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TITLE APPARATUS FOR THE DYNAMIC AND TOTAL MEASUREMENT OF RETAINED FISSION GAS

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ABSTRACT

This versatile apparatus provides a quick, accurate and inexpensive determination of fission gases Kr and Xe in irradiated nuclear fuels. Samples are heated to 2000°C in a vacuum furnace under controlled temperature-time conditions and the released Kr and Xe are dynamically and integrally measured by a quadrupole mass spectrometer.

INTRODUCTION

Measurement of the quantities of fission gas as Kr and Xe retained in irradiated, experimental Fast Breeder Reactor (FBR) fuels is essential to establishing their irradiation stability. To now, the measurement technique has been mainly acid dissolution in a closed system with added amounts of enriched Kr and Xe (^{82}Kr and ^{130}Xe), nitrogen sweep trapping of the gases on a cryogenic charcoal absorbent, heating of the trap to release the gases to a chromatographic column, collection of the separated Kr and Xe and their measurement by a mass spectrometer. This technique, although highly reliable is expensive. The apparatus described[1] in this paper has been developed to provide a lower cost measurement. It also uniquely provides a dynamic measurement of the Kr and Xe released under controllable temperature and time conditions to better assess the gas retention properties of the fuel. The apparatus is located in the Bldg 9 hot cell Facility in the Chemistry and Metallurgy Building at the Los Alamos National Laboratory. A prototype of this apparatus has been described previously.[2]

DESCRIPTION OF APPARATUS

Major components are 1, a quartz furnace with vacuum locks for introduction and removal of samples, 2, a Uthe International Model 1000 quadrupole mass spectrometer, 3, a Digital Equipment Corporation PDP 11/20 minicomputer data acquisition and mass spectrometer control system, 4, a Datalink temperature time program set to control the furnace induction heater, and 5, a Leybold Heraeus Model PHP 150 turbomolecular pump. All components, except the furnace are located outside the hot-cell. Fig. 1 is a photograph of the hot-cell installation.

The quartz-tube furnace, shown in Fig. 2 features, 1, a small internal volume of 250 cm^3, 2, an upper operating temperature of 2000°C (up to 2200°C for limited periods), 3, a maximum heating rate of 3000°C/min, 4, a vertical orientation for gravity introduction of samples through vacuum locks, 5, a continuously maintained vacuum of 1×10^{-7} Torr (leak condition), 6, high sample throughput rate, 7, acceptance of intact and powder samples having volumes up to 1 cm^3, and 8, total recovery of heated sample.

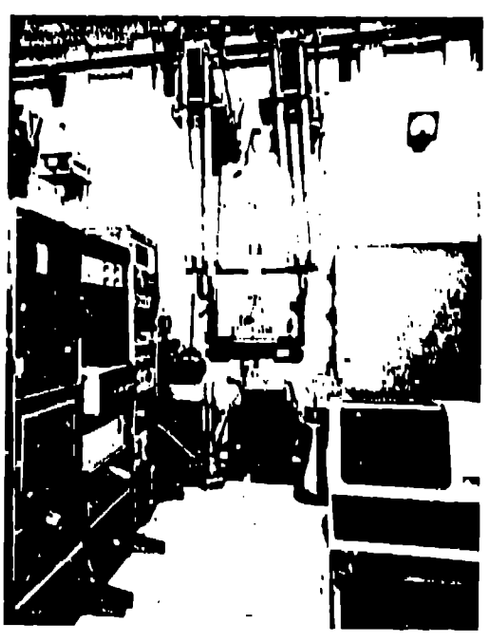

Fig. 1. Overall view of the dynamic fission gas release measurement system.

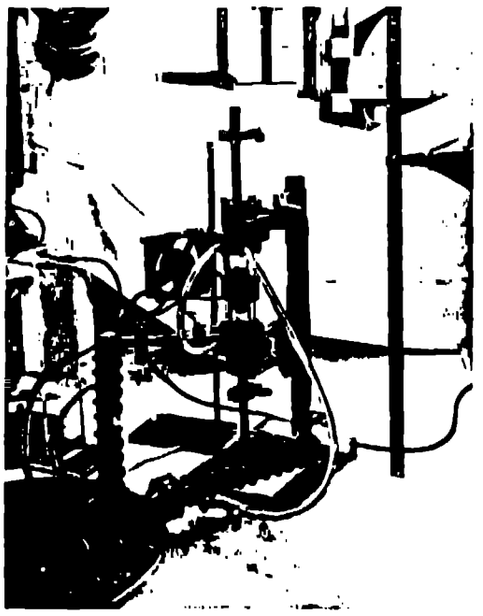

Fig. 2. Hot-cell installed, induction heated furnace.

The furnace is a 25.4-cm-long, 1.8-cm-diam quartz tube, sealed at both ends to water-cooled stainless steel flanges by conical O-ring seals. A six-turn R.F. coil (0.8-cm-diam copper tubing) is wound around the tube at mid length. The furnace is air cooled, rather than water cooled, so that radioactive material is not transported outside of the cell in case of a furnace rupture. An internal 2.5-cm-diam quartz tube shields the outer tube from thermal radiation and metal vapor deposition. This internal tube thermally contacts the lower furnace flange. The furnace connects to the quadrupole mass spectrometer with 1.5 m of 1.91-cm-diam vacuum piping as shown in Fig. 1.

A 15.2 cm-long, 1.1 cm-diam tubular tantalum crucible with a tungsten liner is located at the furnace center. The crucible with sample is dropped into and out of the furnace upon actuating the vacuum lock valves.

The vacuum locks are 1.9 cm butterfly valves sealed to the upper and lower furnace flanges by o-rings. The vacuum locks are sequentially pumped by the vacuum system shown in Fig. 1. KF-quick disconnect flanges (Varian Vacuum Products) seal all vacuum connections except vacuum piping external to the hot cell. External vacuum connections are

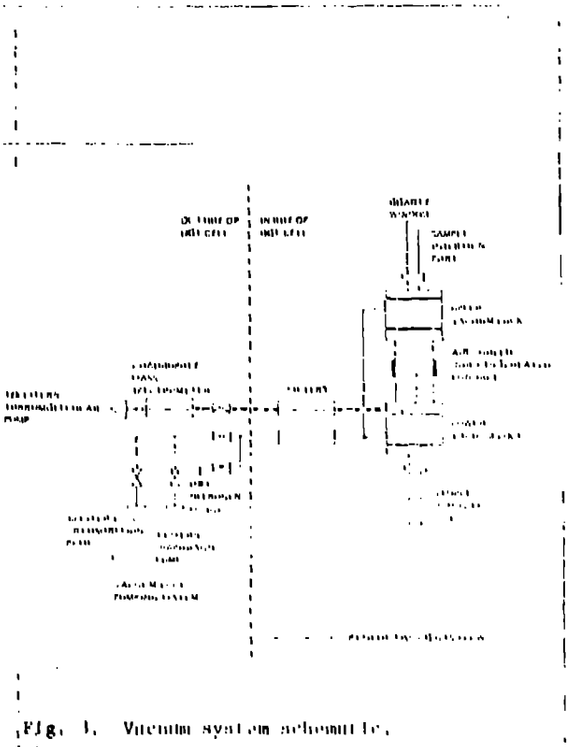

Fig. 1. Vacuum system schematic.

sealed by knife metal gasket flanges. One of KF flanges proved very successful with respect to ease of operation and integrity of seal. Most KF flanges used are also KF 16 (1.9-cm-diam). The clamps used to seal the KF flanges were modified to keep the clamps in the closed position using music wire springs. This simplified their use by requiring only a single manipulator hand for installation.

Two high-flow, 2 μ sintered stainless steel filters in the high vacuum lines prevent transport of radioactive particles from the hot cell (see Fig. 1). These filters are water cooled to contain volatile fission products other than Kr and Xe.

All components are made of low out gassing material. Most furnace components and vacuum piping are type 304 stainless steel. All high-vacuum piping is bakeable at 250°C.

A Digital Equipment Corporation PDP 11/34 minicomputer system controls the quadrupole mass spectrometer, acquires on line gas release data, and displays results. This real time system has a central processor with a 28K memory, a dual RF-11 hard disk drive system capable of storing 2.4 X 10^6 words, a fast 12 bit AD 11 K analog to digital converter for peak height acquisition, a 12 bit AA 11 K D/A converter and DR 11 A I/O interface to control the quadrupole mass spectrometer, a VT 100 terminal, a KW 11 P programmable

clock for event timing and a IDS-460 line printer. The on-line software has a timed task-swapping routine inwhich data acquisition and mass spectrometer control are interrupt driven. Data reduction, storage and display are concurrent with data acquisition and mass spectrometer control.

OPERATION OF APPARATUS

Apparatus operates in a sequential and cyclic manner. The general sequence is

a. Load a tungsten-tantalum crucible, containing the sample to be analyzed, into the upper vacuum lock.

b. Pump-down vacuum locks to < 5×10^{-5} Torr.

c. Transfer the crucible to the furnace by opening and then closing upper vacuum lock valve.

d. Program heat sample and measure release of Kr and Xe. Display and store analytical results.

e. Eject crucible from furnace after conclusion of sample heating by opening and then closing lower vacuum lock valve.

f. Vent vacuum locks to atmospheric pressure on dry N_2 and recover analyzed sample.

g. Repeat procedure starting at step (a) for next sample.

The time required to pump-down vacuum locks to < 5×10^{-5} is typically 10 - 15 minutes. At < 5×10^{-5}, opening the vacuum locks to the furnace does not significantly affect furnace pressure.

DATA RECORDING AND TREATMENT

The mass spectrometer is programmed to scan and measure release rates of Kr and Xe gases usually at 1 - 10 s intervals. The usual isotopes measured are ^{83}Kr, ^{84}Kr, ^{85}Kr, ^{86}Kr, ^{131}Xe, ^{132}Xe, ^{134}Xe, and ^{136}Xe. The gain of the mass spectrometer is automatically adjusted by the real-time software to give maximum sensitivity for each isotope peak height measurement. Maximum scan rate is 10 amu/s, set by the response speed of the ion detector electrometer. After completion of a scan, the dynamic release rates in Torr-L/S units and the integrated volumes in STP liter units are computed and printed for each gas isotope. At the completion of an analysis, values for the total volume and the isotopic composition for Kr and Xe are printed in tabular form. A plot of the release rate for each isotope versus time and/or temperature also is produced.

INSTRUMENT CALIBRATION

Flow sensitivity factors are established by flowing research grade natural Kr and Xe into the mass spectrometer through a molecular leak. Stability of the sensitivity factors typically is less than 5.0% over 48 h operation.

APPLICATIONS

Total Kr and Xe volumes of $1.0 \times 10^{-[illegible]}$ cm^3 have been measured with about 3% relative reliability. This provides adequate sensitivity and reliability to determine 10% retained Kr and Xe in a 1-g sample of FBR mixed uranium-plutonium oxide fuel having 1% burnup. The uncertainty in the measurement of isotopic composition, based on tests with natural Kr and Xe is about 1.0% for the major isotopes.

SUMMARY

This fission gas measurement apparatus provides a quick, accurate and inexpensive determination both of total and dynamic Kr and Xe released from nuclear fuel samples heated under controlled temperature-time conditions. The obtained data is essential to establishing the irradiation stability of experimental [illegible] fuels.

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