separate potential between adjacent grid lines. Under such a geometry, the low-mobility carriers (holes in the case of CZT) did not contribute to the detector induced charge, and only the electrons contributed to pulse formation. Electrons excited in the device were swept to the grid region, and then collected by the grids with the highest positive potential. When operated in differential mode, the induced charge signal did not begin to form until the electrons were moving through the local vicinity of the grid; hence holes moving in the bulk did not affect the pulse formation, and the detector operated as a single-carrier device. Position-dependent charge collection effects were reduced under such conditions, thereby allowing for the realization of thicker detectors than conventional planar designs without degradation in the energy resolution.

Paul Luke from Lawrence Berkeley Laboratory (LBNL) originated this design for CZT and demonstrated improved energy resolution. Initially, energy resolution of 3.7% was obtained for the 662 keV peak of ^{137}Cs using a CZT detector with size 5 mm x 5 mm x 5 mm. Luke also reported an energy resolution of 2.5% with a 1 cm^3 CZT detector by

modifying the grid design. A group from the University of Michigan reported further improvement in energy resolution using a sophisticated pulse processing method. They showed energy resolution as high as 1.25% at 662 keV for a 1 cm^3 coplanar CZT detector. Their theoretical calculations suggested that with proper coplanar grid design, energy resolution of 0.5% could be achieved. Coplanar detectors with a detection volume of 1.5 cm x 1.5 cm x 1 cm were being fabricated. Although coplanar detectors provided high-energy resolution above 500 keV, they exhibited relatively poor energy resolution at low energies (<200 keV) due to electronic noise.

An LBNL group developed a single-electrode readout for a coplanar detector to reduce the electronic noise. All these results strongly suggested that coplanar detectors had great promise for high-energy radionuclide spectroscopy.

Though existing portable systems demonstrated the potential of CZT detectors, for many field applications large volume (>2 cm^3) detectors were necessary. Since storage containers had thick walls, low energy gamma peaks were attenuated and the intensity of high energy gamma peaks was low. Large volume detectors were necessary to increase the energy efficiency and reduce measurement times. Another limitation of existing CZT based portable systems was the software for analysis of spectra. Most of the software programs were developed for HPGe detector spectra. Since HPGe detectors had high energy resolution with Gaussian photopeaks, fitting was accurate. On the other hand, CZT detectors exhibit moderate energy resolution with peaks that were not Gaussian; therefore, the fitting programs had to be modified. Existing portable systems did not have any built-in software to analyze uranium enrichment.

Spire proposed to advance the capability of portable systems by using a large volume CZT detector and a software program dedicated to uranium enrichment analysis.

C. Description

The overall goal of this project was to demonstrate a compact gamma-ray spectroscopic system with better energy resolution and lower costs than scintillator-based detector systems for uranium enrichment analysis applications.

Spire Corporation built a compact, portable, reduced-cost gamma-ray spectroscopic system with the required energy resolution and detection efficiency for uranium enrichment analysis. The overall performance of the prototype system was reasonable considering the quality of the detectors and the type of electronic packaging utilized. Unfortunately, the performance of the prototype system did not meet the minimum specifications required to perform uranium enrichment analysis using codes developed at LLNL, although it would most likely be adequate for use in "Meter Method" based analysis.

Given the system in its current configuration, there is substantial potential for improvement in performance. It is tremendously encouraging to see a system that integrates multiple large volume Cadmium Zinc Telluride (CZT) detectors providing reasonable resolution with off-the-shelf components. Further attention to the design of the preamplifiers, circuit board layout and mechanical design of the detector assembly should realize significant and necessary improvements in energy resolution performance.

D. Expected Economic Impact

The most significant shortcoming of CZT detector systems is the relative small size (crystal volume) of high quality detector elements that directly results in lengthy spectral acquisition times. In most cases, it is desirable to obtain the energy spectrum in as short a time as possible to facilitate rapid evaluation, efficient inspection activities, and minimized radiation exposure to inspection personnel. Additionally, small volume detectors are not suitable for search activities, as these efforts require large detection efficiency for reasonable performance in this role. Provided a substantial improvement in detector area is realized, this detector system could be used for both limited search activities as well as subsequent spectral analysis reducing the required number of detection systems for a given application. Without exception, all current commercial CZT instruments utilize small volume detectors that do not lend themselves to rapid evaluation and therefore have limited utility.

Successful implementation of Spire Corporation detector fabrication techniques coupled with signal processing circuitry and spectral analysis software developed at LLNL could be rapidly configured as a commercial product. The resulting product would have high potential for use in any application requiring rapid nuclide evaluation in an environment requiring portable instrumentation such as decontamination and dismantlement (D&D) activities, container/transportainer inspection, and portal monitoring.

E. Benefits to DOE

Applications of this system for other nonproliferation and international and domestic safeguards include:

- Detection of nuclear smuggling/illicit nuclear materials
- Routine or special nuclear inspections
- Portable quantitative assay of holdup and in-process inventory
- Monitoring and verification of nuclear inventories
- Continuous, unattended gamma-ray monitoring
- Domestic safeguards accountability close-out in decontamination and decommissioning
- Surveillance

F. Industry Area

The commercial application of this instrument is in the areas of environmental remediation, radioactive contaminant characterization at waste sites, and to screen small baggage for radioactive materials.

G. Project Status

Phase I of this project is completed. Phase II work will continue under a new CRADA currently being negotiated.

H. LLNL Point of Contact for Project Information

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I. Company Size and Point(s) of Contact

Spire Corporation is a publicly held company with annual sales of $12 million. The company employs 88 people.

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J. Project Examples

There are no project examples.

K. Subject Inventions

This small value contractual mechanism did not anticipate any generation of Intellectual Property (IP) including subject inventions. The LLNL contributors and the company participants both indicate that no new intellectual property was generated.

Documents/Reference List

Reports

UCRL-ID-134587, "A Compact, Portable, Reduced-Cost, Gamma Ray Spectroscopic System for Nuclear Verification," Anthony Lavietes, 11/24/99.

Patent/Copyright Activity

No software was developed during this CRADA.

Generated Information

No intellectual property was generated or disclosed during the execution of this CRADA.

Background Intellectual Property

LLNL

None

SPIRE

U.S. Patent Application 08/949,015 *Semiconductor P-I-N Detector and Method of Manufacturing the Detector*, Sudharsanan, R.; Karam, N.

Licensing discussions have not occurred and are reserved until the completion of Phase II work.

L. Release of Information

I certify that all information contained in this report is accurate and releasable to the best of my knowledge.

Karena McKinley, Director — Date: 10/22/01
Industrial Partnerships and Commercialization

Release of Information

I have reviewed the attached Project Accomplishment Summary prepared by Lawrence Livermore National Laboratory and agree that the information about our CRADA may be released for external distribution.

Nader Kalkhoran — Date: 10-09-01
Spire Corporation

Anthony Lavietes, Principal Investigator — Date: 10/16/01
Lawrence Livermore National Laboratory