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Pacific Northwest NATIONAL LABORATORY

# Fractional release of argon from activated rocks and powder

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**The emanation fraction of radionuclides has been highlighted as a known source of uncertainty, particularly in the case of activation products. This work serves as a first step in quantifying this release term and demonstrates that measurable quantities of argon may be released.**

## Objectives

Measure the emanation of:

- $^{37}$Ar produced in various rocks and powders during irradiation
- $^{37}$Ar from different rock grain sizes
- $^{39}$Ar produced in various rocks and powders during irradiation

## Introduction

Estimations of radioactive argon concentrations, from background sources and from an underground nuclear explosion (UNE), are limited by a lack of knowledge of the emanation fraction.

Current estimates either provide only the expected production rate in various geologies or they use the emanation coefficients of other noble gases such as radon.

In order to accurately predict signatures of a UNE, a better understanding of the emanation of radioactive argon isotopes is needed.

## Method

Samples were irradiated on the Godiva assembly at the Nevada National Security Site's Device Assembly Facility (DAF)

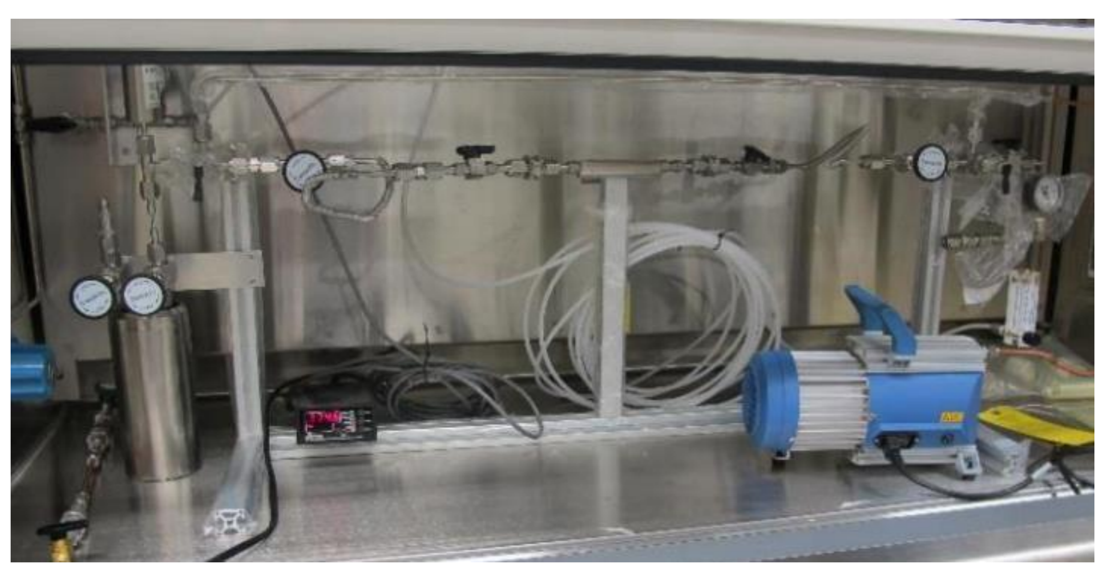
Seven sample materials sealed in quartz ampoules:

- Calcium Carbonate ($CaCO_3$) powder
- Potassium Carbonate ($K_2CO_3$) powder
- Limestone
- Dolostone
- Obsidian
- Rhyolite
- Tuff

Irradiated samples processed to extract gas and isolate argon[1]

- Gases extracted by flowing high purity helium over the sample and through a cryo-trap
- Argon then separated and additional argon make-up gas added (from room air)

Argon loaded into an Ultra-Low Background Proportional Counter (ULBPC) and measured

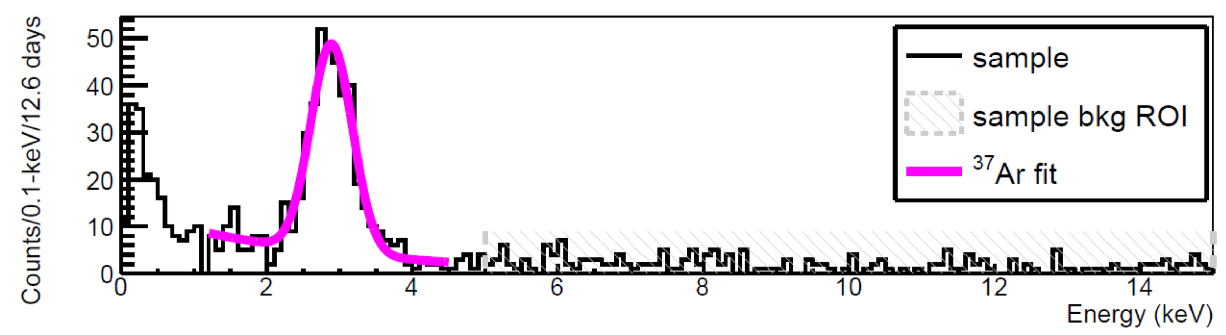


[1]Johnson C, Prinke A, Lowrey JD, et al (2018) A method to quantify the $^{37}$Ar emanation fraction in powders and rocks. J Radioanal Nucl Chem 318:297–303.

## Results

All of the samples measured had statistically significant quantities of $^{37}$Ar.

The smallest measured $^{37}$Ar signals were in obsidian, shown below. Despite the small amounts of $^{37}$Ar present in these samples, it is still detectable above the atmospheric background.




Detectable quantities of $^{39}$Ar were also measured in all of the high-potassium rocks. Measurement of $^{39}$Ar in the samples with potassium contents below 1% is currently in progress.

Average measured percent emanation for $^{37}$Ar and $^{39}$Ar, calculated by

$$\varepsilon[\%] = \frac{Activity_{measured}}{Activity_{produced}} * 100$$

The measured weight percent of Ca and K for each sample is shown. NM stands for Not Measured – early samples were not measured for $^{39}$Ar.

| Material | Ca [%] | K [%] | $^{37}$Ar Emanation | $^{39}$Ar Emanation |
|---|---|---|---|---|
| $CaCO_3$ | 38.0 | - | 2.34% | NM |
| $K_2CO_3$ | - | 56.6 | 50.79% | 22.52% |
| Limestone | 34.2 | 0.3 | 3.80% | NM |
| Dolostone | 16.5 | 0.9 | 1.03% | NM |
| Rhyolite | 0.3 | 4.5 | 34.13% | 0.86% |
| Obsidian | 0.3 | 3.9 | 0.27% | 0.09% |
| Tuff | 0.1 | 3.8 | 10.75% | 0.92% |

## Discussion

The primary $^{37}$Ar producing reactions applicable to this work are listed below along with their flux-weighted average cross sections.

| Reaction | Cross Section [b] |
|---|---|
| $^{40}$Ca(n,α)$^{37}$Ar | $2.27 \times 10^{-02}$ |
| $^{39}$K(n,n+d)$^{37}$Ar | $2.48 \times 10^{-08}$ |

While production of $^{37}$Ar is greatest from high calcium materials, the highest emanation was measured from samples with high potassium content (obsidian excepted).

For $^{39}$Ar, the primary production reactions are:

| Reaction | Cross Section [b] |
|---|---|
| $^{39}$K(n,p)$^{39}$Ar | $1.48 \times 10^{-01}$ |
| $^{42}$Ca(n,α)$^{39}$Ar | $1.66 \times 10^{-03}$ |
| $^{40}$K(n,n+p)$^{39}$Ar | $1.32 \times 10^{-04}$ |

The emanation of $^{39}$Ar has been measured to be significantly lower than that of $^{37}$Ar. Work is ongoing to determine the reason for this difference.

## Key Findings

- Measurable quantities of $^{37}$Ar and $^{39}$Ar are released from irradiated rocks
- Measurable quantities of $^{37}$Ar are produced in materials containing no Ca
- The emanation fraction of $^{39}$Ar was measured to be lower than that of $^{37}$Ar
- The emanation fraction of both $^{37}$Ar and $^{39}$Ar may vary by orders of magnitude across rock types

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