Germanium Detectors in Homeland Security at PNNL

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Abstract. Neutron and gamma-ray detection is used for non-proliferation and national security applications. While lower energy resolution detectors such as NaI(Tl) have their place, high purity germanium (HPGe) also has a role to play. A detection with HPGe is often a characterization due to the very high energy resolution. However, HPGe crystals remain small and expensive leaving arrays of smaller crystals as an excellent solution. PNNL has developed two similar HPGe arrays for two very different applications. One array, the Multisensor Aerial Radiation Survey (MARS) detector is a fieldable array that has been tested on trucks, boats, and helicopters. The CASCADES HPGe array is an array designed to assay samples in a low background environment. The history of HPGe arrays at PNNL and the development of MARS and CASCADES will be detailed in this paper along with some of the other applications of HPGe at PNNL.

1. Introduction
Neutron and gamma-ray detection is used for non-proliferation and national security applications. Non-proliferation examples include material identification and assay in safeguards applications or air monitoring in treaty verification. In the area of national security, neutron and gamma-ray detection is used in portal monitors to screen commercial and personal vehicles, aerial and ground radiation surveys (both of background radiation or by radioisotope), stand-off detection, and gamma-ray imaging. Pacific Northwest National Laboratory (PNNL) is involved in a variety of areas where high purity germanium (HPGe) presents an excellent solution. The work performed spans the space from commercial off-the-shelf systems to custom systems. In the area of fundamental science, PNNL has contributed to the MAJORANA neutrino-less double beta decay search [1], the CoGeNT detector for dark matter search [2], and assay of materials in a shallow underground lab for use in low background detectors[3]. In addition, there have been contributions to treaty monitoring through the RL-16 certified count lab, the Radionuclide Aerosol Sampler/Analyzer (RASA) [4], on-site inspection work utilizing commercial systems for surveys and research into information barriers.

The focus of this article will be on example applications that utilize HPGe arrays that have been developed at PNNL. It will be demonstrated that the requirements for HPGe arrays in the field are very different from those in a laboratory.
2. High purity germanium

Sodium iodide with thallium doping, NaI(Tl), is commonly used in field and laboratory gamma-ray detection applications due to its relatively low cost and high detection efficiency. Of course, NaI(Tl) scintillators typically have poor energy resolution relative to the gold standard HPGe. The use of HPGe allows the precise characterization of sources of gamma-ray radiation and oftentimes, a detection with HPGe is a characterization. Figure 1 shows gamma-ray spectra collected with the MARS HPGe detector array (described below) and a 2”×4”×16” NaI(Tl) detector while observing the combination of a Pu and a 133Ba source at 5 m. The weaker gamma-ray peaks from 239Pu and 241Am are very difficult to discern in the NaI(Tl) spectra due to the presence of the much stronger peaks associated with 133Ba. However, the HPGe detector is able to easily make the distinction. Small, handheld germanium detectors are available and can be used but have efficiency limitations especially in comparison with available NaI(Tl)-based systems.

3. High purity germanium arrays

The obvious way to increase detection efficiency is to use larger detectors. HPGe crystals are limited in size by current production methods. The largest single crystal HPGe detectors are also the most expensive. A more cost effective solution is to combine multiple smaller crystals together into larger arrays thus balancing cost and size. The details of three such HPGe arrays will be discussed.

3.1. Previous Large HPGe Array

A previous large HPGe array was cooperatively developed by PNNL and Special Technologies Laboratory. It was fielded in the late 1980’s and demonstrated the use of broad-area flights over potentially radionuclide contaminated sites. The Environmental Radionuclide Sensor System (ERSS) [5] consisted of 20 60% relative efficiency HPGe crystals in two 10 crystal arrays. The system had a four hour mission time utilizing 50 liters of liquid nitrogen. The system had a 311 kg pod and three electronics racks for a total mass of 816 kg. Note that this total includes two 3He-based neutron...
detectors and the supporting electronics. However, the majority of the mass was due to the HPGe system. The effects of vibration (specifically microphonics) were mitigated by inserting insulation into the cryostats. While this reduced the vibration effects, it also created numerous thermal shorts which contributed greatly to the heat load. While the system performed adequately, any new system required several improvements.

3.2. Multi-sensor Airborne Radiation Survey

The Multi-sensor Airborne Radiation Survey (MARS) detector [6] was designed using the lessons learned from the earlier ERSS array. MARS consists of 14 35% relative efficiency p-type HPGe crystals located in a single cryostat. Two platters of seven crystals each are tightly packed and minimize the material between crystals. The system was assembled and serviced in a PNNL cleanroom. Each HPGe crystal is individually biased and has its signal sent to an XIA Pixie-4 digitizer where the pulse height and time of the event are recorded. The signal from each crystal is then converted from pulse height to energy units and events within a short coincidence window (typically 1 microsecond) can be combined to improve the full energy photopeak efficiency. The cryostat and electronics have a combined weight of 140 kg in the helicopter mount. The cryogenic performance was improved by a factor of 100 with 46 hours between liquid nitrogen fills of 5 liters. The array attained an energy resolution of 3 keV at 1333 keV and the average energy resolution of each crystal was 2.9 at the same energy.

![Figure 2. The MARS detector mounted in a side-looking configuration.](image)

The MARS detector has had an active operational history. It has been transported across the United States multiple times to field testing in Las Vegas, NV, Charleston, SC, and Newport, RI along with a nuclear astrophysics measurement in Durham, NC and outdoor testing performed at PNNL’s Building 3440 Test Track facility. In addition, it has been transported by air to an overseas maritime demonstration. Throughout all of the transportation, the system has continued to perform well even through the thermal cycle required for the air transport.
For the maritime testing on the US east coast and overseas, the system was operated on three different boats with similar system architectures. The electronics have an independent computer that is capable of acquiring the data and relaying back to land-based computers via wireless communications.

![Annotated, rendered, cutaway of the MARS detector. Inset: The back of one of the platters holding seven crystals and showing custom flexible circuit board wiring.](image)

Figure 3. Annotated, rendered, cutaway of the MARS detector. Inset: The back of one of the platters holding seven crystals and showing custom flexible circuit board wiring.

### 3.2.1. Maritime Applications of MARS

Clearly, shielding and distance work against detector efficiency and identification. The solution is to get the detector closer to the potential sources and to do more with the detected gamma rays. Testing was performed with a variety of sources and with the detector being moved on a boat at different speeds, distances and orientations to the sources which were located on a larger, target vessel. Also, when developing hardware, it is critical to also develop software that is user friendly and is tailored to the target audience.

### 3.2.2. Land-based Testing of MARS

The maritime testing of the MARS detector presented a realistic environment but it was far from reproducible on demand as would be expected in a systematic detector characterization test. This systematic testing was performed on land in a controlled, static environment at PNNL’s 3440 Test Track. The detector’s response to different sources at different distances (5 to 100 m) and with different amounts of shielding was tested over the course of two weeks.

### 3.2.3. Aerial Testing of MARS

Returning to the original aerial application, the MARS detector mount design was modified so that it could be mounted in a helicopter with the detector facing downward rather than sideways. The task was to identify the advantages offered by a large HPGe array versus NaI(Tl) for aerial measurements. Five test flights were flown on and in the vicinity of the Nevada National Security Site (NNSS) near Las Vegas, NV:

- An initial checkout flight to identify and fix any early issues
• An altitude spiral for determining the effects of air attenuation and cosmic ray background as a function of altitude over land and water
• A survey over part of the NNSS
• Flyovers of a variety of radioactive point sources multiple times at a sequence of altitudes
• Survey of a variable natural background region

Each flight served a particular testing purpose. Also, throughout all the flights, a three log RSI NaI(Tl) system was flown and acquired simultaneous data for comparison.

The clearest example of the advantage of HPGe over NaI in an aerial platform is shown in Figure 4. While the overall number of counts from the HPGe array is less than the NaI array, the specificity of the HPGe is clear. The HPGe array is able to do more with the fewer gamma-rays it detects due to the much better energy resolution. The individual peaks from $^{152}$Eu are clearly evident in the HPGe plot and much harder to observe above the natural $^{40}$K, $^{238}$U, and $^{232}$Th background in the NaI array.

The mapping ability of the MARS array was also able to be demonstrated with the natural background, gross counts map. The same features were visible in both the NaI and HPGe maps. Although an HPGe array would typically not be used in a gross counts mode, this test showed that synchronization of the GPS and MARS data streams was good.

The MARS array was also used to generate isotopic maps over the NNSS. While, an isotope with a single isolated gamma ray like $^{137}$Cs is observable with both systems, it was already shown in Figure 4 that a different isotope such as $^{152}$Eu is much more readily mapped with an HPGe system.

![Figure 4. Comparison of aerial data collected with an NaI array and the MARS HPGe array.](image)

3.3. CASCADES HPGe Array

While the MARS HPGe detector array was being developed, a similar array was also being constructed but for an entirely different application. The CASCADES detector [7] is also a 14 crystal array but the two seven crystal platter arrays are in independent cryostats and separated by enough space to allow the placement of samples between the two halves of the array. The entire array is in a lead cave surrounded by an anti-cosmic shield and is located in PNNL’s Shallow Underground Lab. The Lab has approximately 30 meters water equivalent of overburden which is sufficient to decrease the fast neutron flux by a factor of 100 and the muons by a factor of 6.

Signals from each crystal in the CASCADES array are recorded with an XIA Pixie-4 digitizer system. This laboratory-based system has achieved 1.72 keV full width half maximum (FWHM) energy resolution at 662 keV and 2.13 keV FWHM at 1332 keV. Great attention was paid to the
materials used to construct the array so as to minimize the background present in the detector itself. Recent improvements in the background performance of the array are shown in Figure 5. By lowering the intrinsic background of the detector, the minimum detectable activities of various isotopes are decreased. As an example, after the background improvements CASCADES has a minimum detectable activity of 6 mBq for Cs-137 in a 24 hour count. The system can provide useful results for very low activity samples as well as provide a high sensitivity capability to detect fission products.

![Recent Background Improvements]

**Figure 5. Example of the recent background improvements in the CASCADES array.**

Through a high efficiency, high selectivity, and low background technical approach, the CASCADES array can be used in a variety of applications:

- Assay of atmospheric filter papers
- Resolve anomalous results seen in field measurements or normal laboratory assay
- Assay low level environmental samples
- Perform physics measurements related to gamma-ray branching ratios for particular isotopes
- Potential application to International Monitoring System samples

4. Conclusions

Two similar HPGe arrays developed at PNNL for two different applications have been detailed. Use of these arrays in the field and with a variety of samples has improved the understanding of these arrays. While useful, some need for improvements has been identified. For a fieldable array, the need for longer mission times and less logistics is desired. A move to a mechanical cooler would help alleviate the problems associated with procuring and using liquid nitrogen in the field as long as the vibrational effects of the cooler can be mitigated/tolerated.

High purity germanium detectors are already in the field in the form of small, portable detectors that are used for secondary inspections, as an example. However, as long as individual HPGe crystals remain relatively small and expensive, HPGe arrays will continue to have a place both in the field and in the laboratory.
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