### ASSEMBLY AND INITIAL RESULTS: AN ULTRA-LOW-BACKGROUND GERMANIUM CRYSTAL ARRAY FOR HIGH EFFICIENCY AND COINCIDENCE MEASUREMENTS

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#### ABSTRACT

Treaty verification, environmental surveillance, and physics experiments continue to require increased sensitivity for detecting and quantifying radionuclides of interest using gamma-ray spectrometry. This can be accomplished by establishing high detection efficiency and reducing instrument backgrounds. A current effort for increased sensitivity in high resolution gamma spectroscopy will produce an intrinsic germanium (HPGe) array designed for high detection efficiency, ultra-low-background performance, and useful coincidence efficiencies. The system design is optimized to accommodate filter paper samples, e.g., samples collected by the Radionuclide Aerosol Sampler/Analyzer (RASA). The system will provide high sensitivity for weak collections on atmospheric filter samples, as well as offering the potential to gather additional information from more active filters using gamma cascade coincidence detection. The instrument will also be capable of making advances in low-dose neutron activation analysis. The first of two ultra-low-background vacuum cryostats has been assembled, with a second in progress. Traditional methods for constructing ultra-low-background detectors were followed, including use of materials known to be low in radioactive contaminants, use of ultra pure reagents, clean room assembly, etc. The cryostat is constructed mainly from copper electroformed into near-final geometry at Pacific Northwest National Laboratory (PNNL). Details of the detector assembly, vacuum tests, and test measurement results are presented.

# **OBJECTIVES**

Two ultra-low-background cryostats are currently under construction for the NA-22 funded "Radionuclide Laboratories" (RN Labs) project. The design and performance expectations for this instrument have been discussed previously (Keillor et al, 2008 and 2009). Each cryostat will house seven high-purity germanium crystals (HPGe). We are building these cryostats from a limited set of materials that are known to have very low levels of radioactive impurities. The vast majority of each cryostat is made from pure copper (< 1 $\mu$ Bq/kg, see Aalseth et al., 2009) electroformed into near-final geometry at PNNL under class 1000 clean room conditions.

The RN Labs instrument is designed to take advantage of low background performance, high detection efficiency, and  $\gamma$ - $\gamma$  coincidence signatures to provide unprecedented gamma spectroscopy sensitivity. The project is focused on improving gamma analysis capabilities for Nuclear Detonation Detection (NDD) applications, e.g., nuclear treaty monitoring. The instrument also has the potential for basic nuclear physics research. For example, the potential for this detector to measure the half-life of predicted rare decay modes of <sup>130</sup>Te is being investigated. In addition, we have also built and tested a large area multi-wire proportional counter (MWPC) active anti-cosmic detector (Soplata et al, 2009). An active anti-cosmic system is a crucial component to allow the instrument to achieve high sensitivities for NDD measurements.

# **RESEARCH ACCOMPLISHED**

This paper details the acceptance testing of HPGe crystals refurbished by Princeton Gamma Tech (PGT) and crystals purchased new from Canberra Semiconductor. It also provides an update on the current status of copper electroforming for the project. The results of initial tests with plastic scintillator anti-cosmic detectors are briefly discussed, as are the design and testing of analog signal processing electronics. Finally, results of vacuum testing for the first cryostat are covered.

## **Initial Germanium Crystal Testing**

HPGe crystals were obtained from both PGT and Canberra Semiconductor for use in the RN Labs instrument. The crystals are nominally 63-mm-diameter and 70-mm-height semi-coaxial P-type detectors with a guard "ditch" surrounding the central bore hole. The fragile contact is protected by gold plating at the entrance of the bore hole. Eight HPGe crystals available from a previous project (Kazkaz et al., 2003) were available for use on the RN Labs research. A contract was arranged with PGT to refurbish these eight crystals. All eight of these crystals were received, tested, and removed from the shipping cryostats. The agreement with PGT did not include an energy-resolution specification for these refurbished crystals, so acceptance was based on the results of an I-V curve measurement. Figure 1 is an example of a typical I-V curve for these PGT crystals. The crystals were stored in a nitrogen dry box for various periods of time (up to  $\sim$ 1 year) after initial testing and then moved to vacuum storage. The crystals are currently in vacuum storage awaiting installation in one of the RN Labs cryostats.



## RN LABS Crystal 3057

Figure 1. I-V Curve for One of the HPGe Crystals Refurbished by PGT. Energy resolution at 1332 keV is also plotted.

An initial shipment of four new crystals from Canberra Semiconductor was tested against factory specifications. After testing, the first of these crystals was removed from its shipping cryostat. Upon removal, we noted that (1) excessive amounts of indium were used on the crystal surface (inconsistent with a low background detector (Figure 2) and (2) the "pogo pin" style of signal contact was used. This latter point made the crystals unsuitable for use in RN Labs, both due to indium at the bottom of the central bore hole and the lack of protective gold plating at the entrance to the bore hole (required for the wire-loop style contact). These features were not consistent with the order specifications, so all four detectors were returned to Canberra for repair. This issue delayed the receipt of the final eight crystals, however Canberra recognized the discrepancy and has returned the original 4 crystals, along with two additional crystals. These detectors have all performed very well in acceptance testing (Figure 3). The eight PGT and six Canberra crystals give us a full complement of 14; the two final crystals due in from Canberra will give us spares in the event that trouble arises with any of the crystals during or after installation in the array.



Figure 2. HPGe Crystal Received from Canberra. The large indium patch is the silver colored material at the top of the image. The end of the "pogo pin" contact rod is visible inside the bore hole, and the visible end of the bore hole is not plated with gold.



RN LABS Crystal 09089454

Figure 3. I-V curve typical of the Canberra crystals. The energy resolution at 1332 keV is also plotted.

## **Electroforming Update**

In general, electroforming parts for RN Labs has proceeded smoothly, and the vast majority of parts have been successfully completed. Figure 4 shows nearly all of the copper parts required for the first cryostat, along with the electroformed cross arm and cold finger for the second cryostat (still on mandrels in this image).

Unfortunately, we have experienced difficulty in electroforming three specific large copper parts necessary for the project. The affected parts are the infrared radiation (IR) shields and the domed entrance window. The IR shields are very thin parts that take a matter of a few days to electroform, so we anticipate that there is sufficient time to resolve the issues discussed below and complete these parts. The entrance window is a much more challenging part, both due to its geometry and its thickness. This part will take  $\sim$  two to three months to electroform so that completing the thin entrance window within FY09 depends upon rapid resolution of poor copper nucleation and adhesion issues, or upon developing an alternative solution.

Obtaining proper copper nucleation and adhesion to the stainless steel mandrels is essential to proper growth of the part. Figure 5 displays two separate attempts to electroform an entrance window and an attempt at one of the IR shields. These parts all suffered from poor nucleation so that the resulting copper had significant holes. Holes are not acceptable for either the entrance window (which must hold vacuum) or the IR shields (which must prevent infrared radiation from striking the HPGe crystals). We are in the process of determining the source of these issues.

Additives employed in typical electrochemical manufacturing processes aid in the development of nucleation. However, to obtain the greatest purity copper possible, we do not use additives since it has been established that these materials and even inert particles such as alumina (from abrasives used for surface preparation) can be incorporated in the growing copper matrix during electrodeposition.

To determine the nucleation and growth mechanisms at play under the electrodeposition conditions used, various copper samples were analyzed via scanning electron microscopy (SEM) with electron backscatter diffraction (EBSD) to determine their microstructure, including grain size and orientation. Secondary ionization mass spectrometry (SIMS) and laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) were also performed to determine relative surface concentrations of impurities and to map impurity localization on the copper surface.



Figure 4. Completed parts for the first cryostat of the RN Labs HPGe array.





Figure 5. Copper Nucleation Problems. Large electroformed parts that have suffered nucleation issues, resulting in numerous holes in the copper.

For SEM and SIMS analysis, samples were mounted in epoxy for metallographic polishing and given a series of successively finer-grit mechanical polishing steps using diamond grits with a final polish of colloidal silica. For LA-ICP-MS, electroplated copper samples were etched in  $3\% H_2O_2/1\% H_2SO_4$ .

All of the samples showed a very fine grain size at the interior surface directly in contact with the mandrel (Figure 6). After a few hundred microns, this tended to transition to a larger elongated grain structure that often had a strong texture. For some samples, the texture continued to refine itself out to the edge of the sample, producing very large, highly oriented grains (Figure 7).



Figure 6. SEM image showing transition of the copper grain structure.



Figure 7. SEM image showing large, oriented grain structure.

These data provide evidence for instantaneous nucleation and highly selective grain growth during the electrodeposition process. Further work is needed to distinguish between nucleation and growth patterns and the effects of self-annealing or recrystallization.

The solution to the difficulties encountered in electroplating these large parts is to provide more controlled surface conditions that favor uniform nucleation on the stainless steel mandrels. Electropolishing, an electrochemical cleaning process, is being aggressively investigated to resolve the problem. Chemical etching is another possible solution that is also being tested.

We are also investigating alternative plating geometries that would be quicker to produce. Figure 8 shows an alternative entrance window geometry that would allow plating of the entrance window separate from the thicker plate surrounding the window. In this geometry, the window will be electroformed as a 6" diameter part, then e-beam welded into the heavier plate. We are pursuing this alternative in parallel with plating the part as a single unit.



Figure 8. Alternative plating geometry for entrance window. The upper part of the figure shows an entrance window plated on a 6 inch mandrel. This window will then be e-beam welded into the annular plate. This geometry is less likely to suffer from the nucleation issues we have experienced with the larger 11 inch mandrel.

### Anti-cosmic Scintillator Testing

An important part of the overall background performance of the RN Labs instrument is suppression of cosmic-ray backgrounds. Tests have been performed with plastic scintillator cosmic-ray detectors, and a design for an effective active cosmic-ray veto system for RN Labs has been prepared. The results of these initial laboratory tests indicate that two inch thick scintillating plastic panels will provide acceptable cosmic-ray identification while remaining relatively insensitive to external gamma-ray backgrounds (Figure 9). Based on the results of these tests, scintillator paddles for active anti-cosmic shielding have been designed and ordered.



Figure 9. Muon peak in plastic anti-cosmic detector. This figure shows the muon through-peak in a two inch thick plastic scintillator. This spectrum is from "tagged" muons, in other words events observed in two independent scintillators at the same time. Energy is in ADC channel units. The rising continuum at the low energy end is caused by coincident detection of scattered gamma-rays; we anticipate good separation between gamma-rays and muons, as displayed in this spectrum.

#### **Electronics Testing**

Another critical part of the RN Labs array is the amplification and analog signal path for the very small electrical signals that are produced in the HPGe crystals as a result of radiation interactions. A sensitive front-end amplifier made with radiopure materials has been designed and tested, along with a low-background signal cable design.

Final production of the front-end amplifiers for RN Labs has not been completed, but 16 research prototypes have been prepared to support initial instrument testing. These custom PNNL components will be combined with a semicustom commercial preamplifier (BridgePort Instruments) for the final signal conditioning before output signals are digitized for further analysis. Tests with the first revision of the semi-custom preamplifiers are now complete, and several small design revisions were identified to provide better (lower) noise performance. Revised prototypes are expected to be available on time for integration into the RN Labs instrument. A parallel research path using commercial off-the-shelf preamplifiers has also been pursued to reduce project risk and has proven to provide the necessary performance for RN Labs should the revised semi-custom solution not meet expectations.

## Vacuum Testing of Cryostat #1

The vacuum boundary of cryostat #1 has been assembled for vacuum testing. This assembly includes the main body of the cryostat, the cross arm, and the service body (Figure 10). The main body of the cryostat, in this test configuration, requires three seals around the circumference of the chamber. Each seal is ~ 36 inches long. In the final configuration of the cryostat, the plate containing the entrance window will be e-beam welded to the corresponding cylinder, eliminating one of these three seals. Several smaller seals are located on the service body, including the cross arm-to-service body connection, the pump-out port plate, a blank plate, and the service body-to-Dewar connection. The vacuum integrity of welds was tested: e-beam welds on each end of the cross arm, welds on the service body, and a laser weld test for the 50-pin feed-through on the service body. Thunderline Z high-voltage feedthroughs were cemented into the service body with Torr Seal, and these seals were also checked for vacuum leaks.



Figure 10. Vacuum testing of the first RN Labs cryostat. The top plate in this image is a test piece; production of a large, thin entrance window for the cryostat is one of the more challenging aspects of the cryostat production.

The large-diameter seals are created by pressing indium wire into step features on the mating surfaces. Due to the  $\sim 11.5$ " diameter of the cryostat, we have found it necessary to use 0.060" diameter wire. Use of small diameter wire is highly desirable due to the natural radioactivity of indium; the 0.060" diameter is larger than we had hoped for, however is consistent with typical manufacturer recommendations for this size vessel. The 0.49 MeV beta endpoint of <sup>115</sup>In is high enough that some bremsstrahlung contribution from the seals is likely; this will be modeled in a

Monte Carlo simulation in the near future. If it is found that the indium will contribute a significant contribution to the low energy continuum, an alternative metal seal will be investigated.

During leak testing of the cryostat and electrical service body, we were initially unable to achieve the seal for the bottom plate. It appears that the "hoop" warped slightly during e-beam welds of the cross arm and cross arm gussets. A steel form of the correct diameter was machined and pressed into the hoop, correcting the out-of-round condition and allowing the seal to be achieved. Subsequently, the cryostat seals have been leak tested and found to perform at better than 1.0E-10 mb-liter/sec.

The leak testing process also revealed one pin-hole through the electroformed copper material. This hole was found in the large, open ended cylinder forming the side walls of the cryostat. This copper material is 0.125" thick, thus it was somewhat surprising to find a pin-hole (although not unprecedented!). This vacuum leak was successfully repaired with the application of a very small amount of Vacseal to the exterior of the part. The entire copper cryostat was then enclosed in a plastic bag; helium was added to the bag and the leak check response was below 1.0E-10 mb-liter/sec.

Leak testing of the cross arm and service body revealed no response to helium at any of the welds, nor at any of the o-ring seals on the service body. Leaks were observed at three of the high voltage (HV) feedthroughs. These three feedthroughs were accidentally bent when the service body was assembled (see Figure 11); the service body was turned onto the side containing the HV feedthroughs, bending three pins. These three pins were subsequently straightened; however, it is now apparent that the seals between the pins and ceramic insulators were compromised. These feedthroughs will be replaced. We have also fabricated a temporary cover to prevent recurrence of damaged HV feedthroughs.



Figure 11. HV feedthrough pins. Three pins were bent when the service body was accidentally placed with this side down. Leak testing has shown that the vacuum integrity of these feedthroughs was compromised, and they will be replaced. A temporary cover was fabricated to prevent recurrence.

## CONCLUSIONS AND RECOMMENDATIONS

We are progressing rapidly toward construction of the first RN Labs seven crystal array. The vacuum jacket of the first cryostat has been assembled and successfully passed stringent leak testing. Preparations for thermal tests of the Dewar/cold-finger/cold-plate combination are in progress. After successful thermal performance is confirmed, the first half of the array will be populated with HPGe crystals. Preparation of the second cryostat is following shortly behind the first.

Unanticipated challenges with electroforming have delayed the schedule for production of three of the electroformed parts. We expect to resolve these problems with plenty of time to complete the IR shields for the system during FY09. The delays are more problematic for the entrance window because of the long period of time required for it to be electroformed. We have completed a new design for this part, and it is currently being electroformed. In the interim, the existing oxygen-free high-conductivity copper test piece will allow us to proceed with assembly and testing of the first array.

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