Contract No:

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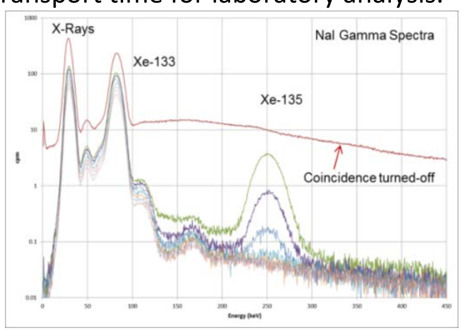
Phase II: Field Development for Undeclared/Declared Nuclear Testing for Treaty Verification

Radioactive xenon isotopes are a critical part of the Comprehensive Nuclear Test Ban Treaty (CTBT) for the detection or confirmation of nuclear weapons tests as well as on-site treaty verification monitoring. On-site monitoring is not currently conducted because there are no commercially available small/robust field detector devices to measure the radioactive xenon isotopes. Xenon is an ideal signature to detect clandestine nuclear events since they are difficult to contain and can diffuse and migrate through soils due to their inert nature. There are four key radioxenon isotopes used in monitoring: ^{135}Xe (9 hour half-life), ^{133m}Xe (2 day half-life), ^{133}Xe (5 day half-life) and ^{131m}Xe (12 day half-life) that decay through beta emission and gamma emission. Savannah River National Laboratory (SRNL) is a leader in the field of gas collections and has developed highly selective molecular sieves that allow for the collection of xenon gas directly from air. Phase I assessed the development of a small, robust beta-gamma coincidence counting system, that combines collection and *in situ* detection methodologies. Phase II of the project began development of the custom electronics enabling 2D beta-gamma coincidence analysis in a field portable system. This will be a significant advancement for field detection/quantification of short-lived xenon isotopes that would not survive transport time for laboratory analysis.



Awards and Recognition

None

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publically published in its current form.

SRNL Legal Signature

Signature **Date**

LDRD-2015-00021
LDRD Report

Phase II: Field Development for Undeclared/Declared Nuclear Testing for Treaty Verification

Project Team: M. R. Kriz (Primary), R.M. Achey, J.R. Cadieux, M.C. Duff, R.K. Huffman, D.B. Hunter, E.M. Kriikku and T. N. Riley

Subcontractor:

Thrust Area: ST3

Project Type: Standard

Project Start Date: October 1, 2014
Project End Date: September 30, 2015

Radioxenon is a critical part of the Comprehensive Nuclear Test Ban Treaty (CTBT) for the detection/confirmation of undeclared/declared nuclear weapons tests. Treaty verification monitoring for radioxenon is another application for post-event monitoring on-site; however, this is not currently conducted because there are no simple portable field devices for measurements. Phase I assessed the development of a small, robust beta-gamma coincidence counting system, that combines collection and in situ detection methodologies. Phase II of the project began development of the custom electronics enabling 2D beta-gamma coincidence analysis in a field portable system. This will be a significant advancement for field detection/quantification of short-lived xenon isotopes that would not survive transport time for laboratory analysis.

FY2015 Objectives

- Development of a small field detector system for beta-gamma coincidence counting that could combine SRNL molecular sieves with *in situ* detection of radioxenon directly from air
- Correlation of the beta signal (time stamp, energy & pulse height) with the gamma signal (time stamp & pulse height)
- Characterization of the beta-gamma detector (quantification of the beta signal, beta signal efficiency and detector linearity)
- Gas transfer mechanism from collection sieve to gas scintillation cell

Introduction

Radioxenon is a critical part of the Comprehensive Nuclear Test Ban Treaty (CTBT) for the detection/confirmation of undeclared/declared nuclear weapons tests. Noble gases are ideal signatures to detect clandestine nuclear events since they are difficult to contain and can diffuse and migrate through soils due to their inert nature. Radioxenon is used instead of other radioactive noble gases due to the poor production rates, short half-lives, high background levels or issues with global scale monitoring of other noble gases [1]. There are four key radioxenon isotopes used in monitoring: ^{135}Xe (9 hour half-life), ^{133m}Xe (2 day half-life), ^{133}Xe (5 day half-life) and ^{131m}Xe (12 day half-life). By measuring the activity ratios of ^{133m}Xe/^{131m}Xe versus ^{135}Xe/^{133}Xe, it is possible to distinguish between civilian sources (nuclear power plants and medical isotope production facilities) and nuclear explosions [2].

The International Monitoring System (IMS) currently uses large laboratory devices located throughout the world for xenon monitoring, such as the Swedish SAUNA (Swedish Automatic Unit for Noble Gas Acquisition), the French SPALAX (Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radioXenon), or the Russian ARIX (Analyzer for Xenon Measurements). The current systems are large laboratory devices that require a climate controlled building. Treaty verification monitoring for radioxenon is another application for post-event monitoring on-site; however, this is not currently conducted because there are no simple portable field devices for measurements. The objective of this research is to develop the necessary components for a portable beta-gamma coincidence counting system that would allow for on-site monitoring for radioxenon.

Savannah River National Laboratory (SRNL) is a leader in the field of gas collections and has developed highly selective molecular sieves that allow for the collection of xenon gas directly from air. Using these molecular sieves, SRNL has the ability to collect radioxenon on location, but the various commercially available components of a portable beta gamma coincidence counting system have not been integrated into a small, portable fieldable device. This work has focused on the development of custom electronics for beta-gamma coincidence counting using a small, USB powered device. Beta-gamma coincident counting is advantageous for two reasons. First, the measurement sensitivity is improved by a lower of the background, relative to gamma counting alone (Figure 1). Second, more isotopic information is available using beta-gamma counting as opposed to gamma counting alone (Figure 2).

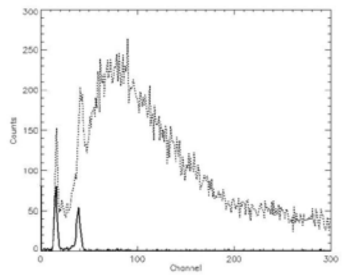


Figure 1: Spectra of ^{133}Xe showing background reduction in coincidence mode

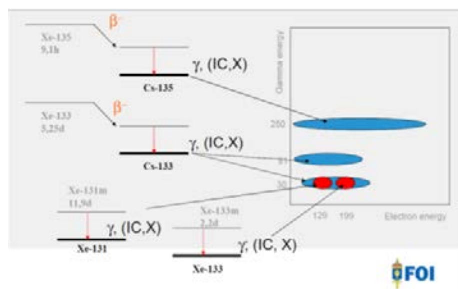


Figure 2: Conceptual 2D beta-gamma spectra [3]

Approach

This project is a continuation of the FY14 LDRD project "Field detector development for undeclared/declared nuclear testing for treaty verification monitoring" and is a collaboration of scientists and engineers. Phase I began the assessment of a small, robust/portable beta-gamma coincidence counting system using the following approach:

- Demonstrating beta-gamma coincidence counting of xenon using COTS rack mount lab scale electronics and an Ortec digiBase multichannel analyzer
- Initial development of the custom electronics necessary to perform beta-gamma counting using one USB interface
- Evaluation of commercially available hardware from Amptek and Ortec for the beta and gamma spectrometry interfaces
- Design and fabrication of a gas scintillation cell

Phase II of this project focused on continued development of the custom electronics and the continued development of the gas scintillation cell and beta-gamma detector characterization. For the development of the custom electronics, two approaches were taken to address the timing challenges and integration of the electronics and data collection. The first was the development of a Visual Studio software application and the second was the design of an embedded hardware circuit board. The continued development of the gas scintillation cell looked and fabrication using three different plastic scintillators. A sample of radioxenon gas was collected and used to evaluate the gas cell and beta-gamma detector system.

Results/Discussion

This project continued the development of a small, robust beta-gamma coincidence counting system, which will combine collection and *in situ* detection methodologies. This will be a significant advancement for field detection/quantification of short-lived xenon isotopes that would not survive transport time for laboratory analysis. During Phase I of the “Field detector development for undeclared/declared nuclear testing for treaty verification monitoring” LDRD, we successfully performed baseline xenon beta-gamma coincidence counting with lab scale electronics and then demonstrated that an Ortec digiBase could perform similar beta-gamma coincidence counting. Using a ^{181}Hf solution, a background reduction of more than 500x was achieved for measuring beta-gamma coincidence signals over measuring gamma signals alone with an unshielded detector resulting in a sensitivity improvement of almost 25x, see Figure 3. The initial development of the custom electronics necessary for beta-gamma counting started with an evaluation of the electronic circuit configuration and selection of the multichannel analyzers for the beta spectrometry and gamma spectroscopy interfaces, Amptek DP5G and Amptek TB-5, respectively, see Figure 4. A gas cell was designed and fabricated with Eljen Technology plastic scintillators. Two different geometry designs for the gas cells were considered: a circular designs and a square cell design, see Figure 5. The two gas cell designs were fabricated and evaluated; and the circular cell design was chosen for further testing.

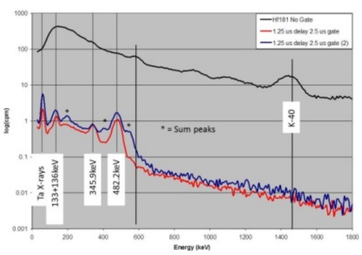


Figure 3: Spectra of ^{181}Hf using no gate delay and a 1.25 us gate delay, a ~500x background reduction is achieved

Figure 4: Electronic components selected for beta-gamma coincidence system

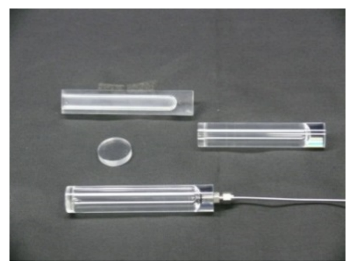


Figure 5: Gas cell protoypes

Phase II of this project focused on the continued development of the custom electronics necessary to perform beta-gamma coincidence counting. The beta-gamma coincidence counting system consists of three basic components: 1) the beta subsystem, 2) the gamma subsystem and 3) a controller to perform basic setup and control of the Beta/Gamma subsystems and creation of a data stream for analysis, see Figure 6. The beta subsystem processes incoming pulses from the beta PMT to create a time record with beta energy. The beta pulse produces an external signal which is applied to the gamma subsystem and serves as a gate to enable gamma data collection (timestamp and energy). The gamma pulses outside this gate window are ignored. Upon receipt of a valid gamma pulse, the controller searches the beta record for a matching timestamp and creates a record of the coincident beta and gamma pulse energies and timestamps. Two approaches were taken to address the timing challenges and integration of the electronics and data collection: development of a custom Visual Studio software application and design of a custom embedded hardware circuit board. The Visual Studio software application is a PC based application that allows the user to control the set-up of both the beta and gamma systems prior to data collection. The software script synchronizes the time bases between the two systems and analyzes the data streams for coincident data sets, see Figure 7. The advantage of the Visual Studio software application is that it allows for better user interaction during the development cycle including enhanced monitoring of the beta/gamma subsystems, more control over hardware setup and operation and easily obtainable data records for faster analysis. The embedded system is a small printed circuit board with an integral microcontroller that will control all beta/gamma subsystem setup and control. It will also produce the output file stream containing the beta-gamma coincidence data. This hardware is tailored specifically for embedding in field measurement systems and provides little in the way of a user interface with its primary interface target being another microcontroller. Both the desktop and embedded solutions utilize the same beta-gamma collection hardware.

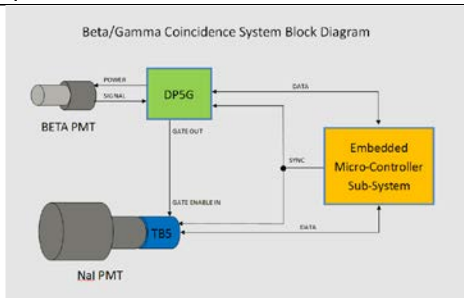


Figure 6: Block diagram for the beta-gamma coincidence system

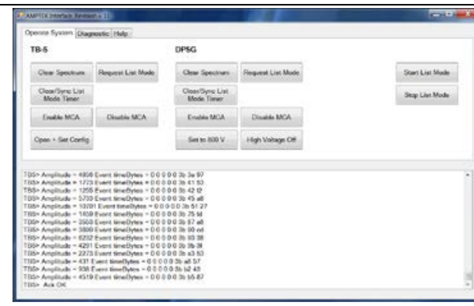


Figure 7: Visual Studio application interface screen.

In addition to the custom electronics development work, the LDRD made significant progress in the development and testing of the gas scintillation cell and the beta-gamma detector characterization. Scintillation gas cells were designed to fit within existing NaI well detectors and were fabricated using three different scintillation plastics from Eljen Technology: EJ-200, EJ-204, & EJ-212. These scintillators were chosen based on their overall properties and are equivalent to other scintillators used in literature (i.e. EJ-204 is equivalent to the St. Gobain BC-404 plastic scintillator used in the SAUNA system, EJ-212 is equivalent to St. Gobain BC-400 used in Phoswich detectors, and EJ-200 is a good general purpose scintillator). The impact of the epoxy used in fabrication of the gas cell and the silicon interface for the PMT on the background of the beta spectrum was investigated and these components were found to have no contribution to the beta spectrum. The Amptek DP5G MCA performance was evaluated using a ^{207}Bi sample and was found to perform in an equivalent manner to the Ortec digiBase. Using the beta-gamma coincidence counting, a reduction in background of 320x was achieved, see Figure 8, and the ^{40}K contaminate peak is no longer present. A screen-shot of the oscilloscope trace detailing the gate window for allowing beta-gamma coincidence is shown in Figure 9.

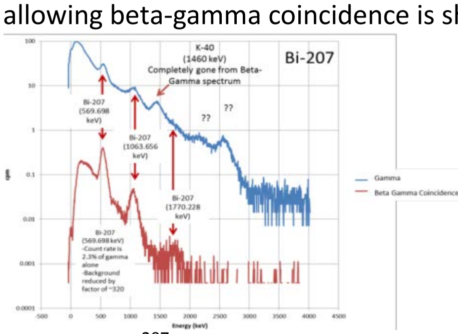


Figure 8: ^{207}Bi spectrum showing a background reduction of ~320x in beta-gamma coincidence mode

Figure 9: Oscilloscope trace showing the beta pulse triggering the gamma

To evaluate the gas scintillation cell and the beta-gamma detector system, a sample of radioxenon gas was collected and analyzed using the experimental set-up shown in Figure 10 below. The sample of radioxenon gas was characterized in an existing HPGe well detector before ~ 3% of the gas was transferred into the gas scintillation cell for analysis.

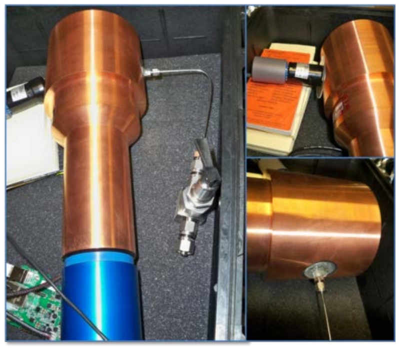


Figure 10:Experiment set-up with gas cell & Amptek components

The resulting beta-gamma coincidence spectrum of the radioxenon gas, Figure 11, shows a significant reduction in background in addition to the appearance of the ^{135}Xe peak. Using the coincidence counting mode, a background reduction of ~320x was achieved. This background reduction corresponds to a ~17.9x improvement in sensitivity based on uncertainty calculated from the square root of the count rate. The counting efficiency of the beta scintillation pulses varied from 50-90% depending on the PMT voltage settings. Spectra were taken over a 14 day period to evaluate the rate of gas leaking from the gas scintillation cell and there was no observed gas cell leakage over the first 48 hours, shown in Figure 12. Based on the 9.1 hour half-life of ^{135}Xe, the CTBT uses a 12 hour count period for samples; therefore the SRNL gas cell design meets requirements for a fieldable system.

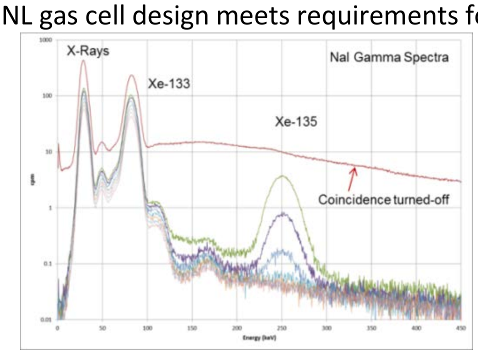


Figure 11: Spectrum of xenon gas showing the ^{133}Xe & ^{135}Xe isotopes in coincidence mode

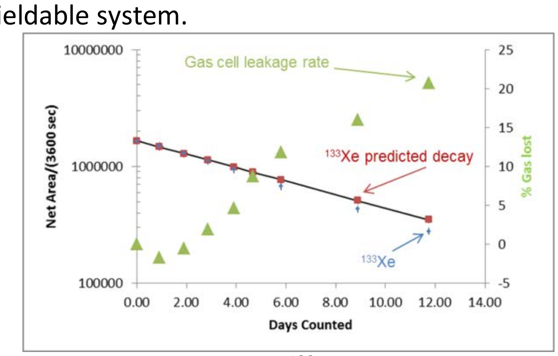


Figure 12: Spectra of ^{133}Xe decay over time correlated with scintillation cell gas leakage

The ^{135}Xe peak at 250 keV was used to calculate the MDA in an unshielded system and was corrected for the ingrowth of the ^{133m}Xe peak, see Figure 13. The ^{133m}Xe was less than a percent of the sample at the start of counting, but was subsequently detectable as the ^{133}Xe decayed. As Figure 12 indicates, the gas leakage from the cell during the 5 day counting period was only 5% and did not impact the ^{135}Xe decay and ^{133m}Xe ingrowth.

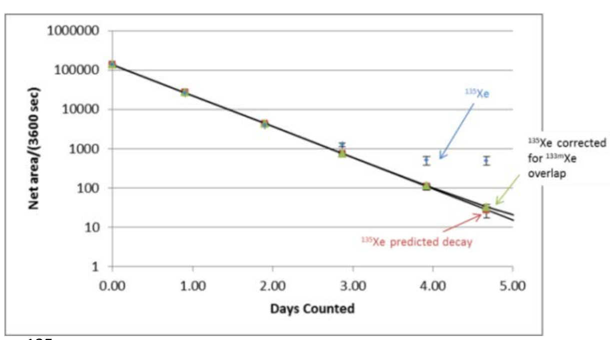


Figure 13: ^{135}Xe decay over time. Blue diamonds show measured peak

area, the green triangles are corrected for the ^{133m}Xe peak overlap and the red squares are predicted ^{135}Xe decay based on the half-life

Analysis of the radioxenon sample in the gas scintillation cell demonstrated that the existing zeolite collection transfer technology is suitable for use with the beta-gamma gas scintillation cell. The experimental set-up shown in Figure 10 demonstrated that beta-gamma coincidence counting is feasible using the Amptek hardware and the development of the Visual Studio application script will demonstrate that pulse height analysis is possible with the Amptek hardware, which will allow for analysis of all 4 xenon isotopes with 2D beta-gamma plots. The embedded hardware circuit board is the implementation of these two concepts in a fieldable form.

FY2015 Accomplishments

- Fabrication of gas cells with 3 different scintillation material (i.e. EJ-200, EJ-204, EJ-212)
- Examined beta spectra of scintillation gas cell components: No contribution to the beta spectrum from the epoxy used in gas cell fabrication or the silicon PMT interface
- Selected Amptek TB-5 for gamma interface and Amptek DP5G for beta PMT interface
- Fabrication of 3 testing systems for electronics development and gas cell calibration/nuclear counting testing
- Proved that beta-gamma coincidence was possible with the Amptek modules through both selective gating and data stream collection
- Initial development of the custom Visual Studio Application software
- Initial design and development of the custom embedded beta-gamma control hardware.
- Tested the gas cell and beta-gamma detectors with radioxenon gas sample
- Beta counting was 50- 90% efficient depending on PMT voltage
- Gas scintillation cell showed no leakage in first 48 hours
- Demonstrated detector linearity over five orders of magnitude for both the beta as well as gamma detectors

Future Directions

- Continued development of the design for integrated microcontroller and fabrication of microcontroller
- Comparison of the EJ-200, EJ-204 and EJ-212 scintillation plastics
- Collection and analysis of all four radioxenon isotopes using the developed 2D beta-gamma system
- Prepare manuscript for publication

FY 2015 Publications/Presentations

1. A mid-year progress presentation was given at SRNL.
2. A presentation detailing the project concepts and the progress of Phase I/Phase II work was given to potential government funding sources to leverage follow-on funding after project completion.

References

1. Zähringer et al., J Radioanal Nucl Chem, 282, 2009, 737-742

2. Kalinowski et al., *Pure Appl Geophys*, 167, 2010, 517-539
3. Fritioff, T. How to detect a nuclear explosion using a SAUNA, 2011 presentation

Acronyms

ARIX: Analyzer for Xenon Measurements
COTS: Commercial off the shelf
CTBT: Comprehensive Nuclear Test Ban Treaty
IMS: International Monitoring System
MCA: Multichannel analyzer
PMT: Photomultipler tube
SAUNA: Swedish Automatic Unit for Noble Gas Acquisition
SPALAX: Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radioxenon
SRNL: Savannah River National Laboratory

Intellectual Property

Invention disclosure will be submitted for the embedded beta-gamma control hardware and its application.

Total Number of Post-Doctoral Researchers

Zero