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UTILIZATION OF THE NOBLE GASES IN STUDIES OF UNDERGROUND NUCLEAR DETONATIONS

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ABSTRACT

The Livermore Gas Diagnostics Program employs a number of rare gas isotopes, both stable and radioactive, in its investigations of the phenomenology of underground nuclear detonations. Radioactive gases in a sample are radiochemically purified by elution chromatography, and the separated gases are radioassayed by gamma-ray spectrometry and by internal or thin-window beta proportional counting. Concentrations of the stable gases are determined by mass-spectrometry, following chemical removal of the reactive gases in the sample. The most general application of the noble gases is as device fraction indicators to provide a basis for estimating totals of chimney-gas components. All of the stable rare gases except argon have been used as tracers, as have Xenon-127 and Krypton-85. Argon-37 and Krypton-85 have proven to be of particular value in the absence of a good tracer material as reference species for studies of chimney-gas chemistry. The rate of mixing of chimney gases and the degree to which the sampled gas

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truly represents the underground gas mixture can be studied with the aid of the fission-product gases. Radon-222 and helium are released to the cavity from the surrounding rock and are therefore useful in studies of the interaction of the detonation with the surrounding medium.

INTRODUCTION

The gas diagnostics program at LLL was established to supplement conventional radiochemical techniques for obtaining device performance-evaluation data in support of Test Program activities. We provide analytical, interpretative, and diagnostic support for Plowshare gas-stimulation experiments, and we also perform a wide variety of service radiochemical measurements in conjunction with laboratory experiments in which gas tracers are used. Much of our effort requires measurement and interpretation of the concentration of noble gases in air samples. These species are present as products of nuclear reactions, as components of any air diluent of the samples, as tracer materials, or as combinations of these possibilities. We use both stable and radioactive components. Although the nature of our diagnostic application of the noble gases is unique, the procedures we have developed are of general applicability. The occasion of this Symposium dealing with many aspects of measurement and application of the noble gases provides an excellent opportunity to describe our use of these materials.

SAMPLING TECHNIQUES

Study of the phenomenology of an underground nuclear detonation requires collection of good-quality samples of the cavity and chimney gases over times extending from a few minutes to several weeks following detonation. The sampling system now used (Fig. 1) consists basically of a reinforced rubber hose containing and fixed to a nonrotating wire rope that provides longitudinal strength and prevents crushing of the hose (Grens, 1972). The rope itself is a reasonably permeable gas path, even where the hose is compressed against it through regions of high soil compaction. Both the rope and hose terminate at a vacuum demand valve placed well downhole beneath the gas-containment stemming plug. This valve prevents passage of gas unless a vacuum can be applied from above, thus precluding the possibility of pressurized gas reaching the surface and providing a fail-safe closure of the system if the surface sampling equipment is not leak-tight. Other built-in safety features include a maximum allowable working pressure of the hose that is well in excess of any previously observed surface pressure. A deeply buried pressure relief valve ensures a safety factor of 6 or more in the upper portions of the sample system. Gas pumped through the system is totally contained either in pressurized sample cylinders or in underground tanks. Sampling operations are conducted in such a manner as to ensure compliance with the concept of total containment that characterizes explosive emplacement techniques currently in use.

Another often-used sampling scheme involves the use of production tubing placed in a postshot reentry drill hole. Although simple in concept, this technique suffers from the time delay imposed by the drilling schedule. At the test site the delay can be days or weeks and means a loss of the information obtained from short-lived radionuclides. In the case of gas-stimulation experiments, the delay has ranged from 1 mo (Gasbuggy) to nearly a year (Rulison) and has precluded detection of all but the longer-lived gaseous radionuclides. To eliminate this loss of information, we emplaced a prompt gas-sampling system at Rio Blanco (Quong, 1972). This novel system consisted of about a mile of half-inch, stainless steel tubing containing a wire rope at its lower end and three gas-block diaphrams that could be manually opened by back pressurization from the surface. Unfortunately chimney gas was not collected by the system. Our first chimney gas samples are expected in early October 1973, following completion of drilling into the chimney.

ANALYTICAL PROCEDURES

Samples are returned to Livermore for analyses. Typical sample volumes range from a few cubic centimeters to a few tens of liters, although several cubic meters of sample have been processed at times. Radionuclide concentrations range from background to a few hundred microcuries per milliliter. Needless to say, our processing equipment exists in isolated high- and low-level specialized versions

The first step in the processing of an unknown sample is a germanium-diode gamma-ray scan of 10 to 20 ml of the gas. We have standardized this process to provide concentration measurements of the short-lived krypton isotopes (87, 88, 85 m) and of xenon-135 against the background of other gamma ray-emitting nuclides in the gross fission product gas. In principle it is possible to determine all of the krypton and xenon isotopes in a gross sample by a judiciously spaced series of counts However, to save time, gain sensitivity, and improve measurement accuracy, we prefer purified krypton and xenon fractions of the sample for measurement of the longer-lived fission product gases.

Radiochemically and chemically pure fractions of a sample are prepared by elution chromatography. Helium is used as a carrier gas, and the separations are accomplished by manipulation of gas flow paths through controlled-temperature activated charcoal and molecular sieve (5A) columns (Momyer, 1960). Thermal conductivity and ionization-chamber detectors are used to monitor the progress of the elution. We have six high-level gas-separation systems and two similar low-level systems.

The desired quantity of sample is mixed with appropriate carrier gases for trace components and collected on activated charcoal at liquid nitrogen temperature. In general the purification process consists of a series of initial group separations from the activated charcoal at temperatures ranging from liquid nitrogen to 350°C (Fig. 2). A molecular sieve is used to obtain the final separation of each of the groups. A much simpler scheme is often used to obtain specific components of a moderately radioactive sample (e.g., krypton can be separated from air in a two-step process using only the charcoal column). Large samples require lower temperatures until the major constituent is eluted and may require multiple elutions from the sieve traps to obtain species such as argon from air or krypton from natural gas in a pure form. Because of the large variety of sample sizes and compositions that we are asked to process, our systems have an excess of built-in versatility and our techniques are modified accordingly. Simpler systems and techniques would suffice in a large number of specialized applications where more-or-less routine analyses are required.

Counting systems used for measurement of the radioactive constituents of the separated gas fractions are chosen according to their activity level. Low-level samples are counted in matched 0.8- and 0.5-liter anticoincidence

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QUANTITATIVE NONDISPERSIVE X-RAY FLUORESCENCE ANALYSIS OF HIGHLY RADIOACTIVE SAMPLES FOR URANIUM AND PLUTONIUM CONCENTRATION

August 14, 1973

Lawrence Livermore Laboratory
University of California
Livermore, California

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED of species of interest in a series of samples is of little value unless one is able to identify and eliminate the effects of dilution, mixing, and chemical reactions that mask the interrelationship between the samples. To accomplish this unmasking we rely on selected noble gas tracers. These species are expected to be uniformly distributed within the cavity, are measurable over a wide range of dilution throughout the desired observation period, and are not subject to excessive and variable background corrections. Three classes of tracers can be defined: relative, internal diagnostic, and emplaced diagnostic.

Application of gas tracers in a relative sense is best illustrated by our gas-quality investigation for Project Rulison (Smith, 1971). During production testing, ^{85}Kr was used as an indicator of relative sample quality and of the extent of dilution due to the influx of formation gas into the chimney during production. It was also used as a common denominator for ratios used to investigate the significance of the changing composition of the produced gas.

A diagnostic tracer is a material present in the cavity in a known amount. It can be produced by the explosive or emplaced with the explosive and, of course, must also meet the requirements previously stated for a relative tracer. The "device fraction" is the ratio of the concentration of the tracer species in a sample to its known total in the chimney. Its reciprocal is the volume of chimney gas at standard conditions. If estimates of chimney pressure and temperature are available, the reciprocal device fraction can be interpreted directly as chimney void volume and can be used to estimate the cavity radius. Most importantly for diagnostic purposes, the total of any well-mixed component of the chimney gas is given by its concentration in a sample divided by the device fraction. These totals are the most tractable quantities available for providing device-performance estimates and for studying underground nuclear phenomenology.

Totals of internal diagnostic tracers must be either calculated or measured. The Rulison ^{85}Kr total, for example, was obtained from release estimates after production testing and was verified from chimney volume, pressure, and temperature estimates and the initial concentration. Krypton-85 was also used as a secondary tracer for the Gasbuggy gas-quality program (Smith, 1970a, 1970b). In this case ^{127}Xe (the ^{127}Xe was produced by neutron capture in a xenon sample that had been enriched in the lighter xenon isotopes) ws emplaced with the device to serve as the principal tracer. Because of its 36-day half-life, the ^{127}Xe was useful during only the initial shut-in period (6 mo). Subsequent diagnostic tracing was based on the experimental total ^{85}Kr measured relative to the ^{127}Xe during this period. Internal tracer totals are sometimes estimated by calculation when measurements based on emplaced tracers are unavailable

Normally we prefer to emplace a unique tracer material with the explosive so that its total can be determined under controlled conditions. Materials that have been used include ^{127}Xe, ^{85}Kr, ^{222}Rn, and stable neon, krypton, and xenon. All except ^{127}Xe require a background correction. This is obtained for ^{85}Kr from the ^{85m}Kr seen in the samples: for radon from helium, using the known preshot radon/helium soil ratio; and for the stable gases from argon in air or helium in natural gas, using known preshot tracer/argon or tracer/helium ratios.

Due in part to the expense and handling difficulties of radioactive tracers and principally to the development of reliable trace-gas measurement techniques by Newton et al., we now rely almost entirely on stable gas tracers.

The most notable example is the Rio Blanco gas-stimulation experiment, where each of the three explosives was emplaced with a unique tracer gas. When production testing is begun, we plan to use these tracers to monitor the degree of inter-chimney communication and, if possible, to gain some insight into characteristics of the individual chimneys. For gas diagnostics of test program events, we often use stable krypton and/or xenon as the emplaced tracer material.

NOBLE GASES FROM A NUCLEAR EXPLOSION

Our studies of nuclear detonations must treat at least five distinct sources of noble gases: 1) The fission process generates many krypton and xenon isotopes both directly and as a consequence of radioactive decay of their precursors; 2) helium isotopes are among the products of thermonuclear reactions; 3) neutrons generated by the detonation interact with components of the explosive and of the surrounding medium to create noble gas isotopes through nuclear reaction processes; 4) the detonation itself vaporizes, melts, and fractures the medium surrounding it, causing the release of soil gases to the chimney voids; and 5) air normally enters the chimney region to replace the condensing steam during the cooling process and in doing so introduces an often significant quantity of noble gases that may be generally termed "background." Each of these sources can be characterized and quantized, using the analytical procedures mentioned previously. The results of such a characterization constitute a viable gas-diagnostic program

To obtain performance estimates for the explosive, we rely principally on those species expected to be present in the cavity at relatively early times following detonation. Prior to chimney collapse, dynamic mixing within the cavity assures a reasonably uniform mixture of detonation products and tracer gases, thus fulfilling a basic prerequisite for meaningful results. In practical terms this limits diagnostic fission measurements to the krypton isotopes of mass 85, 87, and 88. Three of the four are short-lived, necessitating rapid sample recovery and analysis. If sampling times are early enough so that xenon isotopes measured in the gas are attributable to their independent fission yields, then these species can be used in diagnostics. At intermediate times, interpretation of the measured totals of xenon isotopes is complicated. The xenon precursor nuclides are predominately deposited on cool rock surfaces when the chimney collapses. Since chimney-gas mixing rates are considerably slower than cavity rates and some entrapment of the gas may occur, the diagnostic application of the xenons during this period is questionable. In general we prefer to use these isotopes to study the phenomenon of gas mixing rather than for diagnostics. However, in some cases where late-time observations have been possible, we have been able to demonstrate that an appreciable fraction of the xenon was mixed with the gas and have been able to extract diagnostic information from its measurement.

Since a significant fraction of the yield of a thermonuclear explosive is due to the fusion of deuterium and tritium to produce ^{4}He and a neutron, an experimental measurement of the device-produced helium is potentially useful in diagnostics. The measurement is complicated by the presence of "background" helium in the chimney gas from air and from soil. The argon content of a sample is a useful indicator of the amount of helium (and other noble gases) present due to air dilution. Amounts of helium and radon in the soil gas are related, since both are radiogenic. If the preshot helium/radon ratio for the detonation environment is known, then the radon in a sample becomes a useful indicator of the background helium contributed by the soil and is also a useful indicator of the amount of rock that has been fractured by the detonation.

Neutrons produced by the detonation are eventually captured by materials in the device and its immediate surroundings. Although certain isotopes of each of the noble gases are potential products of these reactions, the most abundant such activation product is ^{37}Ar produced principally from calcium. Production of the ^{37}Ar occurs at zero time and within the rock sphere about the explosive that will become molten. It is, therefore, uniformly distributed and well mixed with other detonation products in the expanding cavity. Because of this, we have found ^{37}Ar to be an extremely useful relative gas-tracer material, and we have also used it as an internal diagnostic tracer for samples that are too diluted to provide a useful stable gas-tracer measurement. In the latter application, the total ^{37}Ar must first be known from the results of measurements on good-quality samples, since calculational estimates of activation processes are quite imprecise.

CONCLUSION

Throughout this presentation I have attempted to provide a generalized summary of our gas-diagnostics program and of its utilization of the noble gases in studies of underground nuclear detonations. The results of some of these investigations have been published in connection with the Gasbuggy and Rulison experiments (Smith, 1970a, 1970b, 1971, 1972). The specifics of our investigations do not easily lend themselves to generalization, as each event is unique unto itself in many ways. Such detailed presentation of our results is necessarily relegated to individualized topical presentations. My goal here has been to draw upon our experience in gas analysis techniques to describe concepts that are of general interest and applicability.

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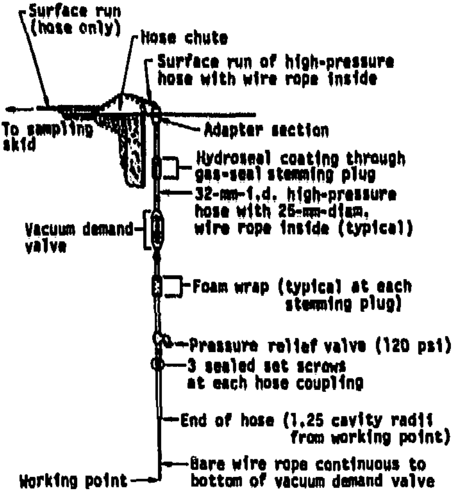
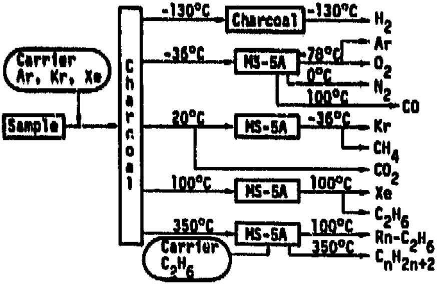
FIGURE CAPTIONS

Fig. 1. Gas-sampling system.

Fig. 2. Gas-analysis separation scheme.


Surface run (hose only)
Hose chute
Surface run of high-pressure hose with wire rope inside
To sampling skid
Adapter section
Hydroseal coating through gas-seal stemming plug
32-mm-i.d. high-pressure hose with 25-mm-diam. wire rope inside (typical)
Vacuum demand valve
Foam wrap (typical at each stemming plug)
Pressure relief valve (120 psi)
3 sealed set screws at each hose coupling
End of hose (1.25 cavity radii from working point)
Bare wire rope continuous to bottom of vacuum demand valve
Working point

Smith - Fig. 1


Carrier Ar, Kr, Xe
Sample
Charcoal
-130°C
Charcoal
-130°C
H_2
-36°C
MS-5A
-78°C
Ar
O_2
0°C
N_2
100°C
CO
20°C
MS-5A
-36°C
Kr
CH_4
CO_2
100°C
MS-5A
100°C
Xe
C_2H_6
350°C
MS-5A
100°C
Rn-C_2H_6
350°C
C_nH_{2n+2}
Carrier C_2H_6

Smith Fig. 2