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ANTICOSMIC-SHIELDED ULTRALOW-BACKGROUND GERMANIUM DETECTOR SYSTEMS FOR ANALYSIS OF BULK ENVIRONMENTAL SAMPLES

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A low-level gamma-ray counting system has been developed which reduces system background, relative to other typical low-background systems, by a factor of ten in the energy region below one MeV, and by as much as a factor of forty at higher energies. This germanium-diode gamma-ray spectrometer was constructed for a modest investment above that required for a conventional germanium detector. The techniques involved use: (1) materials of known radiopurity to surround the diode, (2) an active external anticosmic shield to reduce the background continuum due to interactions of cosmic particles with the detector and passive lead shielding, and (3) nitrogen exhausted from the cryogenic dewar to minimize the introduction of ubiquitous radon decay nuclei into the sample counting chamber. A novel method for handling samples prior to counting is presented. Also, some of the difficulties encountered in calibrating a system intended for bulk samples are discussed.

Introduction

Several applications for routine low-level radiation counting using a germanium diode have evolved during the last two decades. The foremost function has been environmental monitoring, particularly in the neighborhood of nuclear facilities such as power plants or sundry national laboratories. Low-level counting has also been used routinely to establish an environmental baseline prior to construction of power plants, both nuclear and coal-fired. Additionally, low-level counting has been useful in the selection of radiopure materials for use in the construction of improved low-level counters, in radiochemical neutron-activation analysis, and in the measurement of low concentrations for several daughters of primordial radionuclides in site characterization studies for proposed nuclear waste repositories. The recent disaster with the Chernobyl reactor has also precipitated the need to monitor

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agricultural commodities derived from certain European sources. All of these applications can benefit from the improved radionuclide-detection sensitivities which are realized by increasing the detector efficiency, by reducing the background inherent in the detector system, and by counting larger aliquots.

Methodology

In general, three factors need to be controlled when dealing with low-level counting systems:[1-2] 1) selecting a diode with optimal efficiency and resolution, 2) reducing the background to as low a level as practical, and 3) increasing the amount of signal seen by the detector. The optimal size and resolution of the detector is generally fixed at what the manufacturer can provide when meeting required specifications. However, the other two factors, reducing the system background and increasing the sample signal, can readily be dealt with by knowledgeable construction, handling, and preparation on the part of the researcher.

One of the most critical factors involved in reducing the system background is the judicious use of construction materials with high radiopurity. Work at the Pacific Northwest Laboratory (PNL) facility[3] has shown that many commonly used materials are contaminated with primordial radionuclides and their progeny, and that stainless steel and iron generally contain ^{60}Co. Much of the lead used for bulk shielding is contaminated with ^{210}Pb and its daughters. The bremsstrahlung radiation from ^{210}Bi can be a significant portion of the background spectrum below one MeV. The current materials of choice for providing a bulk shield surrounding the diode are centuries-old lead, electrolytic copper, or mercury. Work performed at PNL has indicated that some pre-1945 battleship steel is no longer state-of-the-art material as it contains significant quantities of thorium, uranium, and their decay products. Use of electrolytic copper is avoided, when possible, because of the presence of detectable quantities of radioisotopes such as ^{56}Co, ^{57}Co, ^{58}Co, and ^{60}Co which result from cosmic-ray bombardment. The use of mercury may be undesirable due to its inherent toxicity and volatility, and the difficulties presented in containing the liquid metal with radiopure materials.

Another major contributor to system background is cosmic radiation. The contribution from muonic interactions within the germanium diode and the contribution from prompt gamma rays and fast neutrons produced by cosmic-ray interactions in the mass shielding can be reduced if counting is suspended for a brief period of time following detection of an event in an external scintillator. This technique is referred to as anticosmic (or anticoincidence or veto) shielding. Anticosmic shielding is most effective at reducing the background due to high-energy charged particles, such as muons. However, radiation originating from the decay of radionuclides within the detector and shield (whether present as contaminants or as activation products of neutron absorption) cannot be cancelled using this technique; such radionuclides decay with a characteristic half life independent of the interactions that produced them.

A third technique to reduce the system background is to minimize the presence of airborne contaminants, particularly ^{220}Rn, ^{222}Rn and their daughters.[4] Other workers have addressed this problem by filling all crevices of the bulk shielding with a polymer, such as Lexan, which sets up to form an airtight mass. A more versatile technique for handling the radon contamination problem is to flush the air-tight compartment surrounding the diode with boiled-off nitrogen from the cryogenic dewar. This technique has the added advantage that gaseous airborne contaminants which are introduced by changing a sample are flushed from the vicinity of the diode. Protecting the samples from airborne contamination prior to counting is also important since non-gaseous radioactive daughters of the radon gases may attach themselves to the samples or containers.

The amount of signal seen by the detector can be increased by reducing the sample-to-detector distance and/or by increasing the sample mass. It is critical that geometries be accurately calibrated and reproducible. Appropriate steps must be taken to assure sample homogeneity, and corresponding uncertainties should be determined. At PNL, typical samples consisting of large-volume liquids, some vegetation, soils, adsorption media, etc., are contained within Marinelli beakers constructed from low-atomic-number (low-Z) materials. (A Marinelli beaker is a right-cylindrical can with a well formed in one face to accommodate placing the sample directly over the diode.) Inhomogeneity of the sample, as well as packing density irregularities, can lead to erroneous results when counting non-liquid samples. Pressing light-weight vegetation samples into the Marinelli shape or into a hollow right-circular cylinder will reduce the average sample-to-detector distance by decreasing the volume of voids.

Experimental

The work described in this study was performed using a high-purity germanium (HPGe) diode with an efficiency of 31.5 %, and a resolution of 1.98 keV for the 1332 keV ^{60}Co gamma ray. The system design is illustrated in Figure 1. The cold finger, vacuum jacket, end cap, and diode support cup were fabricated from electrolytic copper of known radiopurity. Aluminum alloys are the materials of choice due to their strength, low density, low-Z, and low cross section to fast neutrons. However, a source has yet to be identified which can supply a substantial quantity with the requisite radiopurity, although some manufacturers now rumor that quantities will be available in the very

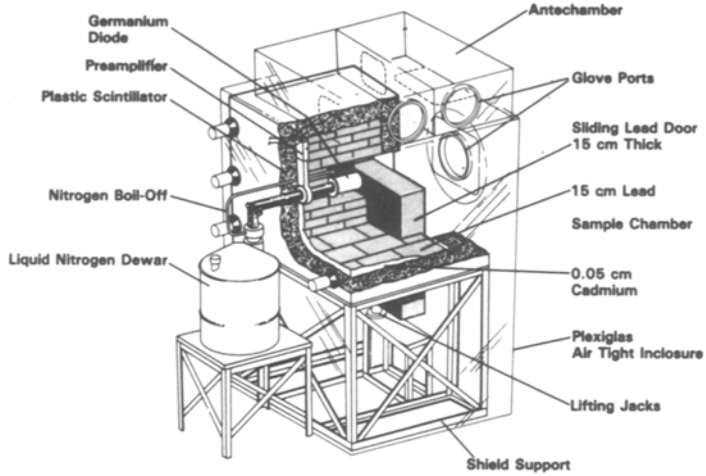

Fig. 1. Shielding configuration of the PNL low-level germanium detector system

near future. The diode manufacturer was provided with solder made from radiopure tin and centuries-old lead for use on electrical connections made near the diode.[5] Flanges were connected to the various sections of the cryostat barrel by electrodeposition of copper (electron beam welding would have been an acceptable alternative) rather than by conventional welding which could have introduced thorium and its daughters. The electronic components, normally located within the cryostat, close to the germanium detector, were removed to the preamplifier (with a sacrifice in resolution of about 0.2 keV F.W.H.M. at 1332 keV.) The preamplifier was located such that a minimum of 15 cm of lead shielded the germanium diode to minimize the effects of radiation from ^{40}K and the radioactive daughters of thorium and uranium which are intrinsic to some of the electronic components (resistors, circuit boards, etc.). To further reduce the effects of the seemingly omnipresent ^{40}K, the detector housing and the 15 cm of lead bricks surrounding the diode were washed, etched with high-purity nitric acid, and rinsed first with doubly distilled water and then with absolute ethanol. The cleansed bricks and detector housing, once sanitized, were handled exclusively with gloves known to be radiopure.

The background due to cosmic-ray interactions was minimized by the use of an anticosmic plastic scintillator located outside the lead shield, as illustrated in Figure 1. The rationale is that a cosmic ray passing through the scintillator and detector produces events in both that are in coincidence (occurring within the resolving time of the electronic system). Additionally, high-energy cosmic rays will produce high-energy gamma rays, high-energy neutrons, etc. Occasionally, the neutrons interact with shielding and construction materials to produce other penetrating radiation which may be counted by the detector. Much of such secondary radiation occurs during a small window of time following the cosmic-ray event. A time window 100-microseconds wide is sufficient to include a large fraction of such secondary events and cause only a small loss in counting time, as the muon flux on the entire scintillator face is roughly 150 per second.

Events occurring within the anticosmic shield generate flashes of light which are converted to electrical signals by photomultiplier tubes located along the edges of the scintillator material. The signals produced in the plastic scintillator are routed to a dual discriminator. The discriminator levels are set such that a signal corresponding to an energy deposition in the plastic scintillator greater than 2.6 MeV results in a logic pulse at an upper-level output, while other pulses which are above a discriminator setting result in a logic pulse at the lower-level output. The logic pulses from the two outputs of the dual discriminator are routed to logic circuitry which generates a 100-microsecond blocking signal if the pulse was from the upper-level output and a 5-microsecond blocking signal if the pulse was from the lower-level output. These blocking signals are used to turn off the ADC and live-time clock for the duration of each signal.

Cosmic rays passing through the scintillator produce pulses representative of energy in excess of 3 MeV, while most gamma rays present in the environment deposit less than 3 MeV. A threshold just above 2.6 MeV was selected for the upper-level discriminator setting in order to avoid high count-rates from ^{208}Tl, a universally occurring decay product of the primordial-thorium chain. Most of the signals which clear the lower-level discriminator setting arise from low-energy gamma rays in the surroundings and result in excessive dead time if the 100-microsecond blocking signal is applied uniformly. However, using a 5-microsecond blocking signal results in a further reduction in the background, with only about 1% additional dead time. This reduction is probably due to the blocking of secondary radiation produced by cosmic-ray interactions in the laboratory surroundings. Five microseconds was chosen because of the simplicity of the requisite electronics. Blocking signals of much shorter duration require electronics of greater complexity and are necessary only if an excessive count rate is produced from radiation in the vicinity of the scintillator. The total dead-time resulting from all these interruptions was about 2% for the PNL system.

The anticosmic shield used in this study is composed of 10-cm thick plastic scintillator with eight photomultiplier tubes coupled at the edges. The scintillator is located external to the bulk shielding, rather than close to the diode as in other low-level counting systems.[6] The placement of the anticosmic shield external to the lead shielding reduces the background which would otherwise come from the ^{40}K and thorium/uranium decay products present in the glass, photocathodes, and other components of the photomultiplier tubes.[7] Also, many cosmic-ray interactions which occur in the mass shield will not be detected by a scintillator which is located inside this mass.

A lining of cadmium metal was placed between the anticosmic shield and the bulk shielding to facilitate maintaining an airtight counting chamber, as illustrated in Figure 1. The cadmium lining also absorbs ambient thermal neutrons arising from cosmic-ray-induced spallation and from naturally occurring spontaneous fission as well as high-energy neutrons which become moderated by the low-Z materials in the plastic scintillator. Activation of the germanium diode and other materials inside the sample chamber by absorption of thermal neutrons is therefore diminished. However, inelastic high-energy neutron scattering and spallation activate the germanium[6] so that beta and gamma rays emitted from within the diode itself are a significant part of the background remaining in the PNL system. Other low-level counting systems at PNL have used liquid-scintillator-based anticosmic shields with equal success. Liquid scintillator is less expensive, but requires that a low-refractive-index container be fabricated with sufficient strength to support the weight of the massive shield. (Stainless steel and other metals are poor container materials due to their high refractive index but may perhaps be lined with appropriate low-refractive-index materials.)

To reduce airborne contaminants, the entire sampling system (diode, lead shield, and anticosmic shield) was enclosed within an airtight container fabricated from Plexiglas sheets. The continuous positive pressure exerted by the nitrogen exhausted from the cryogenic dewar helps to maintain an atmosphere free of airborne contaminants. The dry nitrogen gas passes through a small diameter copper tube to the lead shield and enters the sample chamber through a small passage (0.5-cm diameter) in the lead shield. Detector exposure to unshielded line-of-sight gamma rays which might enter through this passage is avoided by drilling two holes whose intersection forms a right angle at a point located 5 cm above the chamber near a back corner of the chamber. Most of the nitrogen exits the sample chamber through the small openings around the lead door and then through an antechamber located on top of the system. The door is fabricated and positioned such that no line-of-sight gamma rays enter the sample chamber when the door is in the closed position. The antechamber is composed of two compartments which facilitate sample storage prior to counting on the diode. Samples are introduced to the system through the nitrogen exhaust port. A series of samples is therefore flushed with the exhausting nitrogen for an extended period of time prior to counting, which results in further reduction in the quantity of airborne contamination entering the sample chamber. Before installing the nitrogen purge, the diode and inner surfaces of the shield were protected from airborne contaminants by radiopure plastic coverings.

The level of detection for the system is further reduced, whenever practical, by using samples in bulk. The Marinelli shape allows the greatest quantity of sample to be closely presented to the active volume of the diode and works satisfactorily for homogeneous liquid samples that do not precipitate or migrate during the counting interval. The outer dimensions of the one-liter Marinelli-shaped containers used in this study were 13.3-cm diameter by 9.53-cm long with an 8.1-cm diameter by 6.35-cm long right-circular cylindrical void in one end to allow for placement over the detector. A significant improvement in sample count rate was achieved for some bulk vegetation samples by pressing light-weight voluminous aliquots into much smaller Marinelli shapes. In some cases, the original dried sample occupying a volume of 3 to 5 liters was compressed to a volume of one liter by means of a press with 50,000 lbs. capacity (~23,000 kg). The increase in sample signal was not proportional to the increase in sample quantity within the one liter geometry due to additional attenuation of the gamma rays. However, even at energies as low as 100 keV, the low-Z, low-density sample material reduced the count rate by less than 50% for gamma rays originating at the outermost surfaces of the sample. Reduction in count rate for gamma rays with energies above one MeV is only about 25% for the extremities, with an average attenuation of about 15%.

Increased continuum interference produced by Compton interactions of the higher energy gamma rays were observed and also reduced the sensitivity for lower energies. For example, Compton background from the 1460-keV gamma ray of ^{40}K in some samples containing large concentrations of potassium introduced significant continuum interference. Fresh vegetation that had high water content could not be pressed to a density much greater than one g/cm^3, but pressing did reduce packing density errors which are associated with non-uniform hand-packing. Although inhomogeneity continued to be a source of errors, the improved source-to-detector distance reduced the magnitude of these errors. The minimum source-to-detector distance for an aliquot of sample occupying 4 liters when loosely packed was identical to that of the aliquot when compressed to only one liter. However, the maximum source-to-detector distance was significantly reduced.

A varying-density problem was encountered when samples were pressed into a Marinelli shape using a one-pellet operation. The volume of sample in the center portion of the end component was more dense than other fractions of the sample when the pressing pressure was too low to cause the sample to flow. Some samples were very difficult to remove from the pressing mold when the pressure was sufficient to cause flow. Hollow right-circular cylinders were found to be much easier to work with since the sample is easily removed by pressing the sample out of the mold. Other advantages of the cylinder geometry are better homogeneity of sample density and a symmetrical shape for estimating counting errors due to geometry effects.

Discussion

The low-level counting system described in this work provides a significant increase in system performance for a relatively low-cost investment. A forty-fold reduction of system background above one MeV and a ten- to twenty-fold reduction at energies below one MeV is easily achieved by application of the techniques described herein. Typical spectra are shown in Figure 2 for comparison. Counting periods in excess of one week are sometimes required to obtain adequate counting statistics. (Variations in the ambient laboratory temperature must be avoided during such long counts to minimize the effects of any resulting gain shift or effective resolution degradation and the corresponding deterioration of detection sensitivity.[8])

Airborne contaminants are removed by the simple and readily implemented technique of flushing the sample chamber with nitrogen exhausted from the cryogenic dewar. Samples can be protected from atmospheric radon and radon-daughter contamination before loading into the antechamber by encapsulation with polyethylene bags. Although these inexpensive bags are not totally impervious to radon, two sealed bags have proven to be very effective in reducing the concentrations of radon and radon-daughter radionuclides. The introduction of airborne contaminants to the system can be reduced further by temporary storage of samples to be counted in an antechamber which is flushed with the nitrogen exhaust.

The sensitivity of the overall system is additionally increased by the use of bulk samples. Large samples require special care because of errors which may be introduced due to calibration, increased chance of inhomogeneity in the sample, variable densities within the prepared sample, etc. However, the reduced sampling errors made possible by the larger samples and the improved counting statistics will often far outweigh the relatively minor errors introduced by the more massive sample. The geometry that seems most amenable to counting low-density, high-bulk samples is a hollow cylinder because the sample can be significantly compressed and the resulting cylinder can be centered over the active volume of the diode. Optimal placement of the sample is determined by merely adjusting the location of a homogeneous calibration

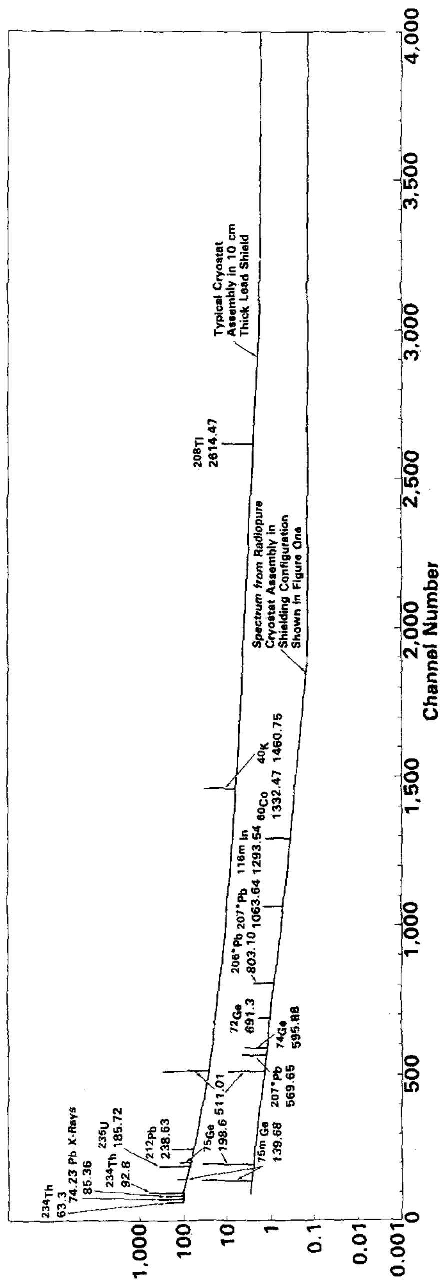

Fig. 2. Background spectra obtained with a single germanium diode (31.5% efficient) in both typical and low-level cryostat and shielding configurations

standard along the axis of the diode so that maximum counting efficiency is achieved. Approximately 16% of a one-liter sample in the current PNL Marinelli-beaker design is located near the face of the detector. This portion of the sample must be distributed to locations farther from the detector when using the hollow-cylinder geometry. However, the higher densities realized by increased die pressures may allow sufficient additional sample to be included in the calibrated geometry to reduce losses from the less efficient counting geometry.

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