

**IMPROVEMENT IN GE DETECTOR COOLING**

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**ABSTRACT**

High purity germanium (HPGe) detector cooling technology used in remote radionuclide assay applications, such as the Radionuclide Aerosol Sampler/Analyzer (RASA) Mark IV, is continually evolving. Cryocooler improvements in recent years have resulted in reduced size, reduced maintenance, improved reliability, and enhanced HPGe detector performance compared to systems based on earlier generations of cooling devices. The Cryo-Cycle™ and Cryo-Pulse® 5 are two such HPGe detector cooler/cryostat systems that we are modifying and evaluating for use in the RASA. The modifications will enhance vacuum lifetime and integrity, improve performance, and resolve the need to procure and handle liquid nitrogen (LN<sub>2</sub>). Both cryocoolers offer advantages over the existing generation of HPGe detector cooler/cryostats used in the RASA and other remote-sensing applications.

## OBJECTIVES

Improvements in HPGe detector cooling technology for use in remote radionuclide assay instruments such as the RASA Mark IV will be demonstrated. Our investigations involve modifying and testing two cryostat/cooler combinations available from CANBERRA Industries, Inc., Meriden, Connecticut. A Cryo-Pulse® 5 HPGe detector system incorporating a 4-watt pulse-tube cooler manufactured by Thales Cryogenics, Eindhoven, Netherlands, is being modified to incorporate ultra-high vacuum (UHV) design and fabrication techniques. An example of the Cryo-Pulse® 5 is shown in Figure 1. The detector chamber configuration is being modified to incorporate a low radiation background remote detector chamber (RDC) in place of the Slimline (SL) chamber shown in Figure 1. The low background RDC allows back shielding to be installed directly behind the detector to minimize streaming through the radiation shield penetration.

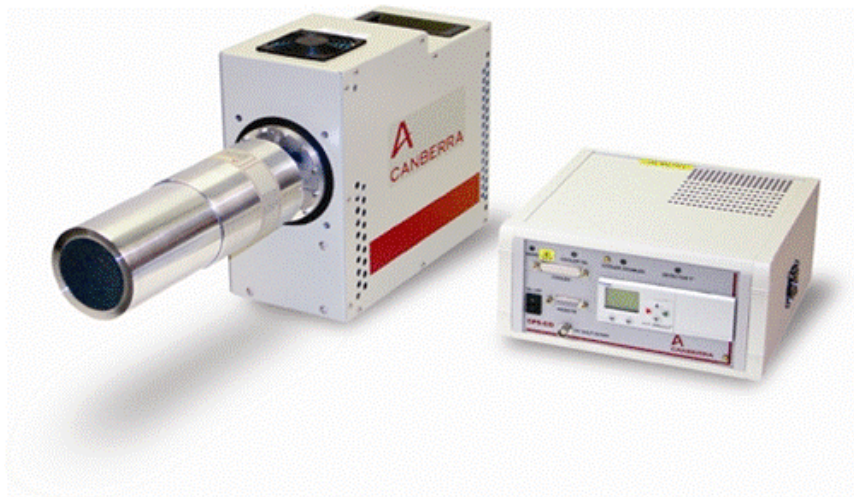


Figure 1. Cryo-Pulse® 5 model CP-5SL (image from CANBERRA website [http://www.canberra.com/pdf/Products/Detectors\\_pdf/CryoPulse5-DET-SS.pdf](http://www.canberra.com/pdf/Products/Detectors_pdf/CryoPulse5-DET-SS.pdf)).

We will also investigate a Cryo-Cycle™ HPGe detector cooler comprising a closed-system nitrogen (N<sub>2</sub>) reliquifier incorporating a 15-watt free-piston linear Stirling cooler manufactured by SunPower, Inc., Athens, Ohio. This hybrid system has the same footprint as a standard 30-liter LN<sub>2</sub> Dewar and receives dipstick HPGe detector cryostats like those commonly used in other LN<sub>2</sub>-based systems. The dipstick cryostat for the RASA system will incorporate all-metal seals and a low-background RDC. The Cryo-Cycle™, shown in Figure 2, continuously condenses the LN<sub>2</sub> boil-off inside a closed Dewar to maintain a 22-liter reservoir of LN<sub>2</sub>. Once charged, the system does not require additional N<sub>2</sub> except in the event of a long-duration power outage or to replace gas loss due to seal leakage. Make-up can be added by topping off with LN<sub>2</sub> or by adding N<sub>2</sub> gas. An N<sub>2</sub> gas generator will be integrated with the standard Cryo-Cycle™ to allow unattended operation in the RASA. In controlled environments such as the instrument bay inside RASA systems, the only maintenance required is semi-annual filter cleaning for both the Cryo-Cycle™ and Cryo-Pulse® 5-based systems. N<sub>2</sub> gas generator maintenance interval and lifetime have not yet been determined. Expected lifetimes for both cryocooler units are in excess of 50,000 hours. Both systems will be tested to validate spectroscopic performance, thermal performance, and vacuum performance for the RASA application. This research will demonstrate improved operational performance (better resolution) and improved operational reliability (less downtime) when compared to previous generations of mechanical coolers used in the RASA system.



**Figure 2. Cryo-Cycle™ shown with model 7500SL cryostat (from CANBERRA website [http://www.canberra.com/pdf/Products/Detectors\\_pdf/CryoCycleCryostat-DET-SS.pdf](http://www.canberra.com/pdf/Products/Detectors_pdf/CryoCycleCryostat-DET-SS.pdf))**

The contract period for this research began in the second week of June 2008. At the time of this writing, two weeks later, no significant research has been accomplished. We will therefore take this opportunity to discuss the nature of the challenges and the methods we will use to address them.

The RASA was developed at the Pacific Northwest National Laboratory (PNNL) in the 1990s to meet Comprehensive Nuclear-Test-Ban Treaty (CTBT) requirements for aerosol radionuclide measurements, as described by Bower et al. (1997) and Miley et al. (1998). Particulate air monitors are installed at each of the 80 radionuclide monitoring stations established worldwide by the International Monitoring System (IMS). The RASA draws approximately 24,000 m<sup>3</sup>/day of air through six parallel filter media strips; every 24 hours the strips are combined and laminated between two continuous Mylar tapes to form a continuous strip of individual pouches, each representing a 24-hour sample. Each pouch is marked with a barcode containing identifying information. Perforations between pouches allow collection as a continuous reel or separation wherever desired. After a sample is collected and laminated, the pouch sits for 24 hours to allow decay of normal atmospheric short-lived radon daughters. It is then transported around an HPGe detector inside a shielded enclosure for a 24-hour measurement of gamma ray emissions. Software analysis of the data is performed, and the raw data and results are communicated to the International Data Centre (IDC) in Vienna. As the continuous tape of pouches exits the machine, it can be automatically reeled and stored, or specific pouches can be manually removed for transport to one of 16 IMS radionuclide laboratories for further analysis. The system operates unattended for 6-month intervals before routine preventative maintenance and replenishment of materials are required. Figures 3 and 4 show the RASA Mark IV pictorially and schematically.



## Department of Energy

A Radionuclide Aerosol Sampler/Analyzer

### RASA Mark 4

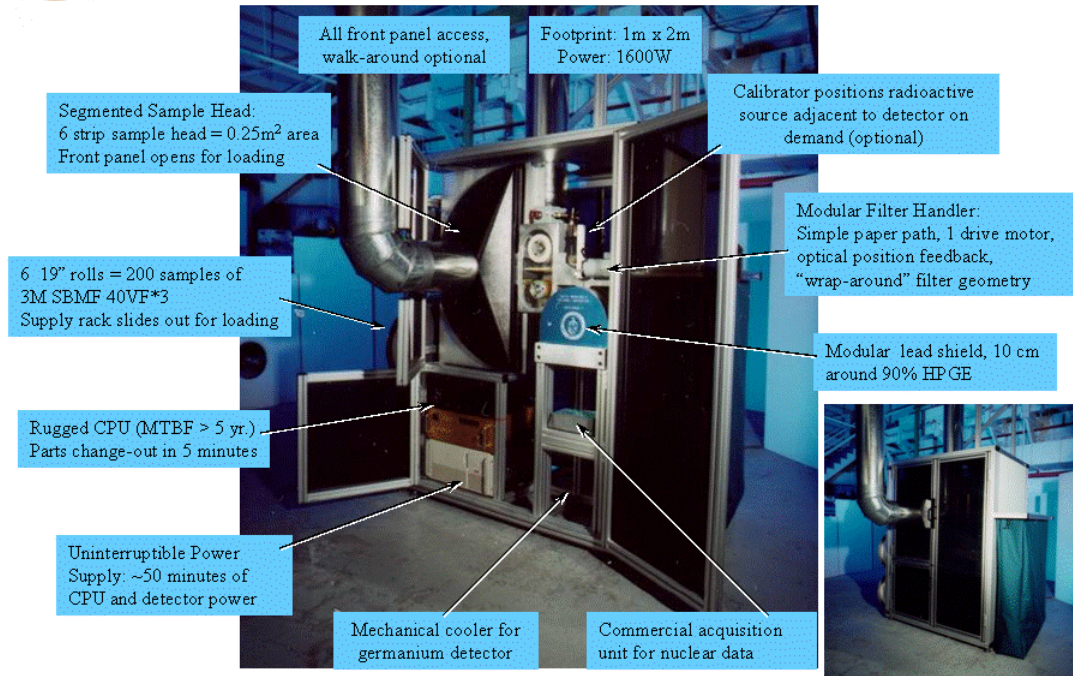


Figure 3. RASA Mark IV.



## Department of Energy

Radionuclide Aerosol Sampler/Analyzer

### System Overview

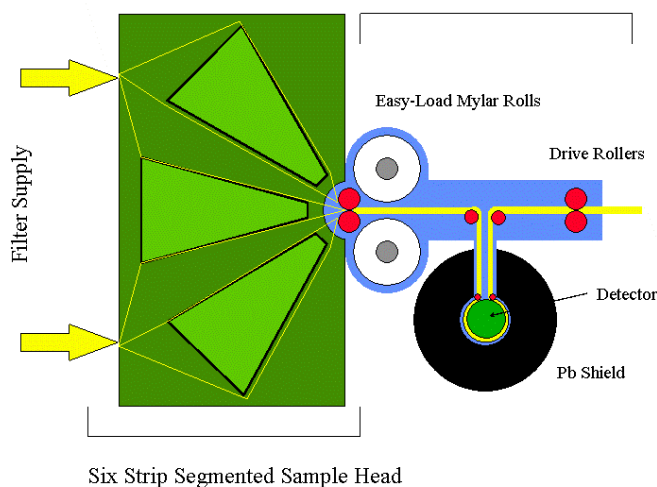


Figure 4. Schematic diagram of the filter strip transport system in a RASA Mark IV.



Radionuclide concentrations in air are quantified in microbecquerels per cubic meter ( $\mu\text{Bq}/\text{m}^3$ ). The sensitivity of the RASA is defined as the minimum detectable concentration (MDC) of relevant fission-product radioisotopes it can measure. The Provisional Technical Secretariat (PTS) requires that the system be capable of measuring an MDC of  $10 \mu\text{Bq}/\text{m}^3$  for  $^{140}\text{Ba}$  in the absence of radon and its daughters and an MDC of less than  $30 \mu\text{Bq}/\text{m}^3$  in operation (Züringer et al., 2007). When measuring low radioisotopic concentrations, it is axiomatic that larger HPGe detectors (i.e., higher relative efficiency for  $^{60}\text{Co}$ , 1.33 MeV), lower background radiation, and better energy resolution lead to improved MDC. Given the existing configuration of the RASA, this translates realistically to detectors in the 100% efficiency range; hardware background radiation typical of industry-standard, ultra-low background materials; and resolution of less than 2.0 KeV for 1.33 MeV gammas. This performance level has been reached with the detectors previously provided to the RASA development effort by the vendors. Long-term mechanical cooler and cryostat performance, however, has been problematic.

Mechanically cooled HPGe detectors for the early generations of RASA application were procured from vendors in the mid- to late-1990s. These detector systems use mixed-refrigerant single-stage or auto-cascade closed-cycle Joule-Thomson (JT) coolers. To remove heat from the detector, the coolers rely on the expansion of gas mixtures or gas and fluid mixtures under high pressure through an orifice or series of orifices. Input power to the compressor is in the range of 0.5 to 1.0 kilowatts in order to provide a minimum cooling power of several watts at the roughly 100°K temperature required for HPGe detector operation. Vibration from the large compressors and from the expanding gas often degrades detector performance. The expansion necessary to accomplish the low temperatures necessitates small JT expansion orifices that are prone to clogging. Water and oil contamination in the JT working fluid can freeze and cause clogging. The working gases are mixtures of flammable gases that can create hazardous situations when leakage occurs or maintenance is required to replace lost gas. Cooling power in JT coolers for HPGe detectors can be marginal. With time, the power is insufficient to overcome the cumulative effects of conductive and convective heat loads caused by vacuum degradation inside the detector chamber. Vacuum degradation is caused by gas permeation through elastomer vacuum seals and from hardware outgassing. For this reason, vacuum service is often required annually to re-evacuate detector cryostats. Molecular sieves used to establish and maintain high vacuum in detector cryostats are prone to outgassing during brief power outages. This then necessitates a 24–48 hr full thermal cycle to room temperature and back to operating temperature in order to clear detector contamination caused by the outgassing. Low cooling power also imposes constraints on cryostat configurations and detector size. Improvements in JT-cooled detector technology have been made, but all shortcomings for the RASA application have not been resolved.

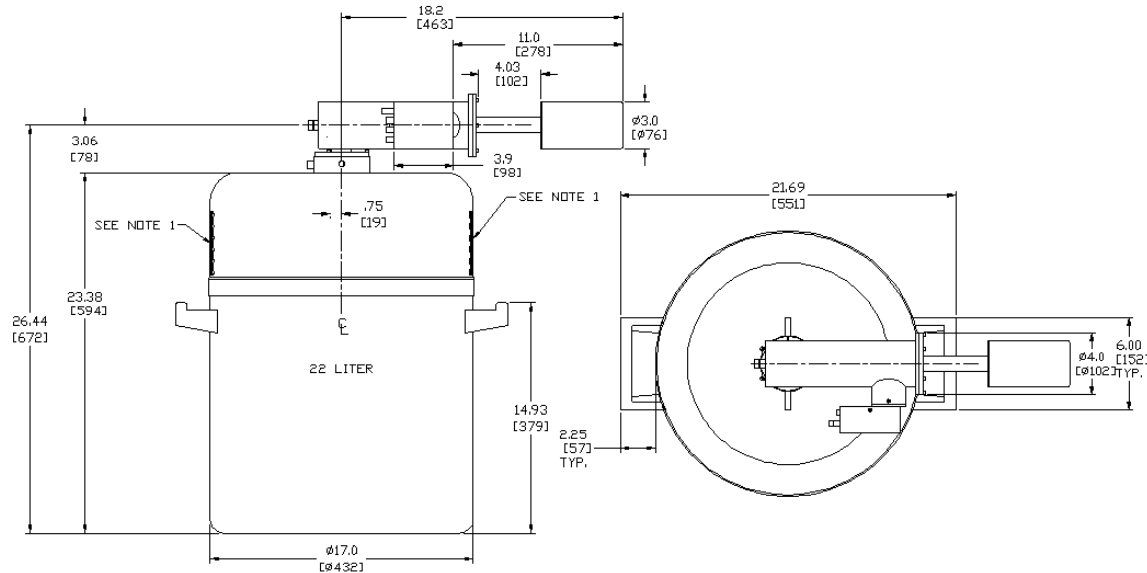
Great strides have been made in alternative cooling technologies in the period since the first RASA systems were built, specifically with linear Stirling and linear Stirling-type pulse-tube coolers. Both types of cooler are inherently more efficient at high and low temperatures than the JT coolers used for HPGe detectors. Until recently, they had not been used to any large extent as HPGe detector coolers because of high cost, short lifetime, and high vibration. Events conspired in the 1990s to give rise to development efforts that removed or substantially lowered these hurdles. At the time, it was thought that many thousands of coolers would be needed to provide cooling for the electronics in cell phone and other communication network stations. This need has not yet fully materialized, but the prospect has led to substantial improvements in Stirling and pulse-tube cooler performance and price. Flexure bearings, gas bearings, magnetic and pneumatic stops, improved motor design, and improved manufacturing techniques have brought these coolers to the point that their costs are below \$25,000, their lifetimes exceed 50,000 hours, and vibrations have been reduced, or at least offset, by the high cooling power available. Stirling and pulse-tube-cooled HPGe detectors are now available from major detector manufacturers in many different configurations. We believe that cooling systems based on the Cryo-Pulse<sup>®</sup> 5 direct-coupled, pulse-tube cooler and the Cryo-Cycle<sup>™</sup> hybrid cooler now represent the best choices for the RASA application.

The Cryo-Cycle<sup>™</sup> is referred to as a hybrid cooler for HPGe detectors because it uses an electrically powered mechanical Stirling cooler, and in this case an  $\text{N}_2$  gas generator, to generate and indefinitely maintain a reservoir of  $\text{LN}_2$  for cooling an HPGe detector. A 15-watt CryoTel GT Stirling cooler is attached to a closed  $\text{LN}_2$  Dewar to reliquify boil-off gas and return it to the 22-liter  $\text{LN}_2$  reservoir. The SunPower cooler incorporates a floating displacer with pneumatic axial centering system, gas bearings, a free-floating magnetic compressor piston, and a linear AC motor to reduce power consumption and eliminate the major causes of wear failures in Stirling coolers. Vibration is reduced by use of a tuned dynamic vibration absorber. Details of the design and references can be found at the SunPower website [www.SunPower.com](http://www.SunPower.com). To minimize the effect of vibration on detector performance, the

Stirling cooler is mounted on vibration isolators and coupled to the Dewar through a gas-tight, vibration-isolating coupling. A dipstick cryostat is inserted into the Dewar through a vibration-isolating gas-tight collar. The closed Dewar operates under 1–2 psig positive pressure relative to the surroundings. The footprint and function of the hybrid Dewar are similar to the standard 30-liter Dewar commonly used for HPGe dipstick cryostats.

Metal-sealed dipstick cryostats have a 40-year history of providing extremely long life and dependable germanium-detector performance. In recent years, some users have found that the administrative difficulties and logistical inconvenience of dealing with LN<sub>2</sub> outweigh the benefits of the metal-seal dipstick design. For remote unattended applications such as RASA, it can be seen that this is a legitimate consideration. However, the hybrid cooler removes the negative aspects of acquiring and handling LN<sub>2</sub>, because the LN<sub>2</sub> is generated and confined inside a closed system. The operator and maintenance personnel never come in contact with it, and transport of LN<sub>2</sub> is not required. A major benefit of LN<sub>2</sub> is that it provides a 7-day reserve of cooling power. If any of the electrical cooling components fail, or if a power failure of long duration takes place, the 22-liter reservoir of LN<sub>2</sub> provides up to 7 days of cooling to the detector with no risk of a partial thermal cycle or warm-up. The detector is immediately available when power is restored. The modularity of the system ensures that, as long as the detector and Dewar are intact, the system will provide a 7-day buffer for replacement of a faulty component. In most cases, downtime is minimized, because the detector does not need to be warmed for repairs. Moreover, the interchangeability of dipsticks and Cryo-Cycle™ coolers allows independent replacement of either of the two most expensive components in the system. The Cryo-Cycle™, with an N<sub>2</sub> gas generator attached and a metal seal dipstick cryostat installed, is a closed system that is constantly maintained by electricity. The simplicity and proven track record of LN<sub>2</sub>-cooled, long-life, metal-seal dipstick cryostats ensure the absolute highest level of dependability and performance.

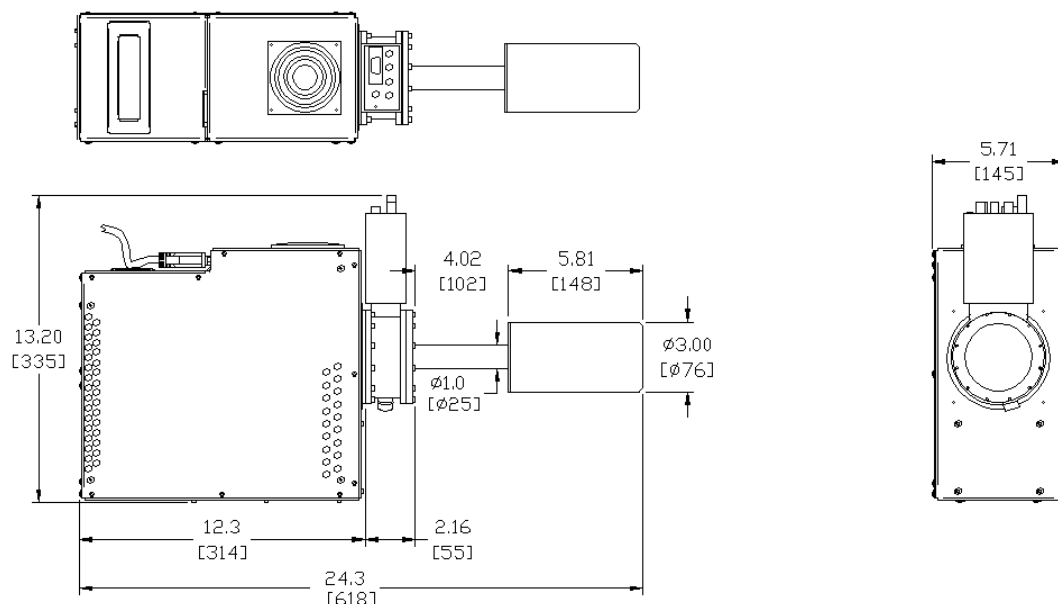
The Cryo-Cycle™ is an off-the-shelf product for use with dipstick cryostats. The difficulties of integrating the Stirling cooler with the detector have been solved. There are no vibration, thermal, or vacuum issues to be addressed. For the RASA application, we will make improvements to the metal-seal dipstick design by developing a small-cross-section metal-seal end cap to be used on the RDC. As mentioned earlier, an RDC is preferred for the RASA because the detector chamber must reach inside a shielded enclosure. The RDC allows the most convenient method for back-shielding the detector. A long end cap that reaches through the shield penetration must include the back-shielding inside the cryostat vacuum space behind the detector. With an RDC, the back-shielding is installed outside the vacuum space behind the RDC. A diagram of a Cryo-Cycle™ RDC is shown in Figure 5. This can be compared to the photos of Slimline end caps shown in Figures 1 and 2. The RDC will contain standard-product ultra-low-background (ULB) hardware to reduce the amount of background radiation inside the shield. We will also use welded UHV electrical feedthroughs and a metal pinch seal on the vacuum pump-out port. Such a cryostat is expected to have a vacuum life in excess of 10 years. Finally, we will integrate an N<sub>2</sub>-gas generator to completely remove the necessity for transporting and handling LN<sub>2</sub>. An off-the-shelf high-purity N<sub>2</sub>-gas generator will provide source gas for the initial charge of LN<sub>2</sub> and subsequent make-up N<sub>2</sub> gas. Excess N<sub>2</sub> gas can be used to maintain a purge in the shield cavity to reduce radon background. We believe that generation rate of several hundred cubic centimeters/minute of 99.995% pure N<sub>2</sub> gas should be sufficient, but this point requires further investigation, as does the maintenance interval for such generators.



**Figure 5. Cryo-Cycle™ model CCHD RDC-4 (image courtesy of CANBERRA).**

Another approach we are pursuing for the RASA application is modification of a Cryo-Pulse® 5 to improve vacuum integrity and lifetime. The Cryo-Pulse® 5, shown in Figure 1, is a compact unit comprising a Thales 4-watt pulse-tube cooler directly coupled to a vacuum cryostat containing an HPGe detector. A separate controller is connected to the cooler by an electrical cable. The cooler/cryostat package with RDC is shown in Figure 6. The Thales cooler incorporates a moving-magnet linear helium compressor and a split pulse-tube cold head. The compressor makes use of dual, opposed permanent magnet pistons that are fully supported by flexures to prevent piston-to-cylinder contact and reduce wear. Stator coils are placed outside the helium pressure vessel, thereby removing a potential source of gas contamination. In addition, the external stator design eliminates the need for a glass or ceramic feedthrough in the pressure vessel. The pulse-tube cold head contains no moving parts, so wear is eliminated and vibration is reduced. These design choices remove the main causes of failure, wear and contamination, and improve reliability. Vibration is reduced by virtue of the canceling effect of the opposed pistons and by the lack of moving parts inside the cold head. The manufacturer states that the cooler has an expected life in excess of 50,000 hours. Details of the Thales cooler can be found at [www.thales-cryogenics.com](http://www.thales-cryogenics.com).

The Cryo-Pulse® 5 is available with detector sizes in excess of 100% relative efficiency and energy resolution of less than 2.0 keV for 1.33 MeV gammas. The ULB RDC is a standard option. End caps and RDC neck lengths are available in different sizes, with a 4-in.-diameter end cap accommodating detectors of up to 130% relative efficiency. An diagram of a Cryo-Pulse® 5 cryostat configuration usable in the RASA is shown in Figure 6.



**Figure 6. Cryo-Pulse® 5 Model CP-5F with RDC (image courtesy of CANBERRA).**

The standard-product Cryo-Pulse® 5 so far discussed uses zeolite molecular sieves to maintain the detector vacuum when cold. As with all sieve-pumped cryostats, the unit is susceptible to partial thermal cycling in the event of a brief power outage. Because the sieves are closely coupled to the cooler cold tip, they are the first things to warm in the event of a power outage. As the sieves warm, they release large amounts of the gases they have captured. The gases then condense on the still-cold detector. If power is restored before the detector reaches room temperature, the detector is said to have partially thermal-cycled, and some gases remain adsorbed on the detector surface, causing leakage current and poor performance. In the event of a partial thermal cycle, the detector may be offline for 1–2 days during recovery.

The standard Cryo-Pulse® 5 has at least one elastomer seal on the vacuum pump-out port sealing plug. RDC-equipped cryostats also have an elastomer end-cap seal. The cryostat electrical feedthroughs are made of glass and sealed with epoxy. Elastomer, glass, and epoxy seals are all permeable to atmospheric gases to some small degree, and they are particularly permeable to helium. Because the partial pressure of helium in the atmosphere is low and the pulse-tube cooling power during cooldown is high, the lifetime of a Cryo-Pulse® 5 exceeds 5 years with normal use. But the demands of the RASA are such that a higher standard of reliability is required. We are currently designing a low-cross-section metal seal for the RDC end cap. A low cross-section is required to maximize the HPGe detector volume (i.e., the relative efficiency) that can be contained in a given end-cap outside diameter (OD). End-cap OD is in turn constrained by the bore diameter of the shield entry port. In the case of the RASA, this is an approximately 4-in. clearance. In addition to the end cap seal, we are replacing the electrical feedthroughs with welded UHV feedthroughs, such as Ceramaseal® UHV feedthroughs by CeramTec North America, Inc., Laurens, South Carolina. A metal pinch tube that forms a cold-weld metal bond when sealed will replace the pump-out port plug. Elimination of highly permeable seals will greatly enhance the vacuum integrity of the Cryo-Pulse® 5 but will not solve the problem of partial thermal cycling in the event of brief power loss. This risk can be mitigated by using an uninterruptible power supply (UPS) and/or back-up generator, but to completely eliminate the risk, we choose to use UHV cryostat design techniques.

The issues associated with gas loads in mechanically cooled HPGe detector cryostats have been elucidated in past proceedings of this conference by Hull et al. (2006, 2007). The intrinsic gas load inside a sealed vacuum cryostat results from leakage, diffusion, vaporization, and surface desorption. All these sources can be minimized by control of design, fabrication, cleaning, and assembly techniques. The most stringent application of these techniques is referred to as UHV technology. UHV denotes the vacuum pressure range below  $10^{-9}$  torr. In order to achieve UHV,



the gas load must be severely reduced. Ekin (2006), O'Hanlon (2003), and Redhead et al. (1967) are only three of myriad references on the topic of applied UHV techniques. Since gas pressure inside the sealed vacuum cryostat is a function of the gas load and rate of gas removal by pumping, the battle is only half fought by controlling gas load. One or more pumping methods must be incorporated into the design in order to permanently remove evolved gases and ensure a long-lived working vacuum.

To minimize conductive and convective heat loads and reduce contamination of the HPGe detector junction surface, the working vacuum inside the detector cryostat must be maintained in the low  $10^{-5}$  to  $10^{-6}$  torr pressure range. We believe that chemical getter pumps and ion pumps are practical for compact detector cryostats because they permanently remove gas molecules from the vacuum space. Other types of pumps, such as cryopumps, turbomolecular pumps, and diffusion pumps, are impractical, primarily because of size or implementation constraints. Cryosorption pumps are impractical because they do not permanently remove gas molecules (with the possible exception of water molecules) from the vacuum space, thereby leaving the detector susceptible to partial thermal cycles. Chemical getter pumps, such as titanium sublimation pumps (TSPs) or non-evaporable getter (NEG) pumps, permanently remove reactive gases when they form chemical compounds on the getter surface. Noble gases are not affected. Alloys of vanadium and/or zirconium are used as the reactive media in NEG pumps manufactured by the SAES Getters Group (<http://www.saesgetters.com/>). In addition to irreversibly pumping many reactive gases, NEG pumps can possess high capacity for reversibly absorbing hydrogen. NEG pumps can be regenerated *in situ* by heating to several hundred degrees Celsius and diffusing the chemical reactants into the bulk of the media, thereby forming a fresh reactive surface layer. A TSP works by heating a filament of titanium that sublimates onto adjacent surfaces, forming a chemically reactive film. The lifetime of TSP filaments is short. Ion pumps remove gas molecules through several different mechanisms after the gases are ionized and accelerated by high voltage applied to the pump. Ionized gas molecules are removed from the vacuum by implantation into the electrodes, burial by metal sputtered from the electrodes, or by chemical reaction with metal sputtered from the electrodes.

In a UHV chamber, with NEG and/or ion pumps installed, the residual gas species and their partial pressures can be identified using a residual gas analyzer (RGA). Pressure can be measured with a modified Bayard Alpert ionization gauge tube or inverted magnetron cold-cathode gauge tube down to the  $10^{-11}$  torr range. Even at these low pressures, knowledge of the exact nature of the residual gases inside the cryostat is important for predicting the lifetime of the pumps and the rate of long-term vacuum degradation. These residual gas species and their partial pressures can be identified using an RGA. We feel that, although we will be applying UHV techniques that have been successfully used on other products, it will still be important to determine the residual gas constituents in order to predict the optimum pumping methods and UHV materials to use. Investigations will be carried out using in-house RGA and UHV facilities to shed light on the nature of the residual gas environment in our modified Cryo-Pulse® 5. It may also be possible to carry out accelerated aging tests by observing the cryostat pressure over a period of weeks with the cryostat at elevated temperature.

### **RESEARCH ACCOMPLISHED**

The project work has just begun. We are currently in the design phase for the RDC end-cap low-cross-section metal seal. Information on small laboratory high-purity nitrogen generators is being gathered. Conceptual design is being carried out for the Cryo-Pulse® 5 UHV cryostat modifications.

### **CONCLUSIONS AND RECOMMENDATIONS**

Products based on Stirling hybrid cooler and pulse-tube cooler technologies show promise as improved HPGe detector coolers for the RASA system. Modification of the existing Cryo-Cycle™ and Cryo-Pulse® 5 products provides a means of avoiding the time and cost of full-scale efforts to develop new cooler technologies for this application. Integration of an N<sub>2</sub>-gas generator with the Cryo-Cycle™ and incorporation of UHV seals and feedthroughs on the dipstick cryostat offer one solution. Enhancement of the Cryo-Pulse® 5 by application of similar and more rigorous UHV design principles to the cryostat hardware and vacuum maintenance methodology offers another solution. RGA analysis of the residual gases inside the cryostats can be beneficial with both products, especially the latter.

Our recommendation and intention are to pursue both solutions by building and testing modified versions of the two standard products. RGA analysis should be used to optimize UHV design.

## **ACKNOWLEDGEMENTS**

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