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A new shallow underground gas-proportional counting lab—First results and Ar-37 sensitivity

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HIGHLIGHTS

- A new PNNL shallow underground laboratory is operational.
- A low-background gas proportional counting system for argon has been prepared.
- First background data has been collected relevant to an Ar-37 signature.
- First calibration measurements of a low-level standard have been made.
- Detector response to Ar-37 has been calculated and Ar-37 sensitivity projected.

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ABSTRACT

A new ultra-low-background proportional counter was recently developed with an internal volume of 100 cm^3 and has been characterized at pressures from 1–10 atm with P-10 (90% Ar, 10% methane) gas. This design, along with a counting system providing event digitization and passive and active shielding, has been developed to complement a new shallow underground laboratory (30 m water-equivalent). Backgrounds and low-level reference materials have been measured, and system sensitivity for ^{37}Ar has been calculated.

1. Introduction

The noble gas radioisotope ^{37}Ar is of great interest in the detection of underground nuclear tests. The production of ^{37}Ar via the reaction ^{40}Ca (n,α) ^{37}Ar has a relatively high cross section and is expected to provide a signature of large numbers of neutrons interacting with the soil (Riedmann and Purtschert, 2011). As a noble gas, ^{37}Ar is expected to migrate to the surface following an underground nuclear test without chemical effects. With a 35-day half-life, it is long-lived enough to allow time for soil–gas sampling to occur, but is expected to be present at relatively low background levels under normal conditions and has a very low atmospheric background. Because argon is abundant in the atmosphere at about 0.93% by volume, enough argon is available in a sample to act as a carrier for any ^{37}Ar and thus simplify chemical separation. The only disadvantage it carries is rather challenging detection, as it decays via electron capture, emitting only low-energy Auger electrons and x-rays. Perhaps the most well-known high-sensitivity measurement of ^{37}Ar was performed as a means of measuring the solar neutrino flux incident on the earth (Cleveland et al., 1998). The major emissions of ^{37}Ar decay are summarized in that work. In this work, only two K-capture decay channels are important, both with total energy summing to 2.82 keV. In the first, 81.5% of decays yield only Auger electrons summing to 2.82 keV. This is the easiest decay channel to detect, as the electrons will deposit their energy in a short range in the proportional counting gas at typical spectrometer pressures. Thus full-energy detection efficiency for the Auger electron emissions will be high. In the second decay channel of interest, an additional 8.7% of decays yield 2.82 keV mostly or completely as x-rays. The efficiency for detecting the full energy of these x-ray emissions will vary by proportional counter operating pressure and geometry; at the 7 atm pressure used in this work the detection efficiency will be high. Thus the overall branching ratio for detectible emissions can be expected to approach 90.2%.

The ideal laboratory capability for ^{37}Ar measurement would allow measurements well into the expected soil gas background range of about 1–200 mBq m^{-3} air (Riedmann and Purtschert, 2011) and would provide sufficient capacity that worldwide background characterizations could be supported. Several laboratories

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worldwide have this capability, notably the University of Bern; in recent years, the authors have noted the need for a U.S.-based capability focused on ^{37}Ar and having both the desired sensitivity and capacity for parallel measurements. Filling this gap was one of the motivations for the recent development of a shallow underground laboratory and new proportional counter design at the Pacific Northwest National Laboratory.

2. Proportional counter design

A long history of successful low-level gas-proportional beta spectrometers precedes this work. Necessarily, this work rests on that solid foundation and the new PNNL design sought to provide a detector capable of good beta energy resolution, low energy thresholds, capable of holding argon samples ranging up to 1.0 l, and providing the best possible background performance through an integrated system of passive and active shielding with digital signal processing of individual pulses.

The point of departure from prior work for the new PNNL design, known as the Ultra-Low-Background Proportional Counter (ULBPC), was to use materials of better radiopurity than previously available. Coupled with suitable active and passive shielding, this would allow thicker wall construction and thus higher pressure operation without background penalty from additional materials. The ULBPC design uses electrochemically-purified copper as the primary construction material. Copper in general can offer high radiopurity, and the PNNL process further reduces bulk and surface backgrounds through electrochemical purification (Aalseth et al., 2005) and chemical surface treatments (Hoppe et al., 2008, 2007). This electrochemically-purified copper offers the potential for very low surface and bulk material backgrounds from alpha and beta emitters. Clean plastic resin (PCTFE) was used for insulating and gas-seal components, taking advantage of its good radiopurity (Zuzel and Simgen, 2009) and extremely low gas permeability. The only other material employed within the active volume of the design is a high-purity niobium anode wire of 25 μm diameter. Details of the design and characterization of this design have been published in Aalseth et al. (2009), including spectroscopic performance and operating voltage over a 1–10 atm fill pressure range with P-10 gas (90% argon, 10% methane).

3. Counting system and data acquisition

To allow the beta spectrometers to reach their full potential, significant shielding and instrumentation are needed. A graded shield was designed with active and passive elements. This, along with data acquisition electronics, is known as the Ultra-Low-Background Counting System (ULBCS). The details are given in Seifert et al. (in press); the main features are summarized here. From the exterior and moving inward toward the detectors the shield consists of: (1) a gas-tight enclosure purged with liquid nitrogen boil-off gas, with airlock for detector entry and exit, (2) an active anti-cosmic veto detector consisting of 5-cm-thick scintillating plastic panels covering five sides of the shield. These panels are instrumented each with two photomultiplier tubes, are gain-matched, and all signals are summed via charge-integrating preamplifier into a global veto signal. The sixth side is left uncovered to allow for a moving door, (3) a layer of 30% borated polyethylene with a thickness (2.5 cm) selected to provide opacity for thermal neutrons, (4) a passive lead layer (15 cm) selected to limit projected gamma interactions to less than one event per day in an ULBPC, given a typical laboratory gamma background source term and (5) a passive high-purity copper inner layer with a thickness (7.6 cm) selected to limit projected gamma interactions to less than one event per day in an ULBPC, given modeled bremsstrahlung gamma production due to ^{210}Pb (~20 Bq/kg) in the modern lead shielding.

The shield is designed to allow up to 12 proportional counters to operate simultaneously. All active preamplifier electronics are external to the passive shielding but located within the radon exclusion enclosure. The potential for radon emanation from preamplifier electronics and cables inside the radon exclusion is mitigated; nitrogen purge gas is introduced into the inner area of the shield and then makes its way to the outer volume, thus providing several liters/minute of continuous flow counter to radon diffusion.

Each ULBPC is instrumented with a charge-integrating preamplifier (Canberra model 2006). After preamplification, detector signals go to high-speed event digitizers, Pixie-4 modules manufactured by x-ray Instrumentation Associates. List-mode time-stamped events are recorded; in addition to time, energy, and coincidence information, a 13 μs digitized trace of each integrated pulse is stored for off-line pulse-shape analysis (Aalseth et al., 2013).

4. Shallow underground laboratory

Due to the environmental restoration of the DOE site where PNNL is located, the prior facilities in which the authors worked were slated for demolition. Thus, design work began in 2004 to plan the "Capability Replacement Laboratory", a set of new facilities to support current and future research. At that time specifications were developed for a shallow underground laboratory for low-background research. The result in 2009 was the PNNL Shallow Underground Laboratory, detailed in Aalseth et al. (2012). Depth and overburden were designed to provide 30 m water-equivalent (mwe) shielding. This was projected to reduce cosmic-ray backgrounds by a factor of six, and high-energy neutron backgrounds by a factor of 100. Important for this work, the observed cosmic-ray rate as seen in the ULBCS active shield was reduced by a factor of 6.1 over surface-lab testing, thus meeting design goals for shielding.

5. Backgrounds

Background data have been collected from ULBPC prototypes operating in the ULBCS. Considered here are high-gain background data, with a spectrometer hardware dynamic range of approximately 0.25–20 keV. For this work, an analysis threshold of 1 keV is used and the range from 1 keV to 15 keV is considered, with events summed into 0.2 keV bins. The high-gain data illustrate the background for an ^{37}Ar measurement in the energy range including the 2.82 keV ^{37}Ar signal.

A series of analysis cuts are applied to the digitized events before a net background is calculated. First, events in coincidence with the anti-cosmic veto shield are rejected; this eliminates about 93% of the gross counts. Next, pulse-shape discrimination (PSD) is applied to identify events not arising from gas gain, i.e. no charge deposition in the argon gas. These events predominantly arise from high voltage micro-discharge, typically amounting to less than ten events per day. Details of efforts to reduce micro-discharge events are given by Mace et al. (in press). Further details of the overall analysis method, including PSD, are given by Aalseth et al. (2013).

The cosmic-ray-vetoed events, pulse-shape-discriminated events, and net background can be seen as a stacked histogram in Fig. 1. In the figure, the dramatic effect of cosmic-ray veto shielding can be seen in the interval between the upper line and the next lower one. Note the rising edge of the cosmic-ray