EVALUATION OF HPXe NUCLEAR EXPLOSION MONITOR APPLICATIONS

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ABSTRACT

The selection of a detector type for field gamma-spectroscopy monitoring applications is usually based on achieving an acceptable balance of detector energy resolution, efficiency, practicality in the measurement environment, and cost. Because energy resolution and photopeak efficiency of the detector are key factors in determining the detection sensitivity limit, these parameters usually must be addressed first to ensure that the measurement objectives of the mission can be met by the system. For automated spectroscopy systems viewing a complex radionuclide mixture, the unmatched resolution of germanium detectors frequently makes the High Purity Germanium (HPGe) the detector of choice, in spite of the relatively high cost, requirements for cryogenic cooling, and limitations on size and ruggedness of germanium detectors. Promising results are being realized for improvements in the performance of several alternate room temperature detector types in areas of compound semiconductors, halide scintillators and high pressure gas detectors.

One detector type that has shown recent progress as a rugged high-temperature detector with better energy resolution is the high pressure xenon (HPXe) detector. Current HPXe detectors deliver energy resolutions in the range of 1.7%–2.0 % at 662 keV and 7–8 keV FWHM for low-energy gamma rays. Although HPXe detector resolution will likely never match the 0.3-0.5 % resolution of large HPGe detectors, the resolution is quite acceptable for many field applications, and provides other benefits for field systems such as large volume and extended stable operation without cooling. Excellent temperature stability (0.2% total over 23°C–80°C) and negligible drift over time are now being achieved, thus facilitating energy calibration and long-term operation of unattended systems. Current R&D programs are expected to produce HPXe detectors with active detector mass in the few Kg range, and surface areas of 1–3 square feet. These promising developments have prompted recent consideration of HPXe systems for specific harsh environment applications in homeland security, power reactor fuel failure monitors, environmental monitoring stations and “bore hole” measurements at nuclear facility cleanup sites.

In this present work, the performance potential for HPXe-based systems for Nuclear Explosion Monitoring (NEM) applications will be evaluated. A candidate HPXe detector is being selected and characterized in a series of test measurements. A NEM-specific Monte Carlo N-Particle (MCNP) model will be developed for use in optimizing and evaluating the performance of promising NEM configurations. Data from the test measurements will be used to verify the accuracy of the MCNP model and to predict detection sensitivity capabilities for selected radionuclides in the presence of expected NEM interference. If warranted by the performance predictions for HPXe-based nuclear explosion monitors, the MCNP model will be used to define an optimized design of an HPXe-based NEM prototype system.

OBJECTIVES

Note: The research and development project described in this publication has been approved for funding, and a contract to initiate the project is expected by mid-July 2006. This paper is a description of the technical basis and objectives of the program, and of the method by which it will be accomplished.

Background

In defining the detector and auxiliary components for a gamma spectroscopy measurement system, the energy resolution, photopeak efficiency, and peak-to-Compton ratios over the gamma energy range of interest are of primary concern in ensuring that the measurement system will have the inherent capability to meet the required detection sensitivity. For an applied gamma spectroscopy system that must perform a single specific mission in a severe environment with limited access to skilled personnel, other detector features such as room temperature operation, system stability, ruggedness, reliability, and ease of maintenance can also be crucial to success. A recent International Atomic Energy Agency (IAEA) program to review the effectiveness of existing field spectroscopy measurement instruments and fixed monitors identified several major applied systems with state-of-the-art measurement capabilities that were removed from operation because their sensitivity provided too many false positives or provided information in a manner than could not be interpreted and constructively used by field personnel (Arlt, 2005). The IAEA report indicated that a system that requires support and maintenance to the extent of interfering with the primary role of the operator will likely have a very poor availability record. The different criteria for success in a laboratory system and a field gamma spectroscopy system requires that much more consideration of operational issues must be included in the design of a field system if it is to successfully meet the mission role for which it was intended.

If the purpose of a gamma spectroscopy system is to provide a variety of low level measurements in the presence of interference from multiple radionuclides, the excellent energy resolution of an HPGe detector system usually makes it the detector of choice because it offers superior detection sensitivity that simplifies the data analysis process. In a research laboratory environment, resources are normally available to provide cryogenic cooling, background reduction, stable power, minimal shock and vibration, access to replacement parts, and a support staff of experts in detectors and instrumentation. Also, in the laboratory research environment, spectroscopy system downtime is inconvenient, but does not represent a potentially life-threatening emergency that can be associated with unavailability of crucial applied nuclear monitoring systems.

By contrast, NEM, and homeland security and nuclear plant applications of automated gamma spectrometer monitors are examples of systems that must perform a fixed crucial mission with very little downtime over a wide range of severe environmental and logistical conditions. In several field applications of automated gamma spectrometry based monitoring, HPGe detector-based systems have been used successfully to demonstrate that the desired measurement can be made in the field if adequate resources for support can be provided. The Radionuclide Aerosol Sampler Analyzer (RASA) system for nuclear explosion monitoring clearly has demonstrated the inherent capability of that system to identify and quantify radionuclides from an nuclear explosion (Miley, 1998). RASA operational problems related to the reliability of the HPGe detectors and the filter system have been addressed and mitigated (Miley et al., 2000).

Tests of several HPGe based Advanced Spectroscopic Portal Monitors have demonstrated the ability to resolve complex spectra, but the HPGe system is not recommended as the detection system of choice by Arlt (2005). For nuclear power plant applications, automated HPGe based systems have provided fuel failure detection (Walker and Mullin 1996) and post accident sampling (Serpa et al., 1981) capabilities for more than two decades. These nuclear plant systems provide regulatory compliance and operational benefit that justifies the added manpower and radiation dose for providing maintenance and continuous cryogenic cooling of the automated HPGe-based systems in high radiation areas.

For each of these field based automated gamma spectroscopy systems, the practical advantages of a rugged room temperature detector of sufficient resolution and efficiency would be significant. Within the last few years, several promising detector candidates have emerged that offer resolution between that of HPGe (0.2%–0.5% at 662 keV) and NaI (6%–8%) (Park et al., 2006). In particular, three alternate room temperature detector types are now available as commercial detector products offering energy resolution in the range of 1.5% to 3% with acceptable

efficiency for many field monitoring applications. CdZnTe semiconductor detectors have been reported with an energy resolution of 1.9% for a 2.25 cc detector, and of 4% for an 11 cc detector (Gostilo et al., 2005). A commercial scintillation supplier recently reported the availability of a 3" x 3" $LaBa_3(Ce)$ scintillation detector with resolution of 2.8%, and the production of a 10.5 cm x 10.5 cm (4.1" x 4.1") detector (Rozsa et al., 2006). HPXe detectors of 1.5" diameter x 3" active length and 1.7% resolution are commercially available from Mirmar Sensor, as are detectors as large as 4.5" diameter by 24". Park et al., (2006) states that, "A recent demonstration at Los Alamos National Laboratory (LANL) showed HPXe detector performance may be superior to that of NaI, plastic scintillators, high-purity germanium, or lanthanum halide/bromide detector materials for many homeland security applications."

Each of the three room temperature detector types now appear to have useful field applications in nonproliferation monitoring, homeland security, nuclear power, and nuclear site cleanup. Their application to practical field systems will require a careful matching of detailed mission requirements with the performance capabilities and operational advantages of these detectors with intermediate resolution. In many cases, the design freedom offered by room temperature detectors will present opportunities for improved sampling systems and shielding configurations to boost detection sensitivity and system reliability.

Specific Objectives of this Project

The general purpose of this R&D project is to determine how to best apply higher resolution, room temperature HPXe detectors to produce rugged, user-friendly field gamma spectroscopy for the NEM program. The project will focus on matching the present capabilities of HPXe detector systems with both the measurement requirements and operational considerations for specific NEM applications. The HPXe system development program will draw heavily on the source description data, operating experiences and lessons-learned from the decade of experience of National Nuclear Security Administration (NNSA) and its contractors in the worldwide deployment of RASA and Automated Radioxenon Analyzer/Sampler (ARSA) systems.

Specific objectives of this project are as follows:

1. Identify an optimum HPXe detector for a NEM application, and to produce measured parametric detector performance data for the detector to allow calculations of detection sensitivity limits for evaluating various NEM applications,
2. Develop and verify an MCNP model for HPXe-based systems that can provide quantitative evaluation of the performance of a variety of HPXe-based system designs for specific NEM applications,
3. Define and optimize a conceptual design for the most promising application of HPXe detectors to an NEM program need, and to propose and predict the performance of a prototype HPXe system to be constructed and tested in a field environment situation.

RESEARCH ACCOMPLISHED

This section will describe portions of previous HPXe detector development results and detector performance of HPXe detectors developed and produced by Mirmar Sensor (Mirmar). The R&D program in basic HPXe detector technology is still in progress at Mirmar, and the HPXe detectors for this pending project will be supplied from the inventory of detectors at the Mirmar facility. The purpose of this current project for NNSA is to define and evaluate the performance of HPXe-based spectroscopy systems for applications in nuclear explosion monitoring. It is being conducted under a joint venture agreement between Mirmar and Quantum Technology, an applied spectroscopy systems company.

Status of Mirmar HPXe Detector Development

Mirmar Sensor now builds xenon detectors in the USA in standard active volume sizes (diameter and length) of 1.5" by 3.0", 1.5" by 6.0", 4.5". by 8.0", and 4.5" by 24". Typical energy resolution is less than 2%, and detectors in the 1.7% range are routinely produced. The Model 121 detectors shown in Figure 1 have an integral preamplifier and high-voltage power supply, and are self-contained in that they requires only ±12 V to operate. They have active detector dimensions of 1.5" diameter by 3" length, and overall dimensions of 1.75" diameter by 19" long. The internal microprocessor raises the high-voltage, then signals (via an LED) that the detector is ready for operation.

Figure 1. Mirmar Model 121 High-Pressure Xenon Detectors.

Figure 2 shows a measured spectrum of a ^{137} Cs source taken with the Model 121 HPXe detector shown in Figure 1. The FWHM resolution at the 662 keV peak is 11.0 keV, or less than 1.7%. Note the distinct xenon x-ray peak at 30 kev below the 662 keV line, which shows that low-energy tailing is not significant for this detector.

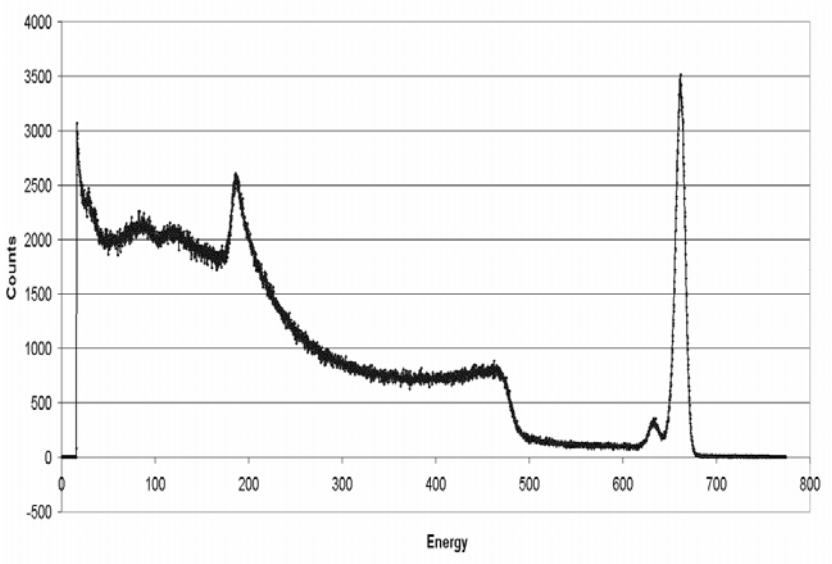

Figure 2. Measured ^{137}Cs spectrum with HPXe Detector Model 121 (Beyerle, 2005).

Stability

The gain of the xenon detectors is quite stable as shown in Figure 3. The time variance of the gain is about 0.02% over a week. This stability eliminates the need for periodic calibration. More importantly, the stability allows the peak location to be very accurately known, which can be useful in quantitative analysis of unresolved peaks. This is important in this application because there are several cases of unresolved or barely resolved doublets.

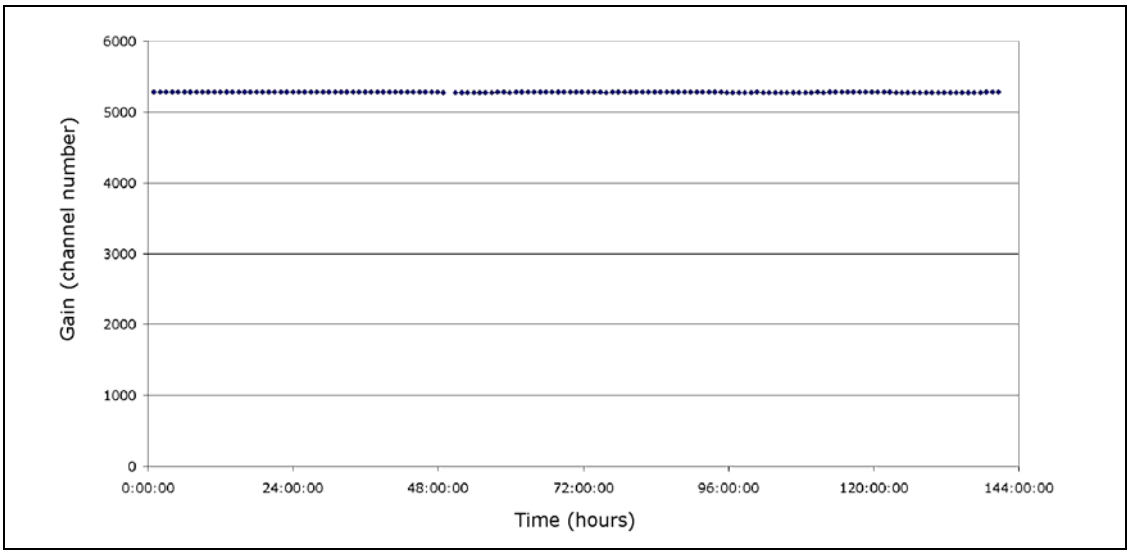

Figure 3. Gain variation over a week for an HPXe detector (Beyerle, 2005).

The temperature stability of the HPXe detector is shown in Figure 4. The temperature stability is about 0.2% over the entire range 25°C–80°C (77°F–176°F). Even this excellent stability range is an upper limit since the precision of the measurement is comparable to the measured variation. Temperature stability is important in applications such as outdoor air monitoring or vehicle portal monitoring systems where the detector temperature may change significantly on a daily and seasonal basis.

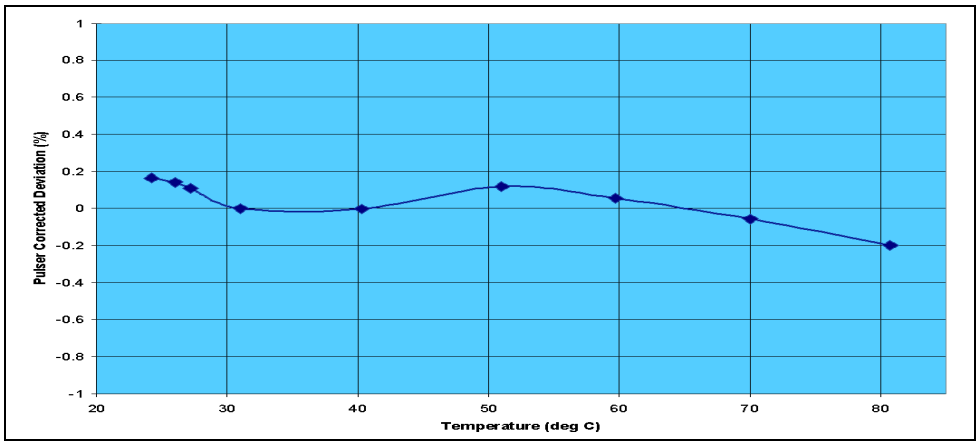

Figure 4. Temperature Stability of HPXe (Beyerle, 2005).

Figure 5 shows a ^{133}Ba spectrum taken with the Model 121 HPXe detector. Significant features of the spectrum include the following:

- The peak at 30 keV is nearly entirely due to the xenon x-ray at 30 keV. The vessel wall is not very transparent at 30 keV. A carbon fiber or aluminum vessel would alleviate this problem.
- The peak at 50 keV is a combination of the x-ray escape from the 81.0/ 79.6 keV peak and the ^{133}Ba 53.17-keV peak.
- The peak at about 80 keV is the ^{133}Ba 81.0 and 79.6 keV peaks. The resolution is not nearly good enough to separate the peaks but the excellent symmetry of the peaks allows some distinction of the closely separated small peak immediately lower in energy than a larger peak.
- The ^{133}Ba peak at 160.6 keV is obscured by the Compton backscatter. This is a reason to use larger diameter detectors.
- The ^{133}Ba peak at 223-keV is barely visible due to its small intensity.
- The standard four ^{133}Ba lines at 276, 303, 356, and 384 are clearly separated. Each has an x-ray escape. Especially noticeable is the x-ray escape of the largest 356-keV peak at 326 keV.
- Higher energy peaks are pile-up.

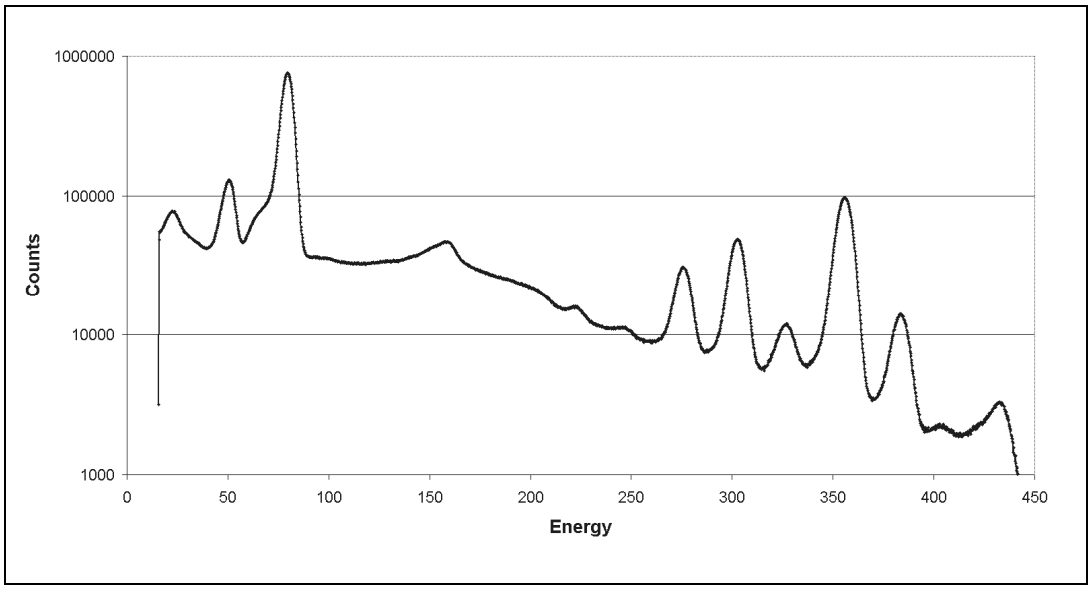

Figure 5. The ^{133}Ba spectrum from a small xenon detector. Low-energy FWHM is 6–7 keV (Beyerle, 2005).

Vibration Issues

Mirmar Sensor has developed a patented technology to greatly reduce the vibration sensitivity of the HPXe detectors. Detectors are generally unaffected by vibration up to about 0.2–0.5 G vibration at any frequency. External damping can further reduce this sensitivity.

Nuclear Test Monitoring

We have performed a first order simulation of the nuclear test air monitoring spectra. This simulation was accomplished by taking a ^{133}Ba spectrum with its gamma-ray lines in the relevant energy regime, and simultaneously fitting all of the peaks. The gamma-ray energies of the ^{133}Ba were replaced with the energies of the actual fallout gamma-rays from Miley (1999). with all of the intensities equal. The resulting spectra are shown in Figures 6 and 7. This was fit over an actual depleted uranium spectrum, hence the fitted peaks (red/solid) bear no

resemblance to the real peaks (blue/dotted) except to give an idea of the statistical noise and the quality of the fit in the one case (U-238 at 1001 keV) where there is alignment.

Note that in each case, there is an irresolvable overlap between the ^{147}Nd and ^{140}Ba at 537 and 531 keV that must be resolved by the ^{140}Ba at 487 keV. The ^{95}Zn (in the fit in red/solid) at 768 keV has a conflict with the natural Thorium line (from the data in blue/dotted), which is resolved by the ^{95}Zn peak at 739 keV.

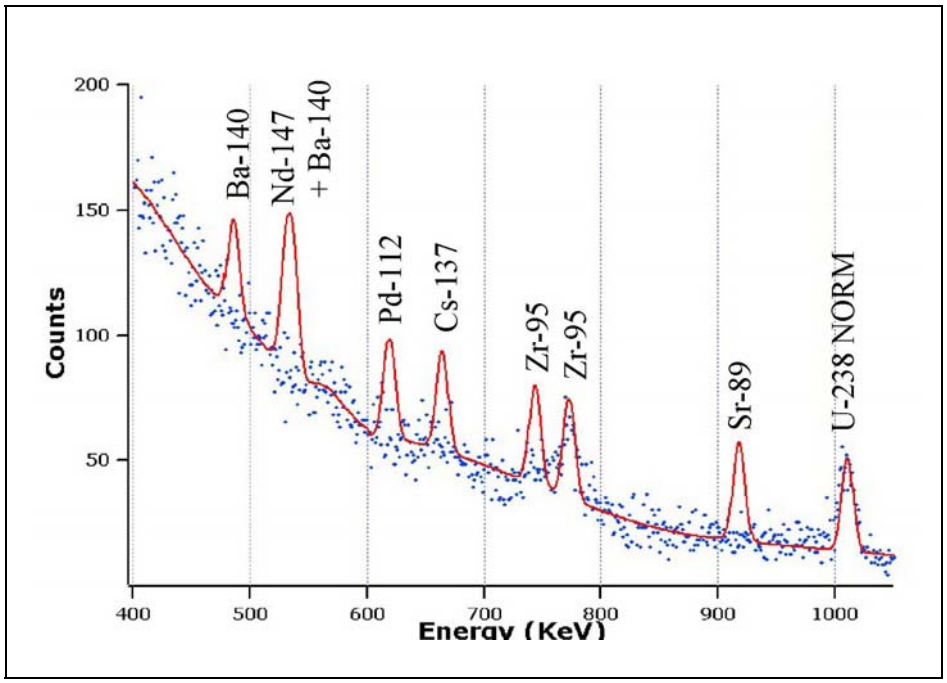

Figure 6. Simulated portion of a spectrum for filter collection 14 days after a nuclear event.

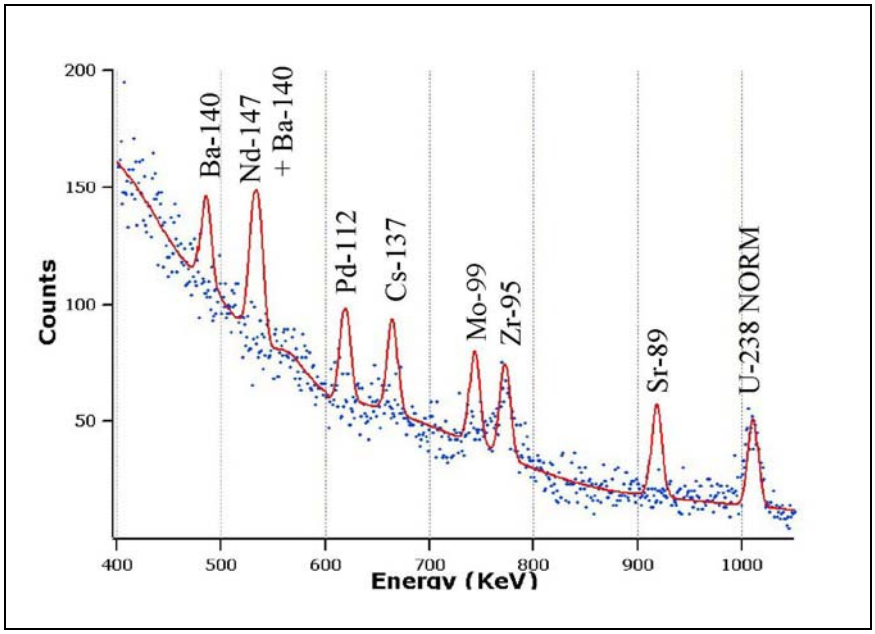

Figure 7. Simulated spectrum for filter collection 2 days after a nuclear event.

Project Plan

The purpose of this project is to evaluate the performance of HPXe-based field spectroscopy systems for nuclear explosion monitoring applications and to advance a selected applied HPXe monitor to the system prototype stage. The first phase of the effort will consist of the following tasks that will be executed in parallel:

1. HPXe Detector Selection and Characterization—HPXe detectors will be selected and used in a series of test measurements with National Institute of Standards and Technology (NIST) traceable sources to generate parameterization data of energy resolution vs. energy, efficiency vs. energy, and Compton baseline over the energy range of 60 keV to 2,000 keV. Data will also be recorded with selected sources representative of NEM and interference sources in field applications. These data will be used to calculate detection sensitivity for cases of interest, and to test the accuracy of Monte Carlo models of HPXe based measurement systems. Data will also be collected to allow prediction of stability vs. time, microphonic effects, and to evaluate optimum pulse processing and data analysis methods for HPXe spectra.
2. Development and Verification of a Monte Carlo Model for Applied HPXe-Based Measurement Systems—A Monte Carlo model will be developed for applied HPXe systems based on the MCNP-5 code. The experimental results from Task 1 will be used to verify the accuracy of the Monte Carlo model, and to estimate the error bounds of the calculated results. Auxiliary programs will be written to transfer calculated spectra into commercial spectroscopy programs to facilitate evaluation of MDC performance and resolution of interference components in the HPXe spectra.
3. Identification and Evaluation of promising NEM applications using HPXe Systems—Promising NEM applications for which HPXe based field systems may offer advantages will be identified based on NEM mission requirements and the data, and operating experiences and lessons learned of the NEM community. Alternate sample collection and data processing and analysis methods will be examined that take advantages of the large size, ruggedness, and flexibility of shielding of HPXe detectors. The MCNP model will be used to quantify the performance of various system configurations, and to optimize system designs for maximum performance. This collective information will be used to select and optimize a prototype design for an HPXe NEM system for specific field applications. The prototype system will be constructed and tested in a field environment situation in subsequent phases of this project.

CONCLUSION(S) AND RECOMMENDATIONS

HPXe detector technology has progressed to a point that justifies an authoritative evaluation of HPXe systems for specific applications in nuclear explosion monitoring. HPXe detectors are viable candidates for nuclear test monitoring because large area spectrometers with excellent stability are available. Detectors have been operated out of doors in air sampling systems successfully for more than a year maintaining 2% resolution, with excellent stability. Their large area makes them the most sensitive choice where a large area sample, such as a large filter, is involved.

This study is a focused program to develop and apply tools to define and optimize HPXe systems that provide effective, practical field systems for NEM applications.

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