T3.1-O6

High Throughput Argon-37 Field System

June 28, 2019

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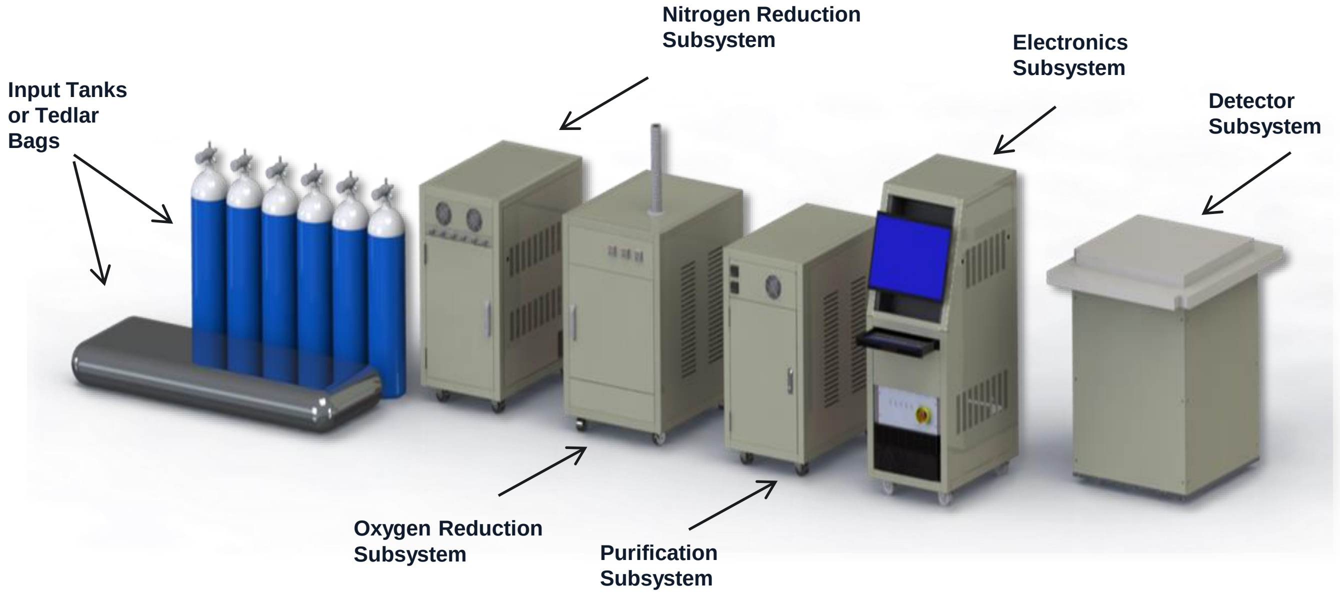
BATTELLE

PNNL is operated by Battelle for the U.S. Department of Energy

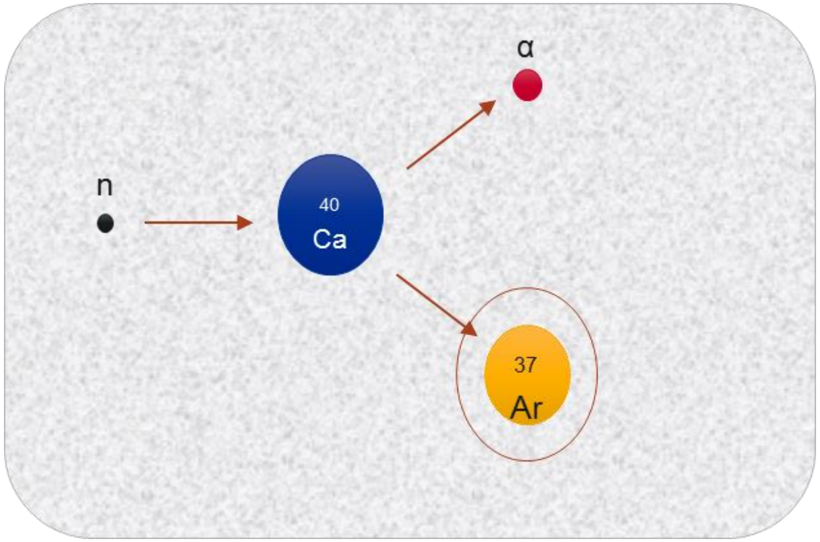
This work was supported by the U.S. Department of Energy, National Nuclear Security Administration

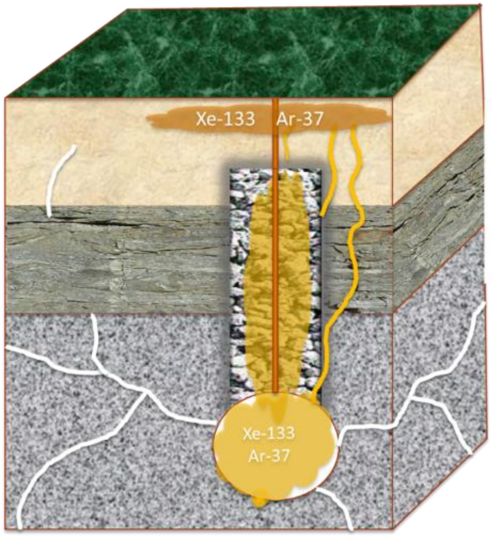
Argon-37 Field System



Argon-37

- Argon-37 (^{37}Ar) is a key indicator of an underground nuclear explosion but is one of the most difficult radioactive isotopes to measure – especially above ground
  - Argon-37 is produced when calcium in the ground is exposed to neutrons produced by an underground nuclear explosion $^{40}Ca(n,\alpha)^{37}Ar$ reaction
  - Detectable for months following the explosion (35.1 day half-life)
  - Most effective when a highly sensitive system is deployed at or near the suspected explosion site within a few months of the explosion





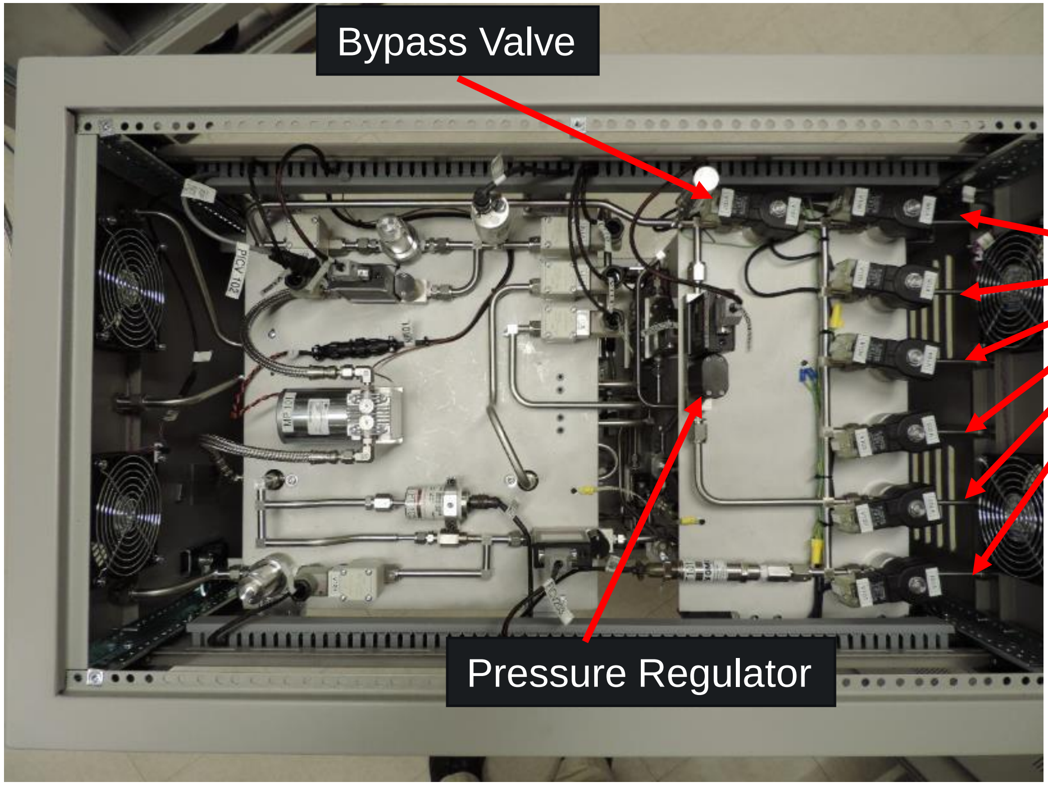
Radioactive argon atoms diffuse toward the ground surface where they are optimally collected in a soil sample to prevent air interference

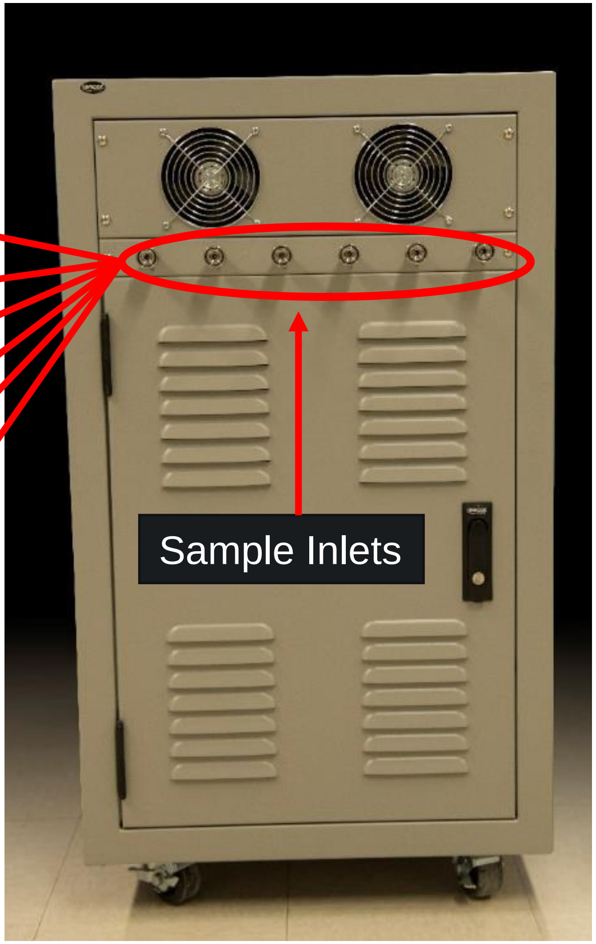
Argon-37 Field System Requirements

- Produce a high-throughput above-ground ^{37}Ar gas collection and analysis capability
- Developed for batch processing, i.e., samples collected and brought back to the system located anywhere
  - near sampling location or in laboratory setting
- Air sample types:
  - High pressure cylinders
  - Atmospheric pressure Tedlar bags
  - Ambient atmospheric air

Characteristics	Threshold (Min. Requirement)	Objective (Desired Goal)
Isotope Measured	^{37}Ar	^{37}Ar
Measurement mode	Auger Electron / X-Ray	Auger Electron / X-Ray
Count time	12 hours/Variable	12 hours/Variable
Minimum Detectable Concentration (MDC)	15 mBq/m^3 per 12 hour count	10 mBq/m^3 per 12 hour count
Operational Mode	With/without xenon system pre-processing	With/without xenon system pre-processing
Data Collection and Analysis	Automated control with operator present	Automated control
Communication	Local/remote control	Local/remote control
Power	National and International power	National and International power

Argon-37 Field System: Sample Inlet

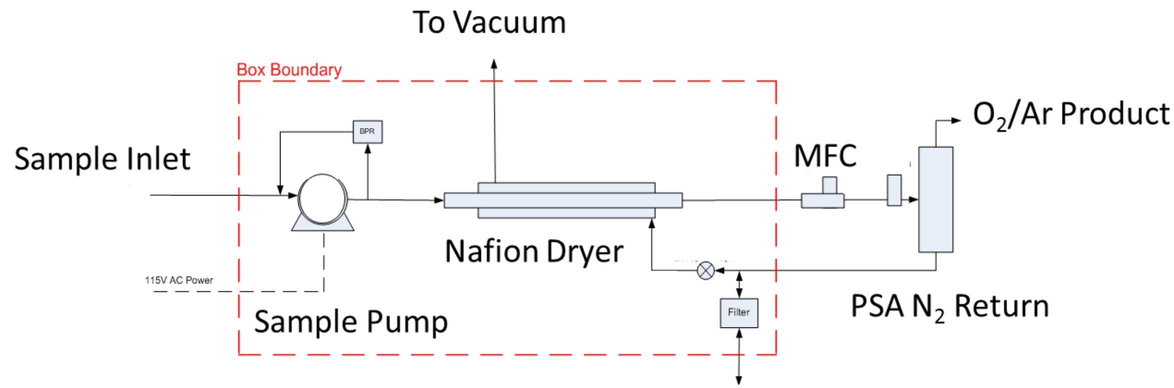


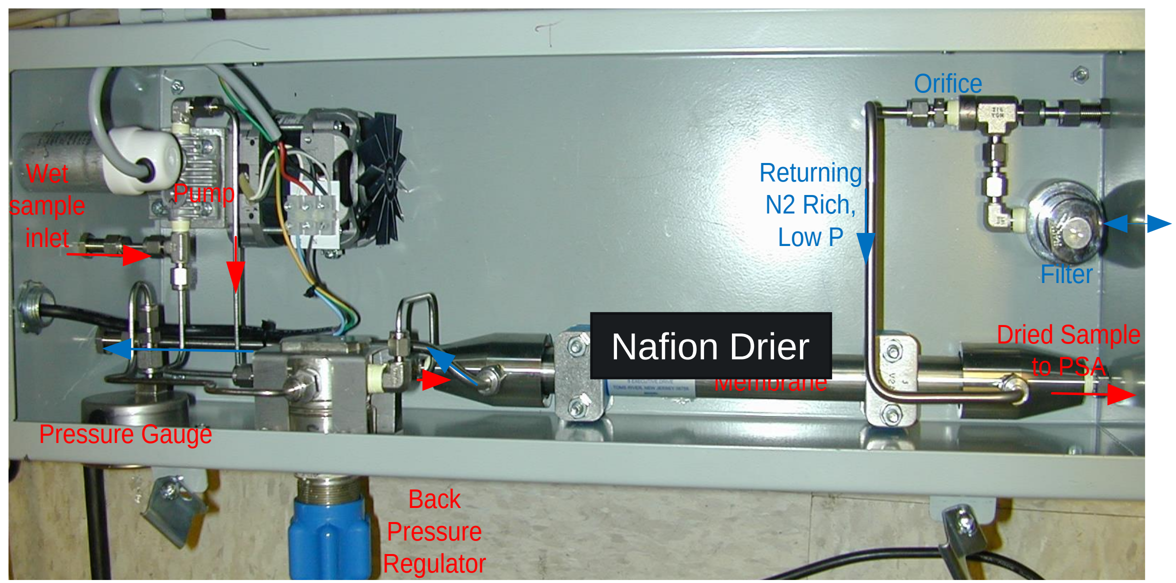


- Six samples (bottles or bags) can be attached each to a sample input port
- Samples are processed sequentially, automatically
- A mixture of Tedlar bags and high pressure tanks can be processed
- GUI used to specify if bottle or bag is attached

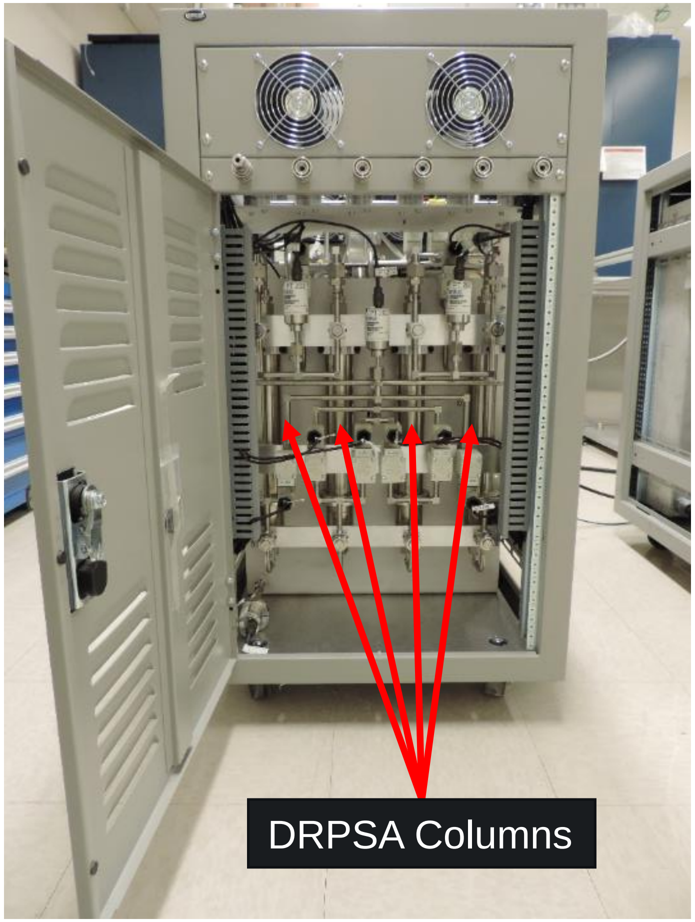
Argon-37 Field System: Water Vapor Removal

- Water content can vary depending on sampling location and sample matrix
- High pressure cylinders have low water vapor content
- Tedlar bags can have saturated water content
- Nafion dryer is used to remove water and stabilize input sample conditions





Argon-37 Field System: Bulk Nitrogen Removal

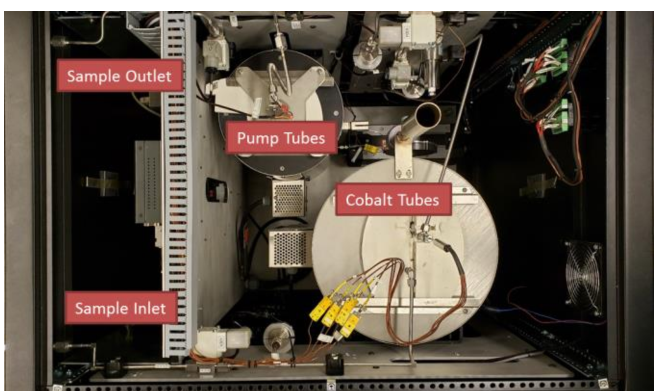


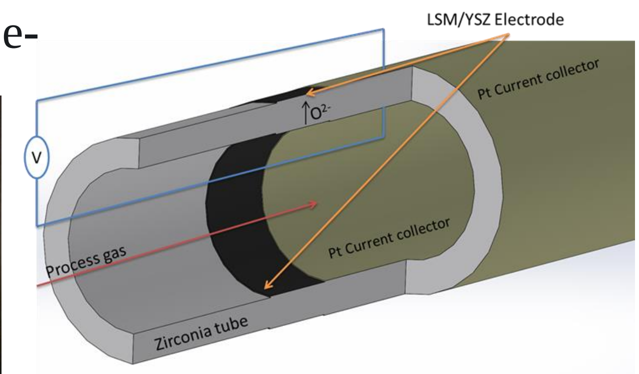
- Nitrogen makes up 78% of air which must be removed from argon. A Dual Reflux Pressure Swing Adsorption (DRPSA) process was developed for nitrogen removal
- The process removes N_2 as well as other preferentially adsorbed (retentate) species in the feed gas

Argon-37 Field System: Oxygen Removal

- Oxygen makes up 21% of air. Need to remove oxygen completely while keeping Ar recovery high
- Challenge: Argon and oxygen are extremely difficult to separate from each other
- Use two steps: bulk (1.) and trace removal (2.)
  1. The gas, containing ~95% O_2, passes over cobalt oxide at ~700 °C resulting in reaction: $CoO + \frac{1}{2} O_2 \rightarrow Co_3O_4$
  2. Yttria stabilized zirconia (YSZ) is an oxygen ion conductor at high temperature. By applying an electrical potential to the tube oxygen will be conducted from the inside to the outside of the tube:

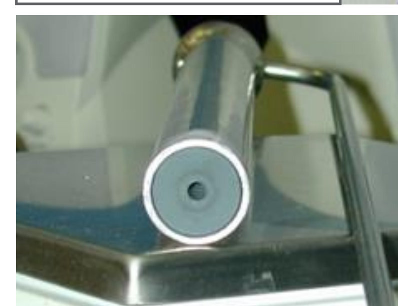
$$\frac{1}{2}O_2+2e- \Rightarrow O_2- \Rightarrow \frac{1}{2}O_2-2e-$$





LSM = Lanthanum strontium manganite
YSZ = Yttria stabilized zirconia
Pt=Platinum

Cobalt Oxide trap assembly



Cobalt Oxide trap assembly

Argon Separation and Purification

- **Separation:** Kr, Xe and Rn may remain in the argon stream, as well as trace amounts of nitrogen. All trace noble gases can be, or are, radioactive and can interfere with the ^{37}Ar measurement.
  - Cold LiLSX adsorbent can selectively remove trace noble gases and nitrogen while allowing argon to pass through the adsorbent.
- **Purification:** Argon is collected (Frozen) onto cold trap from the separation stream.
  - Argon gas quantification is performed prior to counting using high precision pressure gauge which is used to determine methane volume needed to reach P-10 (10% methane).

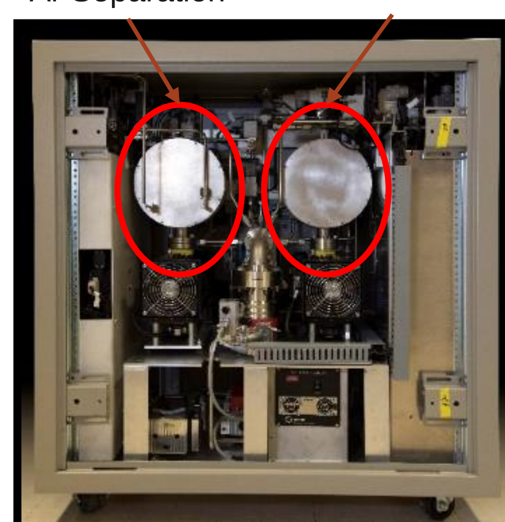
Separation/ Purification module

Ar Separation

Ar Purification

Ar Separation

Ar Purification

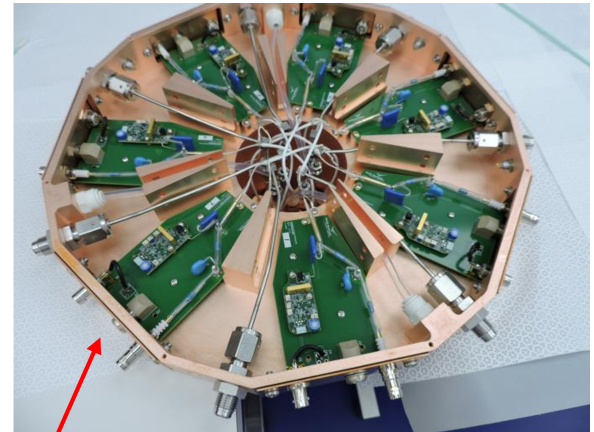


Separation trap module

Argon-37 Field System: Measurement Above Ground

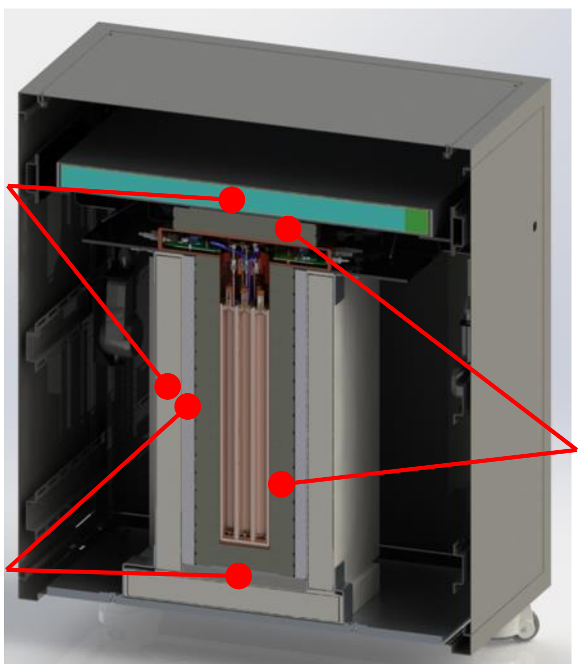
- **Nuclear Detection:** Detection is via Auger electrons, X-rays
  - 81.5% of decays emit only Auger electrons summing to 2.823 keV.
- Essentially 100% of these decays in the active volume of an internal-source gas proportional counter will be seen with a sufficiently low energy threshold.
- Designed with sensible radio-purity for above ground usage:
  - Machined from Oxygen-free high thermal conductivity (OFHC)
  - Low Radioactivity internal springs made from Polychlorotrifluoroethylene (PCTFE)
  - Niobium wire anode
- Unique pre-amplifier boards developed in house
  - Installed near detector

Argon-37 Field System: Detector Design



Electronics platter (with preamps)

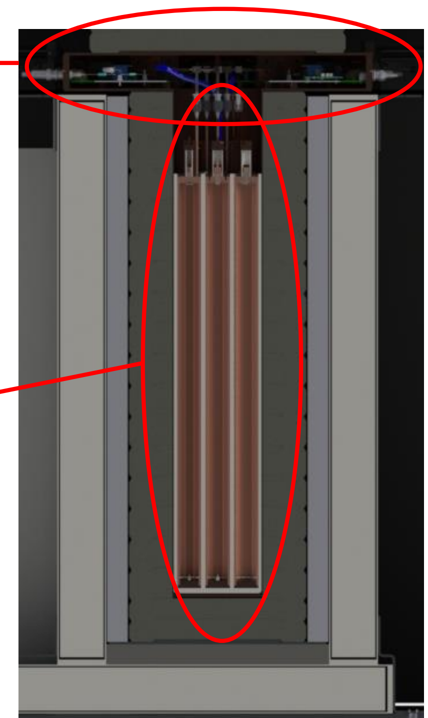
Muon veto panels



Borated poly

Lead shield

Proportional counters



Argon-37 Field System: Testing Conclusions

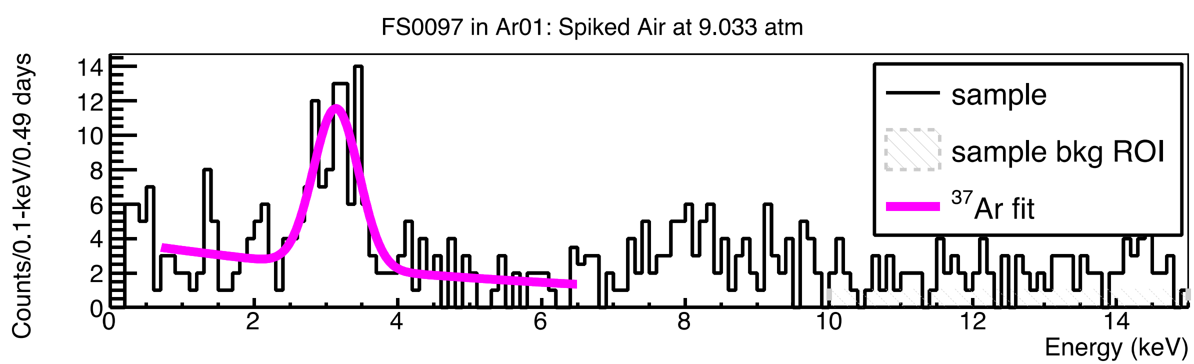
- The Argon-37 Field System was recently tested against strict requirements including verifying the MDC met the requirements
  - MDC requirement: 10 mBq/SCM_{air} for 2-liter STP argon in 12-hour count
- System exceeded MDC Requirement quantifying 10 mBq/SCM_{air} ^{37}Ar air standard

Detector volume: 250.00 cm^3
Fill temperature: 295.20K
Argon fill pressure: 6187.0 Torr
Argon sample: 1908.13 cm^3 STP
STP = 273.15K, 100 kPa (IUPAC)
Collection stop (not specified)
Measurement start (not specified)
Sample LT: 0.49 d (42295.9 s)

^{37}Ar Constants/Fit Parameters
^{37}Ar peak energy: 3.1 keV:
Peak width (σ, from calibration): 0.396 keV
BR (incl. x-ray efficiency) factor: 90.2 %
Fiducial volume estimate: 91.5 %
Overall ^{37}Ar detection efficiency: 82.6 %
Sample Bkg ROI (10-15 keV) =
103.0±10.1 counts (210.4±20.7 cpd)

^{37}Ar Constrained Fit Analysis
(1-σ uncertainty estimates from fit)
Constant bkg = 5.551e-09±2.91
Expo bkg height = 3.911±0.754
Expo bkg scale = 6.086±1.91 (1/keV)
Gaussian Area = 73.12±10.3
Gaussian Centroid = 3.142±0.0513
Gaussian Width = 0.3166±0.12
Gaussian FWHM = 23.72±0.392 %.

DISCOVERY of an ^{37}Ar signal (7.08σ)
^{37}Ar activity = 0.002104±0.000297 Bq
In whole-air (STP) equivalent:
10.26±1.45 mBq/m^3
(corrected to measurement start)



Thank you