

Laboratory Gas Migration Experiments through Intact and Fractured Rock

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ABSTRACT: Presented herein are laboratory gas migration experiments conducted on samples of tuff with varying lithologies mounted within a triaxial core holder. A pressurized gas mixture standard comprised of known concentrations of Argon, Xenon, Sulfur Hexafluoride (used as a tracer), and Nitrogen was chosen based on previous work [1]. The gas mix is applied at known pressure to the upstream side of the samples to induce flow through the pore spaces and/or across fracture surfaces and the gases are detected in real-time on the downstream side using a quadrupole mass spectrometer. Downstream detection under vacuum is possible by precise metering of the gas mixture through a leak valve with active feedback control. Arrival times and time-variant concentrations of the applied gases downstream are collected for comparison between samples. We aim to determine transport properties of noble gases and SF₆, and hypothesize that transport properties vary due to solubility and water content. The parameters derived from this work will provide valuable insight into the three-dimensional structure of damage zones, including fracture networks, the production of temporally variable signatures and the methods to best detect underground nuclear explosion signatures.

1. INTRODUCTION

The detection, location, and identification of suspected nuclear explosions are United States security priorities. Currently, global nuclear explosion monitoring relies on networks of sensors to provide rapid detection, location, and discrimination of suspect events. In some cases, such as underground nuclear explosions (UNEs), prompt signals may not provide sufficiently exact locations or explosion details and the timing and amplitude of non-prompt signals may be difficult to predict algorithmically. Therefore, there is a need to conduct research into improving the post-facto identification of suspect events with emphasis on non-prompt signals. Non-prompt signal analysis can lead to an improved understanding of the detected source to aid in data-informed discrimination (e.g., explosion vs. earthquake or nuclear explosion vs. reactor).

The Underground Nuclear Explosion Signatures Experiment (UNESE) is a nonproliferation effort created to address science and research and development aspects associated with nuclear explosion verification and source analysis based on non-prompt signals. Scientific knowledge and capabilities developed via UNESE will

enhance U.S. capabilities in detecting, locating, and identifying UNEs, with particular focus on non-prompt signals. UNESE R&D results will be applicable in current and future verification and nonproliferation contexts, and as a detection-based deterrent factor for potential proliferant nations or in transparency efforts.

A critical component of the UNESE program is a realistic understanding of the post-detonation processes and changes in the environment that produce observable physical and radio-chemical signatures. As such, an understanding of noble gas migration properties through surrounding geological formations is essential for any UNESE test bed.

The goal of this research is to determine physical constants that can be used by the UNESE community to better understand the processes by which noble gases migrate through a variety of geologic materials and settings. This paper presents the experimental design and preliminary data which will help determine values such as diffusion coefficients for specific rock types at a variety of thermal, barometric, hydrologic, and weather conditions. The samples tested originated from a core hole that is at the same site (U20az) of a field gas migration study to be conducted in the summer of 2016.

2. APPROACH

Noble gas migration field studies have relied on naturally occurring barometric pumping to produce detectable concentrations of radioargon and radioxenon at the surface [1]. Using models based on observed transport characteristics of the U20az site, computer simulations [2] have allowed estimation of the effect of a pressurized explosion cavity as well as thermally driven multiphase convection. Models predict that gases will be detectable in only hours after a UNE in some cases. Other simulations have demonstrated the effect of differential migration of parent and daughter radionuclides on perturbing xenon isotopic ratios [3, 4, 5, 6].

While the understanding of noble gas migration is improving, there remain many unknowns when performing field studies. Taking advantage of testing in a laboratory setting, our goal is to perform gas migration experiments on representative rock samples controlling as many parameters as possible, thus providing a pathway to improved fundamental understanding of field experiments and modeling efforts.

The core samples used in this study are volcanic tuff and rhyolitic lava which originated from core hole U-20az-NG4. This vertical hole was drilled approximately 517 feet from the Barnwell UNE source hole. Core was collected along the entire drilled depth of approximately 1000 feet. Samples were selected from the core boxes for geomechanical testing (triaxial, uniaxial, hydrostatic, tensile, porosity, and permeability), optical microscopy using thin sections, and the noble gas migration studies described here. The samples collected were categorized into four lithologies as shown in Figure 1.

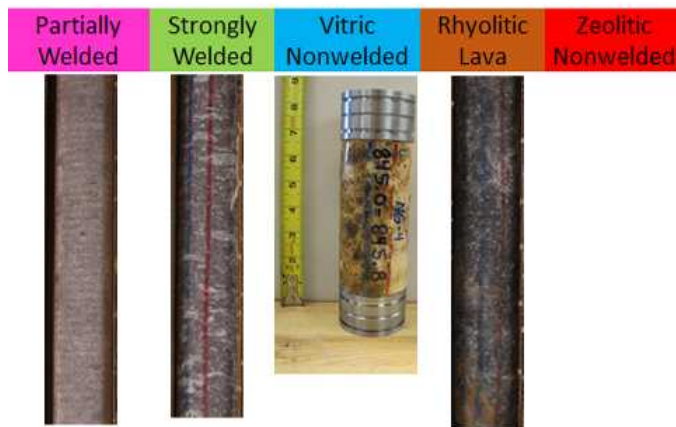


Fig. 1. Different lithologies of tuff for gas migration testing.

U-20az-NG4 is slated to be drilled deeper into Zeolitic Nonwelded tuff to a total depth of approximately 2000 feet but core from this lithology is not yet available.

The noble gas migration tests presented in this paper are from two samples that represent moderate and long

expected arrival times based on permeability testing. Figure 2 shows a plot of apparent permeability versus bulk density for samples from the four lithologies represented in Figure 1. The first sample in the legend was from a nearby but different core hole, U-20az-NG6. The colors in the legend in Figure 2 correlate to those in Figure 1. The gas migration samples are the same as those used for permeability testing and are circled in Figure 2.

Porosity was measured on one inch diameter by one inch long right circular cylinders adjacent to the gas migration samples. Porosities are 8.8% and 2.6% for depths 240.5 and 900.7 respectively.

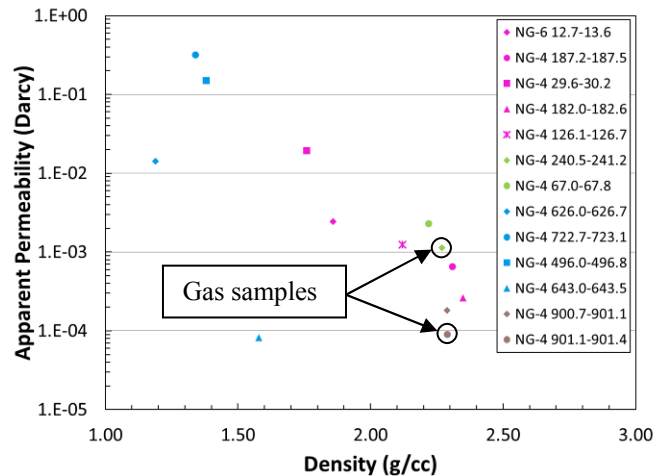


Fig. 2. Apparent permeability versus density on tuff samples of varying lithology.

3. EXPERIMENTAL METHODS/EQUIPMENT

To determine gas migration properties of the rock types, core samples were subjected to lithostatic pressure conditions and exposed to gases of interest by creating a controlled pressure differential across the sample. The downstream gas was sampled by a metering valve and coupled to a quadrupole mass spectrometer to measure the relative abundance of the gases over time.

The gas mixture was chosen based on previous work [1]. The mixture was stored in a gas bottle pressurized to ~2000 psi and contains 6.67% argon, 6.67% xenon, 6.67% sulfur hexafluoride, and 80% nitrogen. All quantities are certified to $\pm 2\%$ concentration ratios. Additional bottles of pure SF₆ and Nitrogen were used for system calibration and sample flushing/purging.

The gas mix bottle is connected to the upstream side of a triaxial core cell rated to 30,000 psi. A load frame of 220 kip capacity is used to counter the internal pressure of the piston entering the top of the pressure vessel. A quadrupole mass spectrometer (QMS) is connected to the downstream side of the sample within the triaxial core cell. Multiple pressure transducers are affixed to

the QMS to aid in metering and measuring the injected gas. One pressure transducer is mounted upstream to monitor gas inlet pressure. Figure 3 shows the three gas bottles, triaxial core cell, load frame, and the QMS system.

The methodology developed for measuring noble gas arrival times and concentration curves as a function of time adheres to the following steps:

- Prior to placing in the triaxial cell, samples are oven dried for a minimum of 24 hours
- Prior to placing in the triaxial cell, samples are placed between porous felt metal plates (to aid in gas distribution) and end caps and then jacketed with an ultraviolet cure urethane
- Sample is placed into triaxial cell and a confining pressure of 500 psi is applied
- Nitrogen is flushed through all plumbing
- The rough pump on the QMS is used to get the system pressure down to approximately 3 mbar absolute
- The QMS is used to monitor the nitrogen level of the test system. The system is deemed ready when $N_2 > 99.4\%$ to ensure nitrogen saturation within the sample and system
- A slug of gas (called a bolus) is created using a fixed volume set to 10 psi with the regulator from the gas mix bottle
- The QMS is turned on and at a specific time the bolus is released
- The QMS is set to detect specific primary isotopes of Ar, Xe, SF_6 , and N_2
- Data are recorded until all gases have reached a near steady state concentration

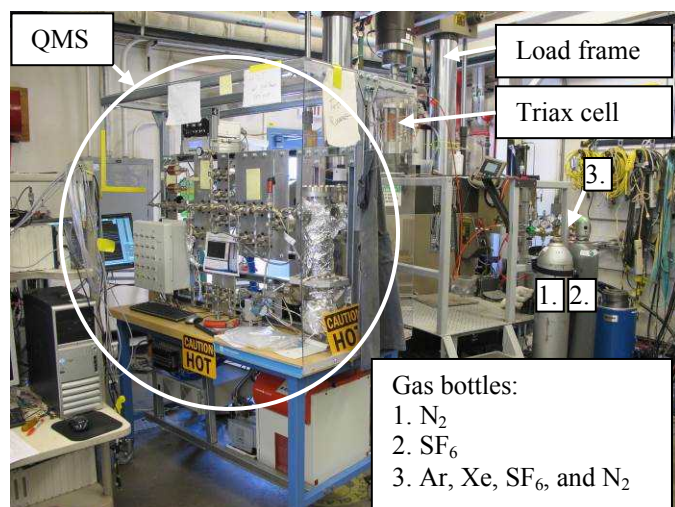


Fig. 3. Experimental setup showing gas bottles, triaxial core cell, load frame and QMS.

The above methodology was developed from much iteration; two noteworthy observations are discussed. Initially, SF_6 was not detectable on the QMS. To investigate, a bottle containing only SF_6 was procured and SF_6 was still not detectable with the QMS parameters chosen at that time. A paper studying electron impact on SF_6 [7] showed that the primary masses that need to be measured are 127 (SF_5^+), 108 (SF_4^+), 89 (SF_3^+), 70 (SF_2^+), and 51 (SF^+); i.e. the filament in the QMS source knocks off a Fluorine atom during SF_6 measurement. Furthermore, it was observed that in static mode, in the presence of the energized filament SF_x gas continues to break down into smaller parts which recombine with other atoms until the presence of SF_6 becomes completely undetectable. Once the QMS was set to detect the aforementioned masses and the QMS was used in dynamic (flow through and replenish) mode SF_6 was detected in the expected concentration of approximately 6.7% from the gas mix bottle.

Another observation was the influx of oxygen and argon when using the rough pump to prepare the gas lines for a test. Oxygen and argon would steadily increase when the rough pump went below approximately 2.5 mbar. It was discovered that when the rough pump approaches maximum vacuum, a cavitation occurs where atmosphere slowly bleeds backwards through the pump and could be detected on the QMS. To alleviate atmospheric influx, the rough pump is used to reach 2.8 mbar and then isolated from the extraction line.

In addition to releasing the gas bolus through the sample, tests were performed releasing the bolus through a precise needle valve with permeability matched to that of the sample so that a system correction could be performed. A system correction is intended to isolate the arrival times and gas concentration curves of the sample.

The needle valve was set to represent the same gas flow as the sample at a given upstream pressure. To do this, nitrogen was flowed through the sample/test system and detected with a flow meter mounted near the QMS so that nearly the same plumbing path was used for both the sample and needle valve. Nitrogen at the same upstream pressure was then directed through the needle valve and measured with the same flow meter. The needle valve was adjusted until the flow meter matched the reading given through the sample to within 0.2 ccm N_2 .

Downstream noble gas detection must be performed while under relatively high vacuum ($< 1E-5$ bar). Because the pressure of the gas on the downstream side of the sample is too high for the QMS, a metering valve couples the downstream chamber to the QMS in order to sample at a low enough flow rate to maintain adequate vacuum. The metering valve provides precise metering of the gas mixture with active feedback control via a pressure transducer. With the combination of the

metering valve and the QMS, arrival and concentration of the noble gases can be measured in real time. The effect of the metering valve on gas fractionation is yet to be determined and could affect the data, however fractionation can be corrected for with established procedure [8]. The needle valve tests may factor out gas fractionation so long as they are repeatable from test to test. Gas fractionation will be investigated further in future studies.

4. RESULTS

Six tests were performed on sample NG-4 240.5; three on the intact rock and three on the same sample with a saw cut surface (Figure 4). NG-4 240.5 is characterized as strongly welded tuff. One test was performed on rhyolitic lava sample NG-4 901.1 (Figure 5). All tests were conducted on oven-dried samples at 500 psi confining pressure.



Fig. 4. Saw cut surface on sample NG-4 240.5 (strongly welded tuff).



Fig. 5. Sample NG-4 901.1 (rhyolitic lava) shown with vented endcaps.

Data were collected from the QMS using a LabVIEW® script that tabulated data into columns of masses of gases of interest. Seventeen distinct masses were

measured. The data rate is controlled by the number of measured masses and the dwell time at each mass and resulted in approximately 3 Hz. The data were converted to an Excel® file and then read into a MATLAB® script.

The MATLAB® script normalizes both Argon and Xenon by multiplying Argon 40 by 1/0.996 and Xenon 132 by 1/0.2689; Argon 40 accounts for 99.6% of the total Argon component and Xenon 132 accounts for 26.89% of the total Xenon component (known isotopic concentrations). SF₆ is normalized by summing masses 127, 108, 89, 70, and 51. A filter with 100 point moving average is applied to the data which are then plotted as ion current versus time. In the future it is planned to make the y-axis dimensionless by normalization to initial concentration so that different tests are more readily comparable.

Figure 6 shows a plot of ion current versus time for the third test on intact sample NG-4 240.5 and the third needle valve system correction test. The curves of gas flow through the sample are lower than the needle valve test and the arrival times from the needle valve are earlier than the sample (shown on zoomed inset plot).

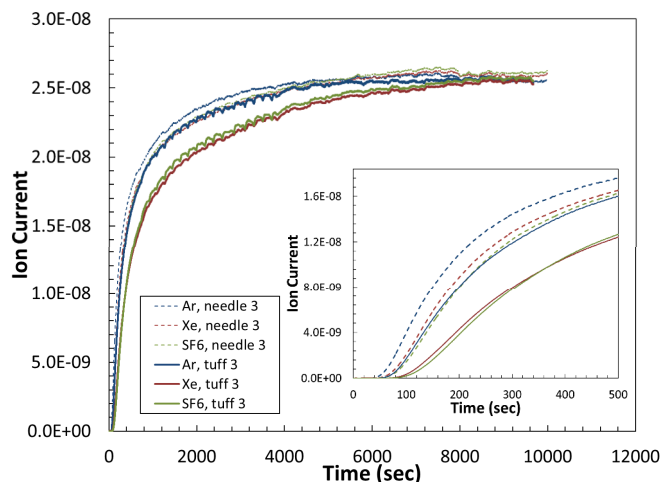


Fig. 6. Typical plot of ion current versus time for sample NG-4 240.5 and needle valve test.

As in Figure 6, Figure 7 plots ion current versus time for the third test on intact sample NG-4 240.5 but here the data are compared to a typical test on the saw cut sample shown in Figure 4. All gases arrive sooner on the saw cut sample compared to the intact test. The final ion current level is lower for the saw cut test but as described earlier, future work will incorporate a dimensionless concentration to improve fidelity of direct comparisons between tests.

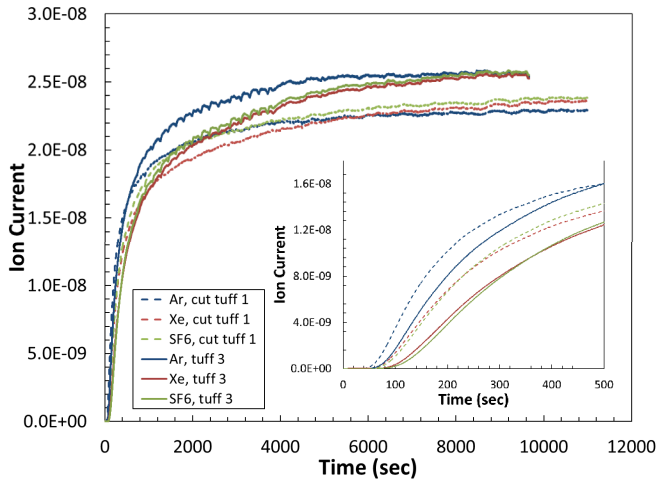


Fig. 7. Typical plot of ion current versus time for sample NG-4 240.5 and saw cut sample.

The test conducted on the least permeable sample from Figure 2, NG-4 901.1 (permeability of approximately 0.1 millidarcy) is shown in Figure 8. Again, ion current versus time is plotted. There is approximately an order of magnitude increase in arrival time for this sample compared with the intact strongly welded sample NG-4 240.5. This could possibly indicate a relationship of arrival time to other material properties such as permeability [9].

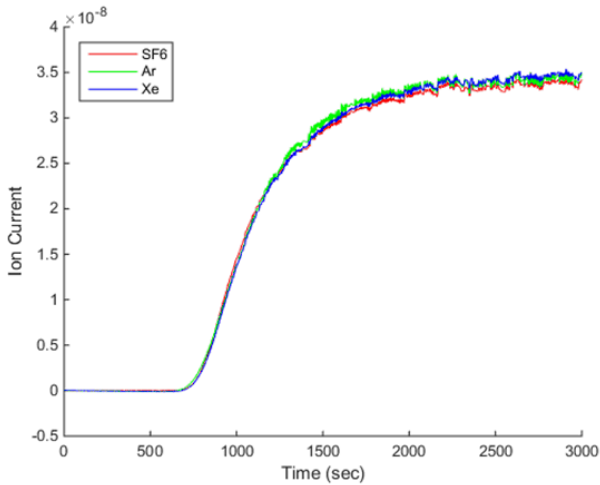


Fig. 8. Plot of ion current versus time for sample NG-4 901.1.

Data consistency is excellent indicating test methods are repeatable. Table 1 shows arrival times for three tests on both intact and saw cut sample NG-4 240.5 and the needle valve to match the intact sample. One needle valve test was performed to match the gas flow through the saw cut sample. Statistics such as average, standard deviation, and coefficient of variation are given except for the single needle valve test to match the saw cut sample.

Table 1. Arrival times for NG-4 240.5 on both intact and saw cut geometries.

UNESE NG-4 240.5-241.2	Arrival times		
Flow path	Ar	Xe	SF ₆
Needle valve (11-20-15)	44.6	60.7	63.3
Needle valve (11-30-15)	45.3	57.7	66.9
Needle valve (12-3-15)	46.3	58.4	64.6
Sample (12-2-15)	64.3	89.2	97.1
Sample (12-3-15)	65.6	86	95.1
Sample (12-7-15)	64.3	86.9	94.5
Needle standard dev	0.85	1.57	1.82
Sample standard dev	0.75	1.65	1.36
Needle average	45.4	58.9	64.9
Sample average	64.7	87.4	95.6
Needle coefficient of variation (%)	1.88	2.66	2.81
Sample CV (%)	1.16	1.89	1.42
UNESE NG-4 240.5-241.2-saw-cut	Arrival times		
Flow path	Ar	Xe	SF ₆
Needle valve (01-07-16)	37.7	48.6	55.1
Sample (01-07-16) run 1	49.9	65.6	68.9
Sample (01-07-16) run 2	49.9	63	67.9
Sample (01-08-16)	53.8	67.9	71.2
Sample standard dev	2.25	2.45	1.69
Sample average	51.2	65.5	69.3
Sample CV (%)	4.40	3.74	2.44

5. FUTURE WORK AND CONCLUSIONS

5.1. Future Work

The work presented here is ongoing. From recent collaboration with personnel performing field noble gas migration experiments and modeling, a set of parameters to vary for future laboratory gas migration experiments has been established. These parameters include:

1. Water content will be varied from dry to as wet as possible. Once the influence of these water extremes is evaluated, intermediate water contents may be investigated.
2. The experiment may be run with either a continuously varying gas pressure gradient or with constant pressure. Data presented here were collected with varying pressure where the gas pressure is at the maximum when the bolus is released and the minimum (because the gas mix is expanding into a lower pressure volume) at the end of data collection. Constant upstream pressure would be maintained with a separate metering valve downstream but upstream of the existing leak valve.
3. Samples will be tested with no fracture, a saw cut, or natural fracture. The saw cut or natural fractures will be vertical in orientation to highlight the difference of gas flow through rock matrix compared to fracture gas flow.

4. Experiments will be conducted at two temperatures, 30°C and 70°C, to investigate the effect of a thermal gradient. Only the triaxial core cell will be heated and the rest of the system will remain at room temperature.
5. On vertical saw cut and natural fractures, confining pressure will be varied to simulate different lithostatic overburdens. The maximum lithostatic pressure at the current test bed would equate to approximately 2500 psi.

The parameters listed above for future work are summarized in the test matrix for one lithology in Table 2 below. All 16 tests in Table 2 would be repeated for each lithology (Partially welded, Strongly welded, Vitric nonwelded, Rhyolitic Lava, and Zeolitic Nonwelded).

Table 2. Planned future work on one representative rock type.

Proposed test matrix for one rock type (Strongly welded)		
Test#	Condition	Legend*
1	SW, dry, ΔP, No fracture, 30°C	First Sample
2	SW, wet, ΔP, No fracture, 30°C	First or Second Sample
3	SW, dry, Pconstant, No fracture, 30°C	Second or Third Sample
4	SW, wet, Pconstant, No fracture, 30°C	Third/Fourth Sample
5	SW, dry, ΔP, No fracture, 70°C	
6	SW, wet, ΔP, No fracture, 70°C	
7	SW, dry, ΔP, vertical saw cut, 30°C, 500 psi	
8	SW, wet, ΔP, vertical saw cut, 30°C, 500psi	
9	SW, dry, ΔP, vertical natural fracture, 30°C, 500psi	
10	SW, wet, ΔP, vertical natural fracture, 30°C, 500 psi	
11	SW, dry, ΔP, vertical saw cut, 30°C, 2500psi	
12	SW, wet, ΔP, vertical saw cut, 30°C, 2500psi	
13	SW, dry, ΔP, vertical natural fracture, 30°C, 2500psi	
14	SW, wet, ΔP, vertical natural fracture, 30°C, 2500psi	
15	SW, dry, ΔP, horizontal saw cut, 30°C, 500 psi	
16	SW, wet, ΔP, horizontal saw cut, 30°C, 500psi	

*Note: Legend indicates how many samples are necessary to satisfy the test matrix. Depending on what conditions are further explored, the estimated maximum for each rock type would be four samples and the minimum needed would be three.

5.2. Conclusions

A critical component of the UNESE program is a realistic understanding of the post-detonation processes and changes in the environment that produce observable physical and radio-chemical signatures. A thorough understanding of rock and fracture properties is needed to best model a UNESE test bed at any stage in its “life”. To best support the need to accurately model UNE scenarios, an experimental program to determine laboratory noble gas migration properties of Barnwell core was developed and presented herein. Seven laboratory gas migration experiments conducted on intact and saw cut tuff have been presented. Test to test variation is excellent indicating good consistency with the established test method. The test method presented will be an excellent starting baseline for future work. These data will improve understanding of noble gas migration methods through various geologic media and ultimately improve modeling efforts.

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