Calibration of an Ultra-Low-Background Proportional Counter for Measuring ^{37}Ar

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Pacific Northwest NATIONAL LABORATORY
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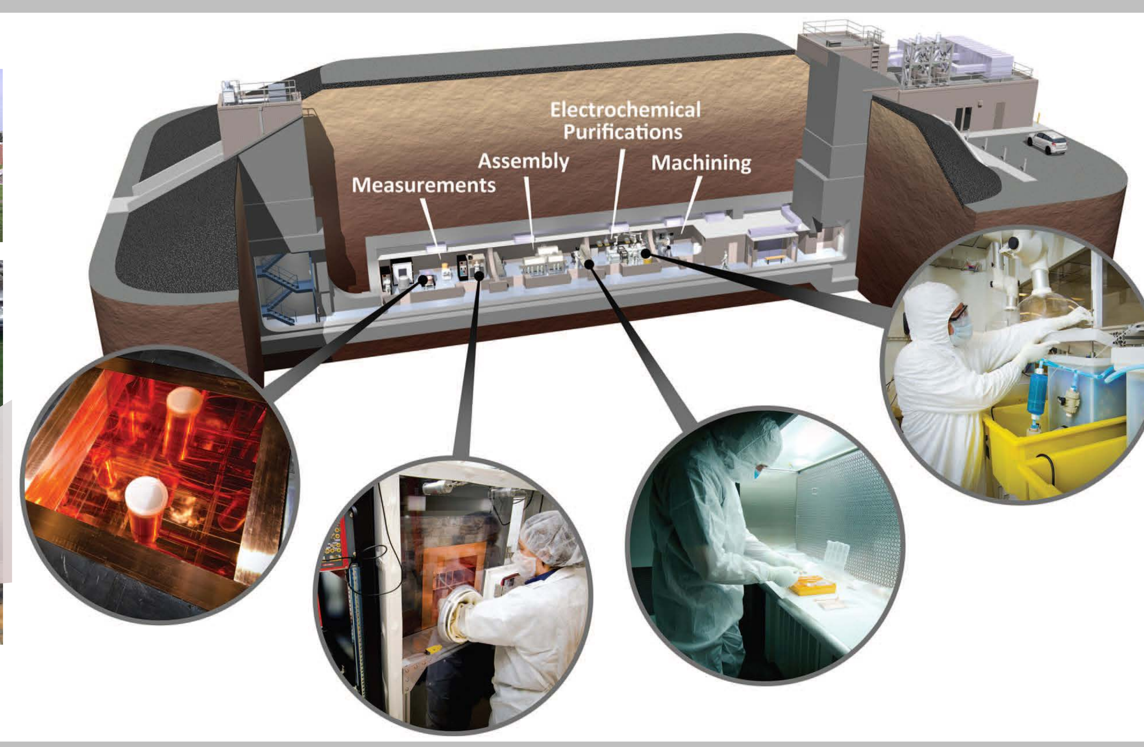
Abstract

An ultra-low-background proportional counter (ULBPC) design has been developed at Pacific Northwest National Laboratory (PNNL) using clean materials, primarily electrochemically-purified copper. This detector, along with an ultra-low-background counting system (ULBCS), was developed to complement a new shallow underground laboratory (30 meters water-equivalent) constructed at PNNL. The ULBCS design includes passive neutron and gamma shielding, along with an active cosmic-veto system. This system provides a capability for making ultra-sensitive measurements to support applications like age-dating soil hydrocarbons with $^{14}C/^{3}H$, age-dating of groundwater with ^{39}Ar, and soil-gas assay for ^{37}Ar to support On-Site Inspection (OSI). On-Site Inspection is a key component of the verification regime for the Comprehensive Nuclear-Test-Ban Treaty (CTBT). Measurements of radionuclides created by an underground nuclear explosion are valuable signatures of a Treaty violation. For OSI, the 35-day half-life of ^{37}Ar, produced from neutron interactions with calcium in soil, provides both high specific activity and sufficient time for inspection before decay limits sensitivity. This work describes the calibration techniques and analysis methods developed to enable quantitative measurements of ^{37}Ar samples over a broad range of pressures. These efforts, along with parallel work in progress on gas chemistry separation, are expected to provide a significant new capability for ^{37}Ar soil gas background studies.

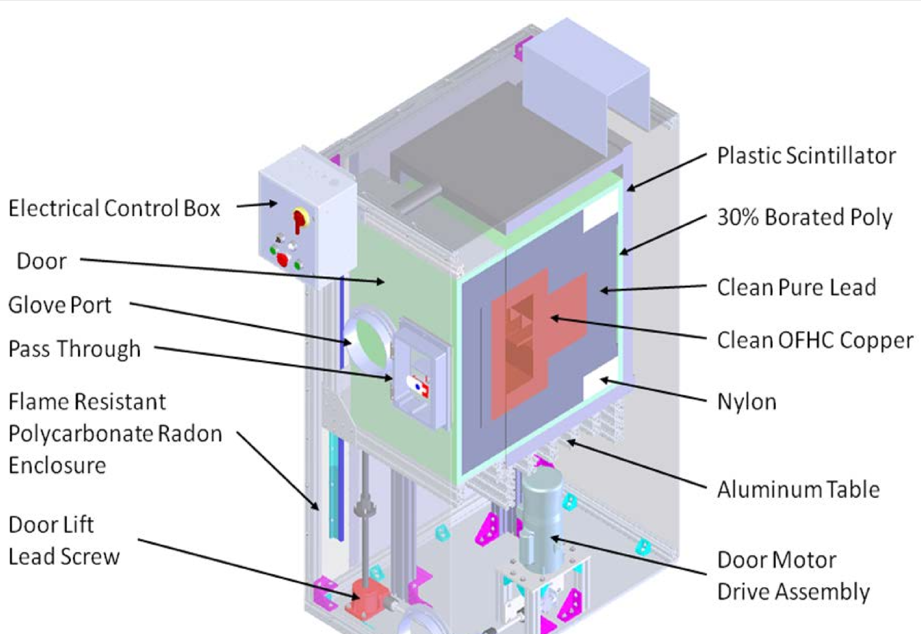
PNNL's New Shallow Underground Laboratory

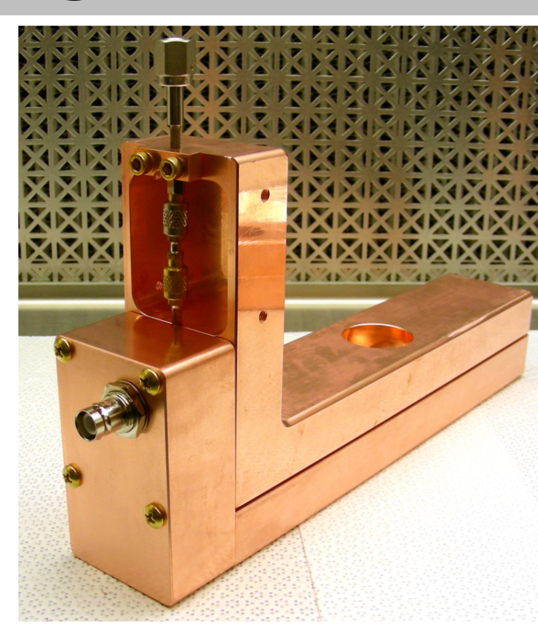
PNNL's Richland campus



- Approx. 2200 sq. ft cleanroom lab space
- Capabilities at ~30 mwe
  - Clean fabrication and machining
  - Electroformed copper production and cleaning
  - Clean detector assembly
  - Measurements hall
  - Underground materials storage
- Approximately 100x fewer fast neutrons and 6x fewer muons
  - Provides capability for low-level measurements "on campus"
  - Enables development of detector technology for deep laboratories

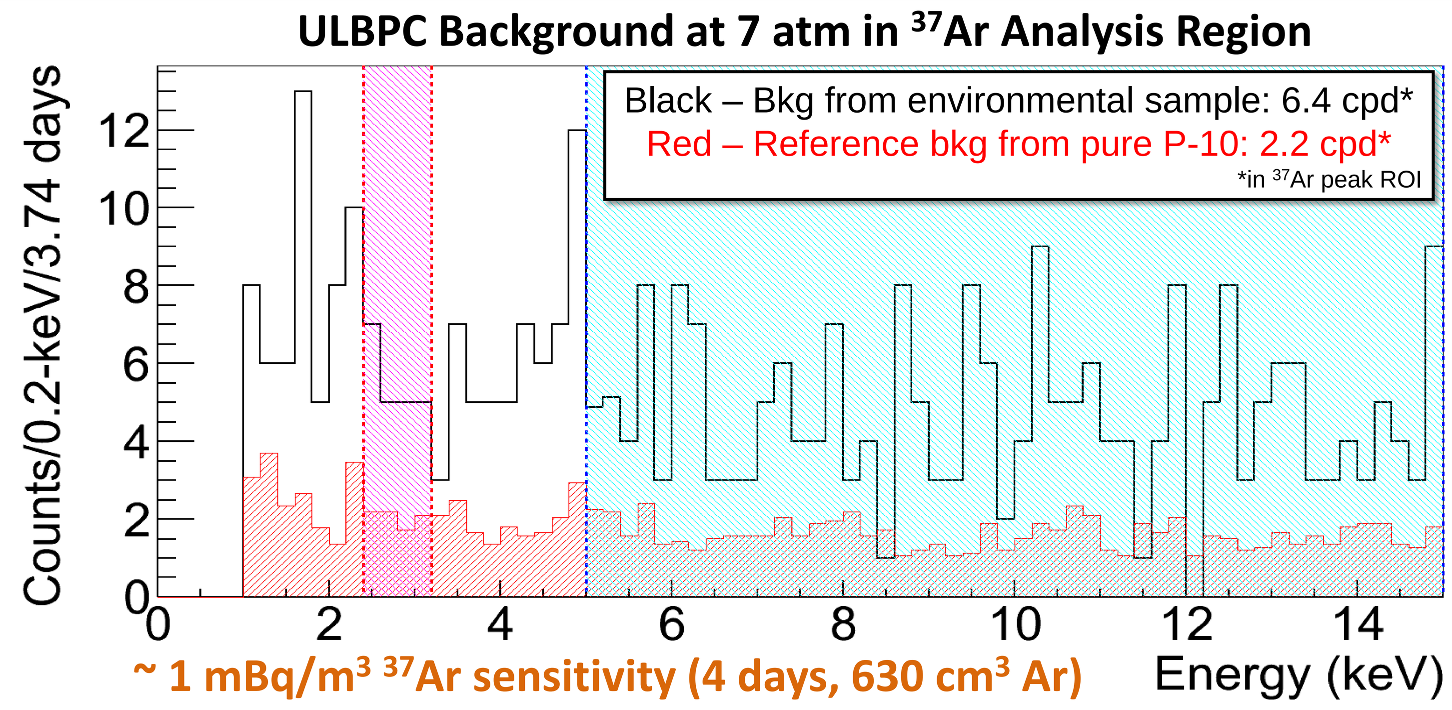
Ultra-Low-Background Counting System





Background: ~180 cpd (3-400 keV) at 3 atm in ULBCS

ULBPC Background at 7 atm in ^{37}Ar Analysis Region



~ 1 mBq/m³ ^{37}Ar sensitivity (4 days, 630 cm³ Ar)

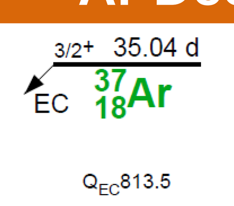
Calibration for ^{37}Ar

Motivation for Measuring ^{37}Ar

- ^{37}Ar can be produced via the reaction $^{40}Ca(n,\alpha)^{37}Ar$
  - Occurs during underground nuclear detonation via neutrons on soil calcium
  - Significant increase in ^{37}Ar activity is unambiguous evidence of neutrons
- ^{37}Ar is identified as isotopic signature of nuclear testing under the Comprehensive Nuclear Test-Ban Treaty (CTBT)
  - Specifically for collection and measurement during On-Site Inspection (OSI)
- ^{37}Ar has near-optimal 35-day half-life
  - Long enough to collect and measure, short enough to identify "recent" activity

^{37}Ar Decay Signature

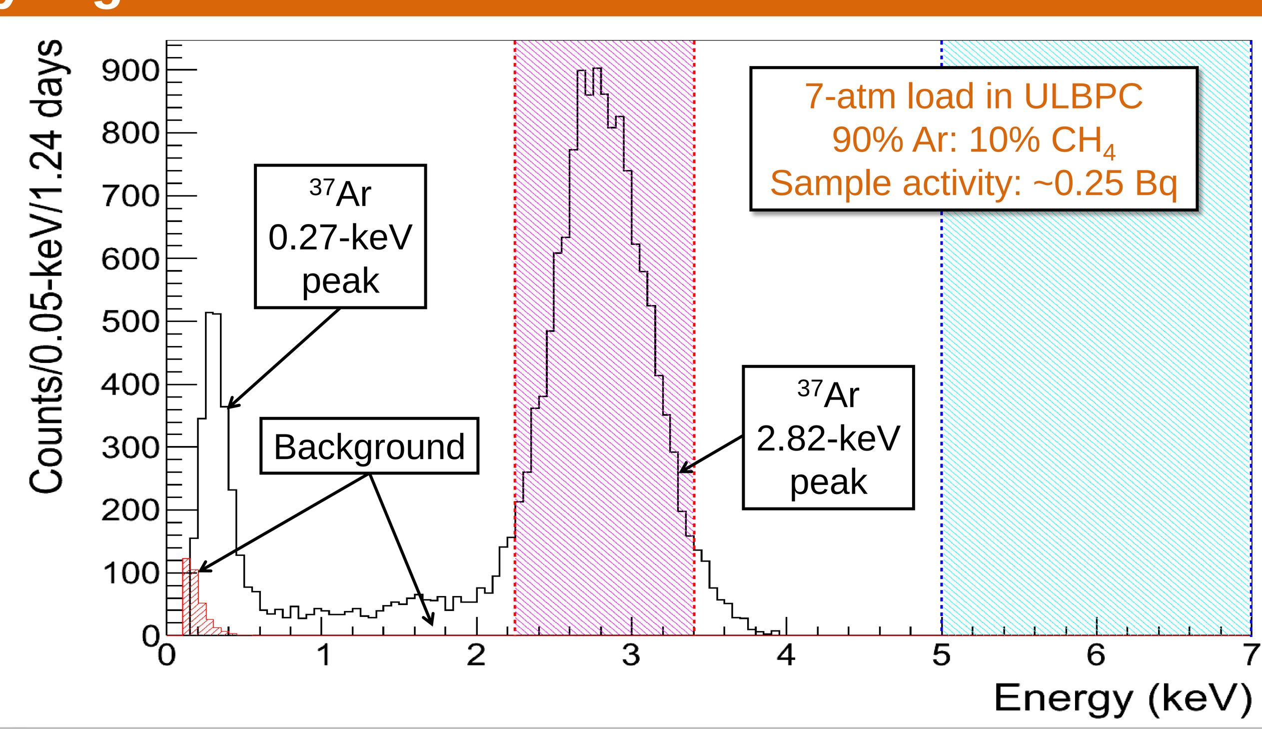
- Decay is via electron capture
  - An ~811-keV neutrino is emitted.
  - EC leaves inner-shell vacancy in ^{37}Cl.
  - Auger electrons and x-rays sum to binding energy of vacancy.
- Detection is via Auger electrons, X-rays



Principal Radiations Produced in the Decay of ^{37}Ar

Decay Mode	Percent of All Decays	Energy of Auger Electrons (keV)	X-ray (keV)
K	81.5	2.823	0.0
L	8.9	0.270	0.0
K	2.7	0.202	2.621
K	5.5	0.201	2.622
M	0.9	0.018	0.0
K	0.5	0.007	2.816

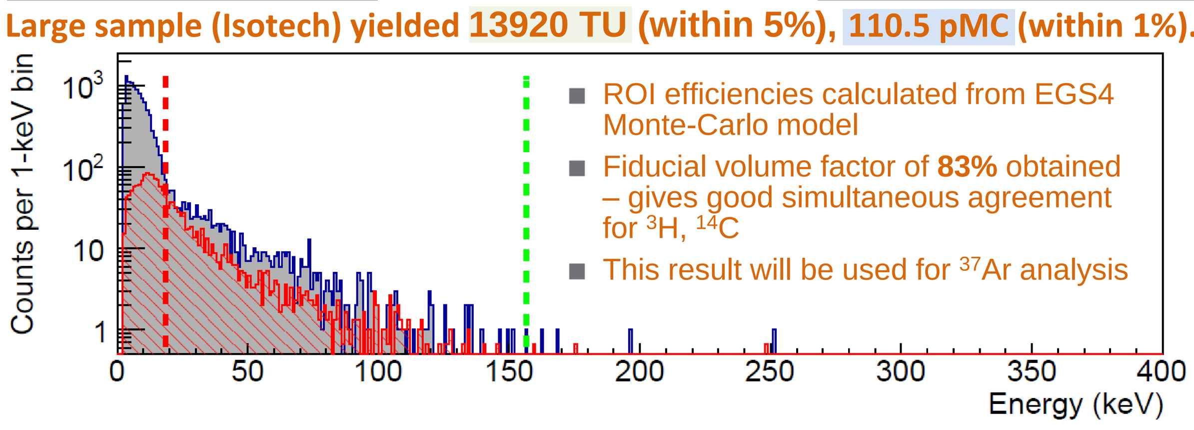
Table from B.T. Cleavland, et al., THE ASTROPHYSICAL JOURNAL, 496:505-526, 1998 March 20
Diagram from R. B. Firestone, Table of Isotopes, 8th Ed., John Wiley and Sons, New York (1996)



Fiducial Volume Calibration

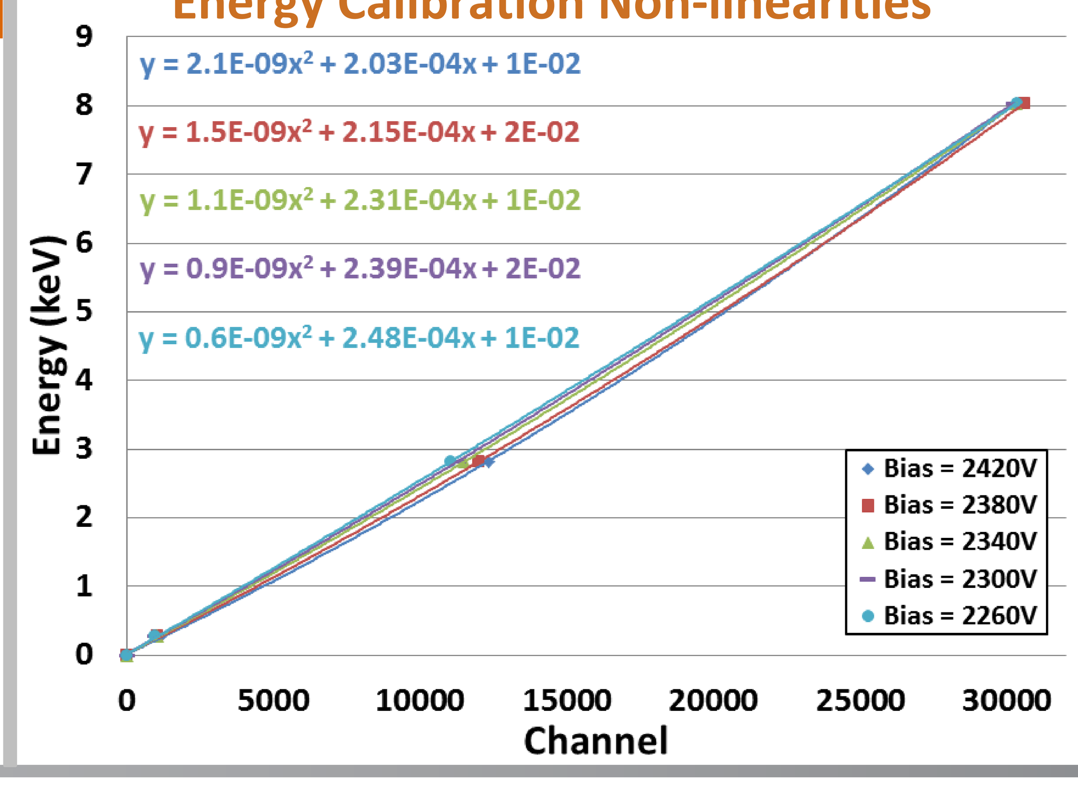
³H Analysis Results	Full ROI Analysis Results	¹⁴C Analysis Results
ROI integral (3.0-19.0 keV): 9372.9±96.8 −1007.6 bkg ±18.3 −135.3 14C ±10.7 = 8230.0±99.1	ROI integral (3.0-400.0 keV): 10416.9±102.1 −1611.7 bkg ±23.1 = 8805.2±104.6	ROI integral (19.0-157.0 keV): 1040.0±32.2 −597.1 bkg ±14.1 = 442.9±35.2
Sample decays: 13889.5±167.3	Sample LT: 6.95 d (600842.5 s)	Sample decays: 728.6±57.9
Sample LT: 6.95 d (600842.5 s)	Background LT: 21 d (1812589.6 s)	Sample LT: 6.95 d (600842.5 s)
Sample volume: 9.12 STP cm³	Gross rate: 1497.9±14.7 (cpd)	Sample volume: 9.12 STP cm³
Specific activity (cpd/cm³): 219±2.64	Net rate: 1266.2±15.0 (cpd)	Specific activity (cpd/cm³): 11.5±0.913
TU: 13227.1±159.3		Approximate pMC: 109.8%±8.7%

Large sample (Isotech) yielded 13920 TU (within 5%), 110.5 pMC (within 1%).



- ROI efficiencies calculated from EGS4 Monte-Carlo model
- Fiducial volume factor of **83%** obtained – gives good simultaneous agreement for ^{3}H, ^{14}C
- This result will be used for ^{37}Ar analysis

Energy Calibration Non-linearities



Next Steps

- Further studies to understand energy calibration non-linearities
  - Prior analyses included looking at k-absorption edge effects *(Monteiro, et al., NIM-A, 505 (2003) 233–237)*
  - Review recent findings in Dark Matter liquid Ar work and apply (if appropriate) to our work *(Sangiorgio, et al. arXiv:1301.4290 [astro-ph.IM])*
- Develop analytical model for energy calibration non-linearities
  - Must be applicable against samples (and, thus, detector fill pressures) of arbitrary size
  - Utilizes per-sample 8-keV calibration peak and prior ^{37}Ar spectrum
  - Incorporate into automated analysis routines
- Test methods for quantitative measurement against known ^{37}Ar calibration standard

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