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IFA-430 FISSION PRODUCT MEASUREMENT SYSTEM

DESCRIPTION AND SCOPING TEST RESULTS

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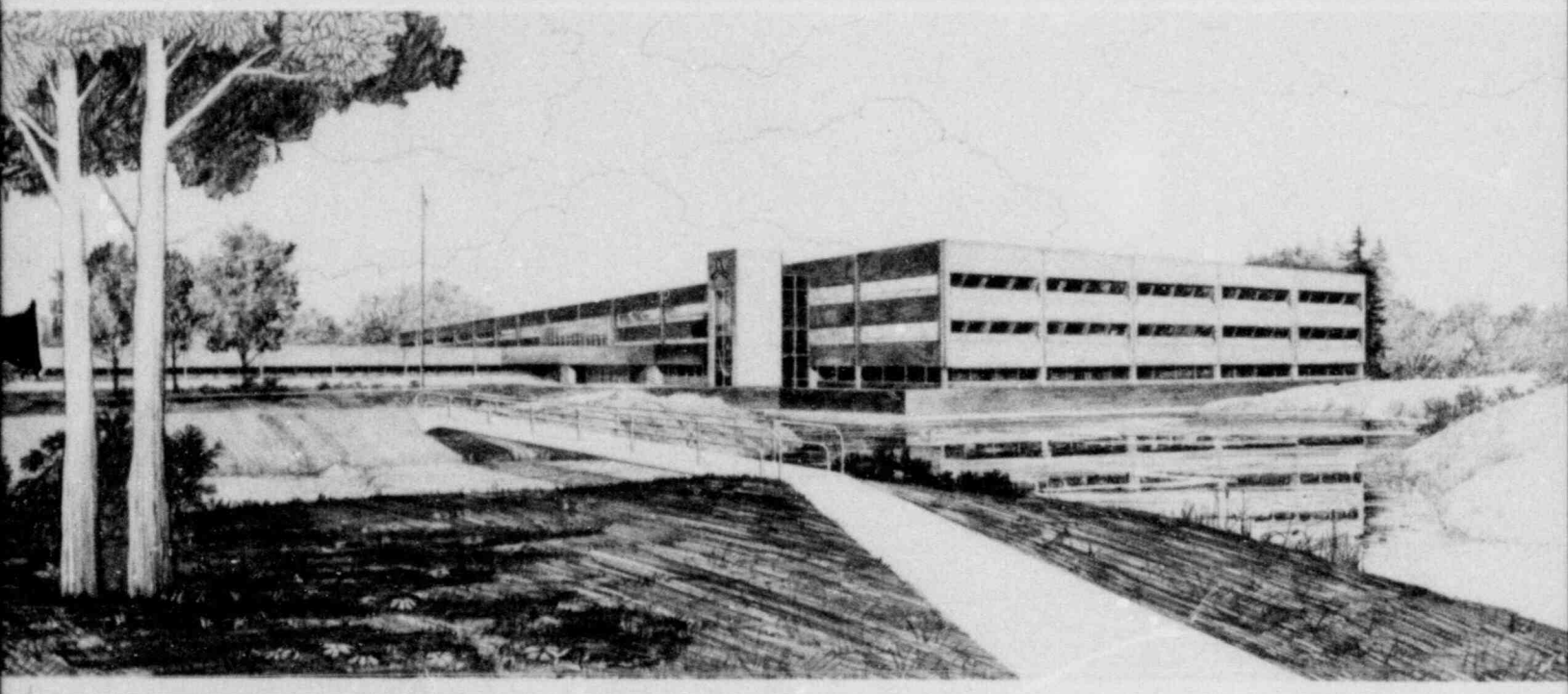
U.S. Department of Energy
Idaho Operations Office • Idaho National Engineering Laboratory



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IFA-430 FISSION PRODUCT MEASUREMENT SYSTEM DESCRIPTION AND SCOPING TEST RESULTS

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December 1979

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ABSTRACT

A fission product measurement system has been installed at the Halden Reactor in Norway to measure the release of Xe, Kr and I fission products from typical LWR fuel pellets during irradiation. A description of the system and the results of two scoping tests are presented. The scoping tests were designed to establish the operating limits and optimize the experiment parameter setpoints. Measured release rates of Kr, Xe and I are presented along with a discussion of the results.

SUMMARY

The United States Nuclear Regulatory Commission's Reacator Safety Research Program is studying fuel rod response for a wide range of operating conditions that could occur during the life of a light water reactor (LWR) fuel rod. As a part of this program, long-term irradiations of instrumented fuel assemblies at the Halden Heavy Boiling Water Reactor (HBWR) in Halden, Norway are being managed by EG&G Idaho for the Nuclear Regulatory Commission. One of these instrumented fuel assemblies, IFA-430, is being used, in part, to study fission gas release. The purpose of this report is to provide a description of the fission product measurement system and to report the results of two scoping tests.

The IFA-430 assembly has the unique capability to provide data to determine the release of fission gases from the fuel to the fuel-cladding gap in a fuel rod of typical light water reactor design during nuclear operation. The assembly contains four fuel rods, two of which are connected to an external gas supply system which allows the fission gases to be swept out of the rods and to an on-line gamma spectrometer where the gas stream is analyzed for Xe and Kr fission gases.

Two scoping tests have been performed to determine the operational limits and optimum parameter set points for fission gas and iodine release measurements. The results of these tests indicate that the system can provide data to characterize the release of Xe and Kr fission gases as well as the release of ^{135}I.

Average fuel surface temperatures in the two gas flow fuel rods were less than 785 K during the scoping test fission gas release measurements. The measured release rates of Xe from the IFA-430 fuel, when compared with release rates expected for recoil and diffusion release, indicate that the dominating release mechanism is recoil at

these fuel temperatures. Kr, however, shows no conclusive trend; this is suggested to be due to precursor effects.

The release-to-birth ratios of Xe and Kr compare well with results of previous work when the data are normalized to the surface-to-volume ratio of the fuel.

The release rate of iodine is being studied by EG&G using the technique in which the daughter product of ^{135}I, ^{135m}Xe, is measured following reactor scram to determine the iodine plateout concentration prior to scram. Results to date indicate that the release rate of ^{135}I for fuel temperatures <785 K is of the same order of magnitude as those of ^{135m}Xe and ^{138}Xe.

CONTENTS

FIGURES

TABLES

IFA-430 FISSION PRODUCT MEASUREMENT SYSTEM DESCRIPTION AND SCOPING TEST RESULTS

I. INTRODUCTION

A major objective of the United States Nuclear Regulatory Commission's Reactor Safety Research Program[1] is to establish analysis codes capable of predicting commercial nuclear fuel performance for a wide range of operating conditions which could occur during the life of a light water reactor fuel rod. An extensive fuel behavior research program has been implemented in which in-pile and out-of-pile experiments are conducted to study fuel rod responses to normal and abnormal operating conditions. The data obtained from these experiments are used in the development and assessment of reactor safety analysis codes. Included in this program are studies of uranium oxide and mixed oxide fuel behavior performed at the Heavy Boiling Water Reactor (HBWR) in Halden, Norway.

The HBWR was built by the Norwegian Institute for Atomenergi, and has been operated by the Organization for Economic Cooperation and Development (OECD) Halden Reactor Project through an international agreement between participating governments and organizations. One of the assemblies being irradiated in the HBWR is the Instrumented Fuel Assembly-430 (IFA-430) managed by EG&G Idaho, Inc., for the Nuclear Regulatory Commission.

The purpose of this report is to provide a description of the IFA-430 fission product measurement system (FPMS) and to report the results of scoping tests performed to establish the operating limits and optimum the experiment parameter setpoints for the FPMS.

IFA-430 was originally designed to study UO_2 fuel thermal performance and gas flow resistance within a fuel rod, and has been modified to include the study of fission gas release. The decision to add a fission product measurement system to the IFA-430 experiment was prompted by the unique prospect of measuring the release of fission

gases from fuel to the fuel-cladding gap in a fuel rod of typical light water reactor design during actual nuclear operation. The data are to be used to calculate release-to-birth (R/B) ratios of Xe and Kr isotopes, assess fission product release codes, and characterize iodine release from the fuel.

In Section II the IFA-430 assembly and fission product measurement system are described. Section III contains the objectives and results of the scoping tests. A discussion of the R/B ratios and fission gas release mechanisms is presented in Section IV and Section V contains the conclusions.

II. SYSTEM DESCRIPTION

The fission gas release experiments are performed using the IFA-430 assembly and fission product measurement system. A detailed description of the design and operation of the IFA-430 assembly is provided in Reference 2; a summary is provided in this section along with a description of the FPMS.

1. IFA-430 Irradiation Assembly

IFA-430 contains four, 1.28-m-long fuel rods containing 10% enriched UO_2 fuel pellets. Two of the rods in the assembly are termed gas flow rods. These two rods each have a fuel centerline thermocouple and three axially spaced pressure sensors and are connected to a gas flow system, shown in Figure 1, which is used during fission gas release experiments to sweep the fission gases out of the fuel rods and to the FPMS. A schematic of the gas flow rods is shown in Figure 2. The other two rods, termed thermal performance rods, are each equipped with two centerline thermocouples and three off-center thermocouples and are pressurized to 0.48 MPa with helium. Table 1 presents the IFA-430 fuel rod instrumentation and design variables; Figure 3 shows the assembly instrumentation.

The gas flow system is capable of providing a wide range of steady state flow rates (0.5 to 50 liters per minute). Helium is presently used during the fission gas release tests; however, future plans include the use of He/Ar or He/Xe mixtures. The gas flow is a once-through process. Gas introduced from regulated high pressure cylinders passes through one or both fuel rods to the FPMS detector station and is then collected in storage cylinders or passed immediatley to the reactor off-gas handling system.

The linear heat rate for each of the four rods in the IFA-430 assembly is determined from axial and azimuthal self powered neutron detector (SPND) signals. To relate the neutron detector signals directly to the linear heat rate, measurement of the absolute thermal

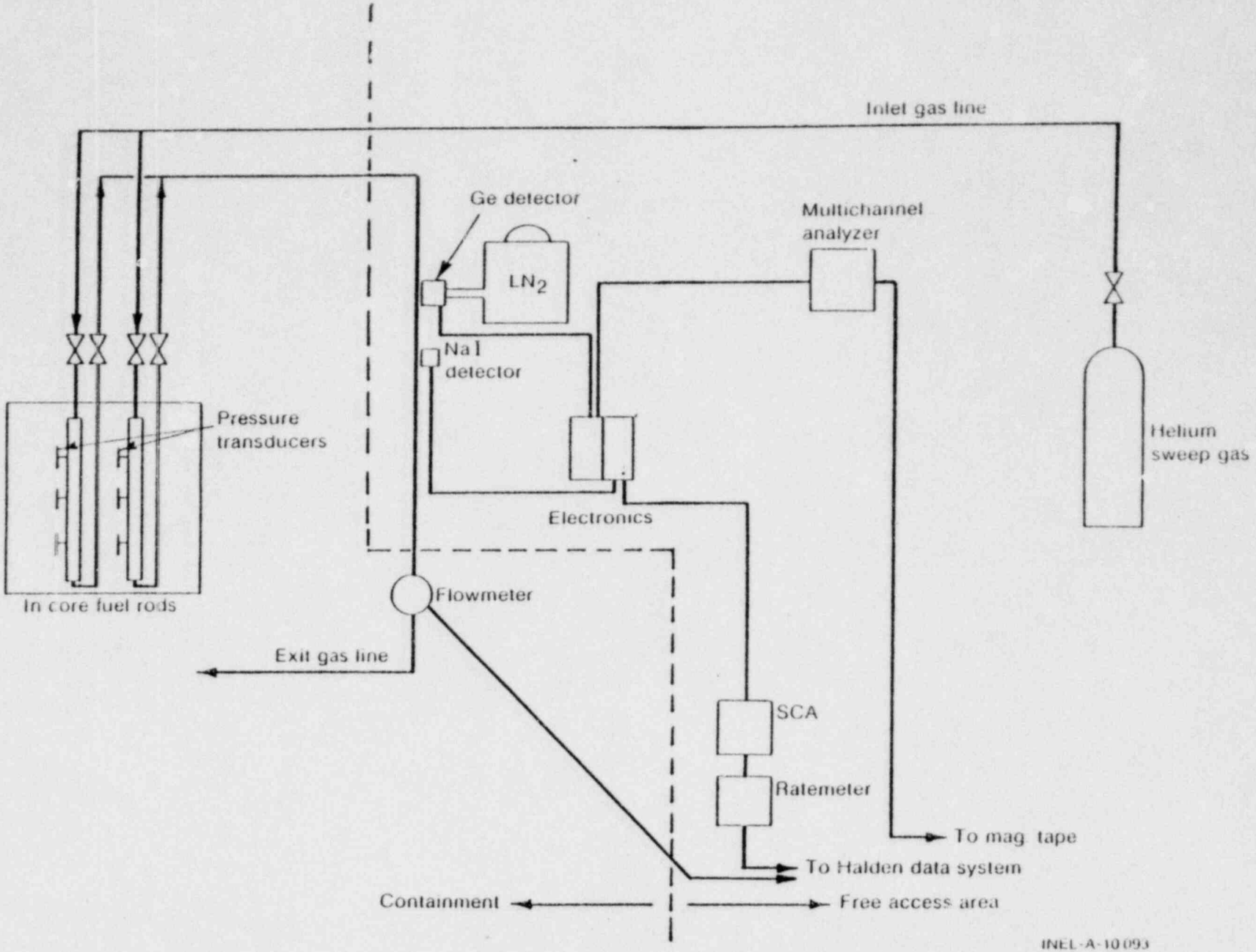

Figure 1 IFA-430 gas flow system simplified piping and instrumentation diagram.

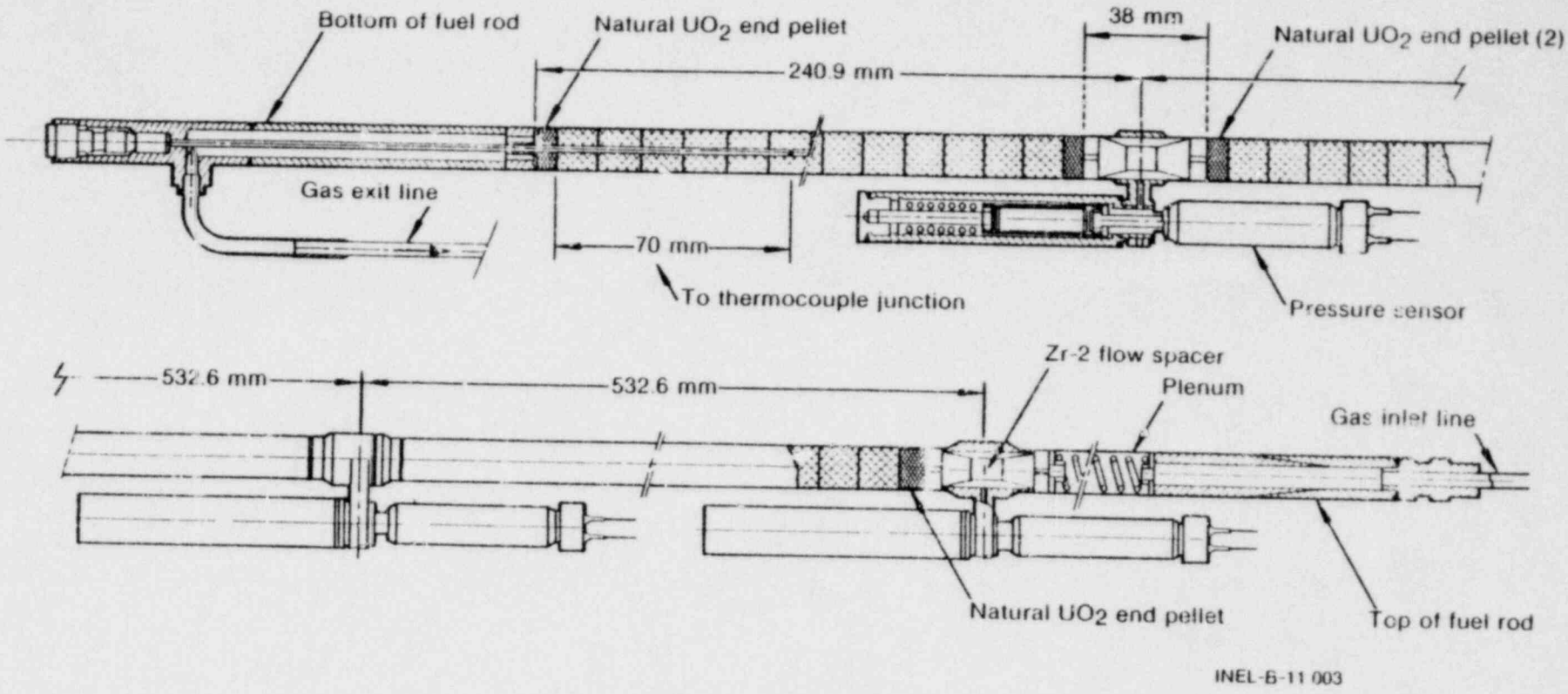

Figure 2 Location of fuel centerline thermocouple and rod pressure sensors on gas flow rods.

TABLE 1. IFA-430 FUEL ROD INSTRUMENTATION AND DESIGN VARIABLES

Rod	Experiment	Diametral Gap	Fill Gas	Instrumentation
1	Fuel temperature	0.229 mm	0.48 MPa He	2 centerline thermocouples, 3 off-center thermocouples
2	Gas flow	0.229 mm	variable	centerline thermocouple, 3 pressure sensors
3	Fuel temperature	0.102 mm	0.48 MPa He	2 centerline thermocouples, 3 off-center thermocouples
4	Gas flow	0.102 mm	variable	centerline thermocouple, 3 pressure sensors

Fuel--Form - pressed and sintered UO_2 pellets
Enrichment - 10 weight percent U-235
Density - 10.412 g/cc (95% of theoretical)
Shape - length, 12.7 mm
- diameter, 10.808 mm, 10.681 mm
- ends, champhered 45° to an approximate depth of 0.12 mm

Cladding--zircaloy-2
OD - 12.79 mm
ID - 10.91 mm

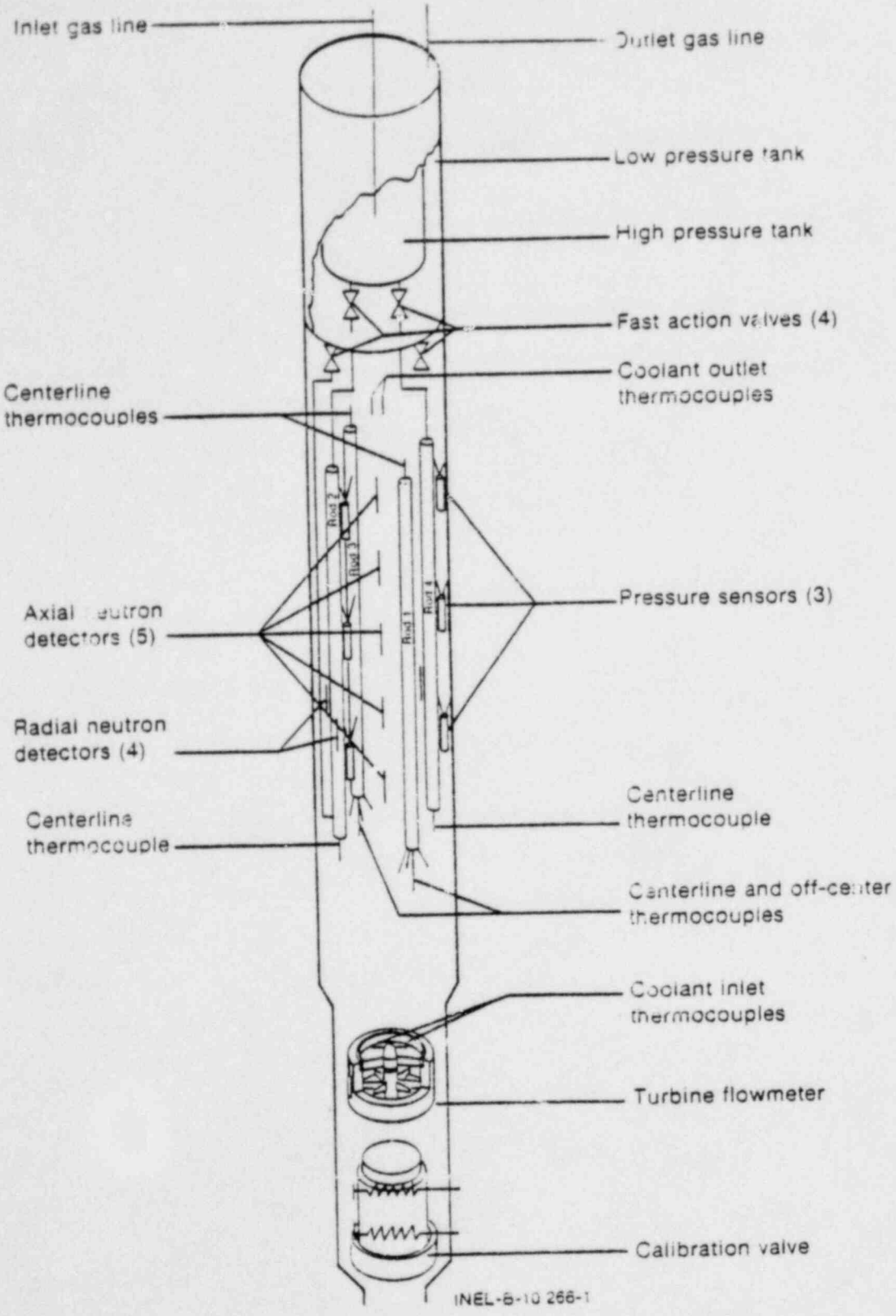

Figure 3 IFA-430 assembly in core instrumentation.

power of the assembly was required. This measurement was accomplished by a calorimetric technique in which subcooled water was introduced into the assembly and the flow rate and temperature rise across the assembly measured. The resulting data permitted calculation of the assembly total power. Heat gain through the walls of the assembly and gamma heating contributions were estimated by analogy to measurements with previous test assemblies.

An assembly power calibration factor was generated which relates the assembly power in kilowatts to the arithmetic average of the neutron detector signals in nanoamps. The five axially spaced and four azimuthally located SPNDs are used to define the axial flux shape, the flux tilt across the assembly, and, in conjunction with the power calibration factor, the rod linear heat rate. To compensate for the slow response time of vanadium the SPND signals are corrected before powers are calculated.

A Fourier sine series is generated using the five corrected axial neutron detector signals to define the axial flux shape and the four corrected azimuthal neutron detector signals are used to determine the tilt of the flux across the assembly. A calibration factor is generated by correlating the measured assembly power to the average integrated flux calculated with the Fourier sine series. This permits calculation of rod linear heat rate for any axial location in the fuel rod, individual sections of the fuel rod and for the rod as a whole. The linear heat rate is used to determine the fission rate and, thus, the isotope production rate.

2. Fission Product Measurement System

The fission product measurement system was designed to measure the quantitative release of fission gases from the fuel to the fuel-cladding gap. Fission gases are swept from the fuel-cladding gap with a carrier gas. As shown in Figure 1, the carrier gas sample stream is routed to the FPMS detector station where the fission gases contained in the gas stream are measured as they flow by the

detectors. At the detector station, located outside the reactor containment, the sample stream piping (6-mm-inside-diameter stainless steel) is enclosed in a lead shield, and viewed by two detectors through collimated apertures.

A schematic of the FPMS instrumentation is shown in Figure 4. Quantitative determination of the fission gases in the sample stream is made with a gamma ray spectrometer consisting of a hyper-pure Ge detector and microcomputer-based data acquisition and reduction system. The Ge detector is a 30-cm^3, 5% efficient, closed-end coax. The detector high voltage supply and preamp are located at the detector station. The preamp drives the signal 100 m to the amplifier, which is located at the data acquisition center. A 50-Hz filter is used between the preamp and the amplifier to reduce noise on the signal lines. The amplified signal is fed into a commercial analog-to-digital convertor and multichannel analyzer (MCA).

Gamma ray spectra generated by the MCA are stored on magnetic disk and tape. The magnetic tape are mailed to the Idaho National Engineering Laboratory and the spectra are analyzed on a large computer using state-of-the-art spectral analysis techniques. To ensure that data collected during experiments are suitable for analysis and to track the behavior of the experiments as they progress, the data stored on disk are analyzed using the data acquisition system microcomputer throughout the experiments.

Data for determining the quantitative release rate of Kr and Xe isotopes and, under special conditions, the release of ^{135}I from the fuel come from the gamma ray spectra. Because conventional gamma ray spectrometry necessitates measuring a sample for relatively long periods of time (many minutes) and then determining the average value during the measurement period, it is important that the sample flow rate be as stable as possible. To monitor the stability of the total fission gas concentration during the acquisition of each spectra, a gross gamma system is included in the FPMS.

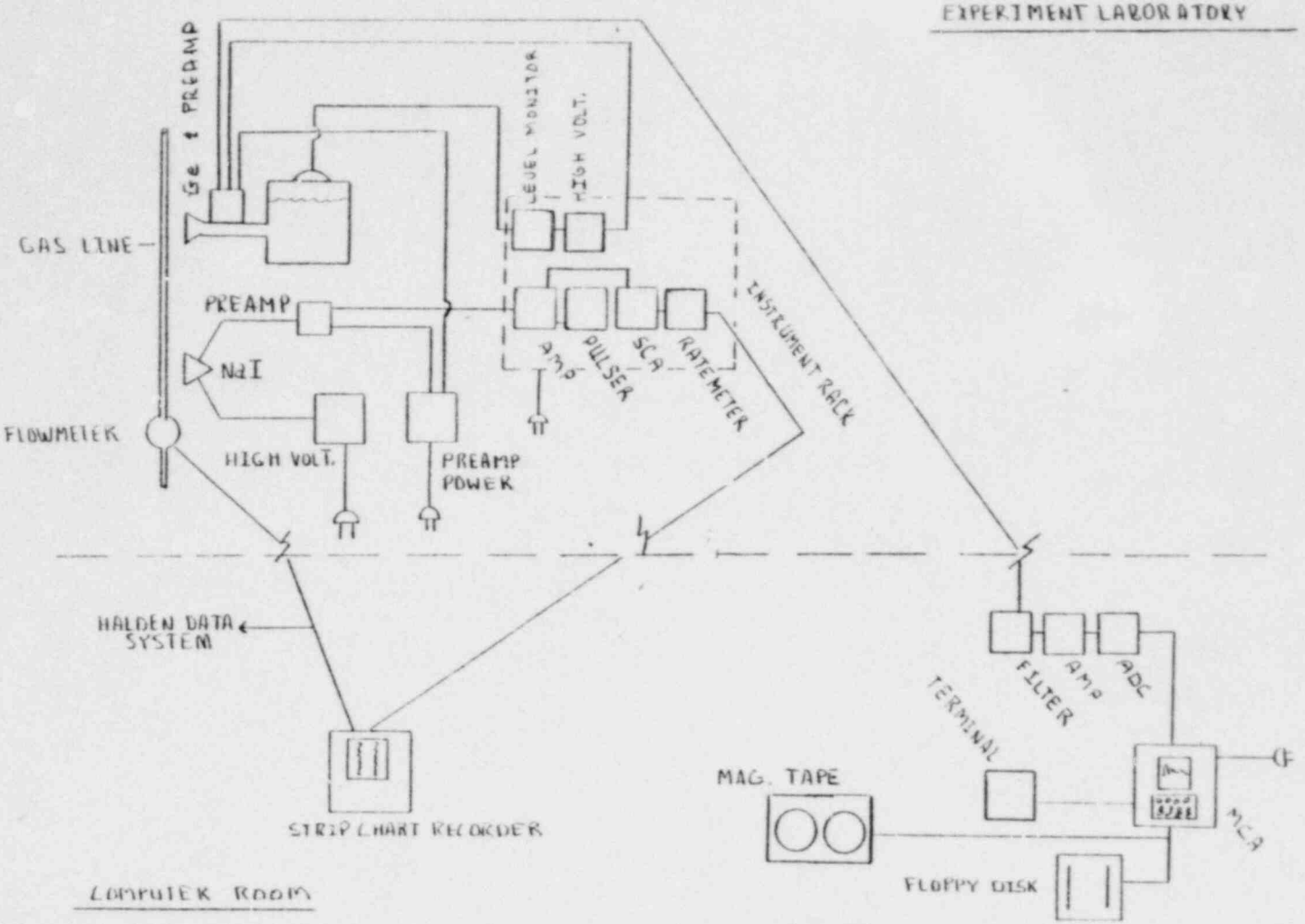

Figure 4 Fission Product Measurement System Instrumentation.

The gross gamma detection system is based on a 2.5 x 3.8-cm NaI crystal which provides a continuous gamma ray count rate. Because of sensitivity to changes in temperature, and the inherent broad resolution of the NaI detector, this system does not provide quantitative information. It does provide data to determine any qualitative changes in the total fission gas release rate. The output of the NaI system along with the sample stream flow rate is displayed and recorded on strip chart at the FPMS data acquisition station. This display allows the experiment operator to continuously monitor the stability of the experiment during each spectral acquisition period.

Parameters important to the measurement of fission gas release include fuel rod power and temperature. The fuel centerline temperature and data for determining the fuel rod power are recorded on one of three Halden reactor data acquisition systems at sample rates ranging from five samples per second to one every 15 minutes, depending upon experiment requirements.

To determine the quantitative isotopic release rate of fission gases from the spectra collected during the experiments, each spectrum must be analyzed. Sophisticated computer codes have been developed[3] for automated analysis of gamma ray spectra; these codes use special fitting routines and background subtraction to determine the areas of the peaks in the spectra from which the release rates are determined. Under optimum conditions the error in the measured release rate is $\pm$20%.

III. SCOPING TEST OBJECTIVES AND RESULTS

The fission product measurement system was installed and two scoping tests performed in August 1979. Scoping Test 1 was performed during nuclear operation and Scoping Test 2 following reactor shutdown. The objectives and results of the tests are presented in this section.

1. Tests Objectives

Scoping Test 1 was designed to determine the operating limits and optimum parameter setpoints for fission gas measurement during nuclear operation of IFA-430. The objectives were to:

1. Determine the fission gas isotopes of interest that are measurable with the system in an on-line mode

2. Determine count rate dependence on flow rate at full power and optimum collimation for full power operation

3. Determine the delay time of the fission gases in traveling from the fuel rods to the detectors

4. Determine any flow-rate-induced effects on volatile fission product measurements and check for consistency in fission gas measurements as a function of flow rate

5. Establish the ability to use He/Xe sweep gas mixtures during fission gas measurement tests.

Scoping Test 2 was performed to evaluate the feasibility of measuring the iodine-135 that plates out in the fuel rod and piping system and of using this measurement to estimate the ^{135}I release rate from the fuel during steady state nuclear operation.

2. Results

Objective 1 was to determine the isotopes of Kr and Xe that are measurable with the FPMS. The measurability of any particular isotope is tied to the energy of the gamma rays emitted from the isotope during decay, the decay rate of the isotope, the delay time from the fuel rod to the detector station, the release rate of the isotope from the fuel and the concentration of other isotopes in the sample being measured. Table 2 presents the isotopes of Xe and Kr which were found to be measurable, in a continuous mode, with the FPMS. The short half-life Xe and Kr isotopes were measurable; however. the long lived ^{85}Kr and ^{133}Xe could not be detected.

Of the isotopes listed in Table 2 ^{135}Xe and ^{138}Xe and ^{85m}Kr, ^{87}Kr and ^{88}Kr have all been used by other researchers to characterize fission gas release from UO_2 and thus will be comparable with the results of the IFA-430 tests. The direct measurement of the short half-lived isotopes ^{139}Xe, ^{140}Xe, ^{89}Kr, and ^{90}Kr has not been possible in previous experiments and application of these data to overall fission gas release characterization will have to be developed.

Determining the dependence of the count rate (gamma's counted per second) on the flow rate of the sweep gas and the optimum collimation for full power operation was the second objective of Scoping Test 1. The count rate dependence on flow rate is important because the accuracy of the measurement system is dependent on the total count rate. In addition, the count rate for each isotope at various flow rates can be used as a measure of the consistency of the measurement system. To minimize the error in measurement, the total count rate should be kept to less than 10,000 counts per second, corresponding to a dead time of <20%. Figure 5 shows the total system count rate as a function of the flow rate when using a 30-cm length of pipe as the sample volume. The count rates for the 30-cm pipe section geometry are unacceptably high, and the trend indicates that flow rates of less than a few tenths of a liter per minute would be

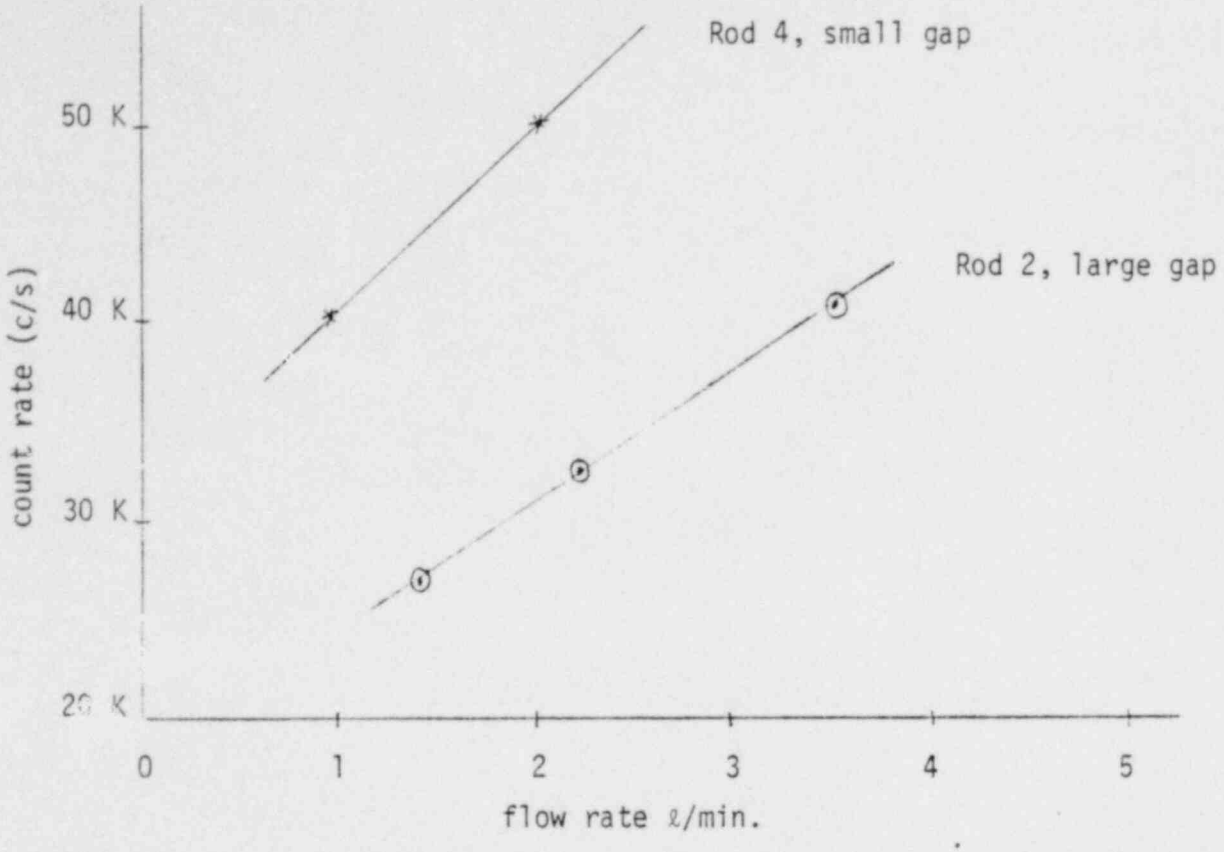

Figure 5 Processed count rate of Ge spectrometer as a function of flow rate at full power for a 30 cm sample pipe geometry.

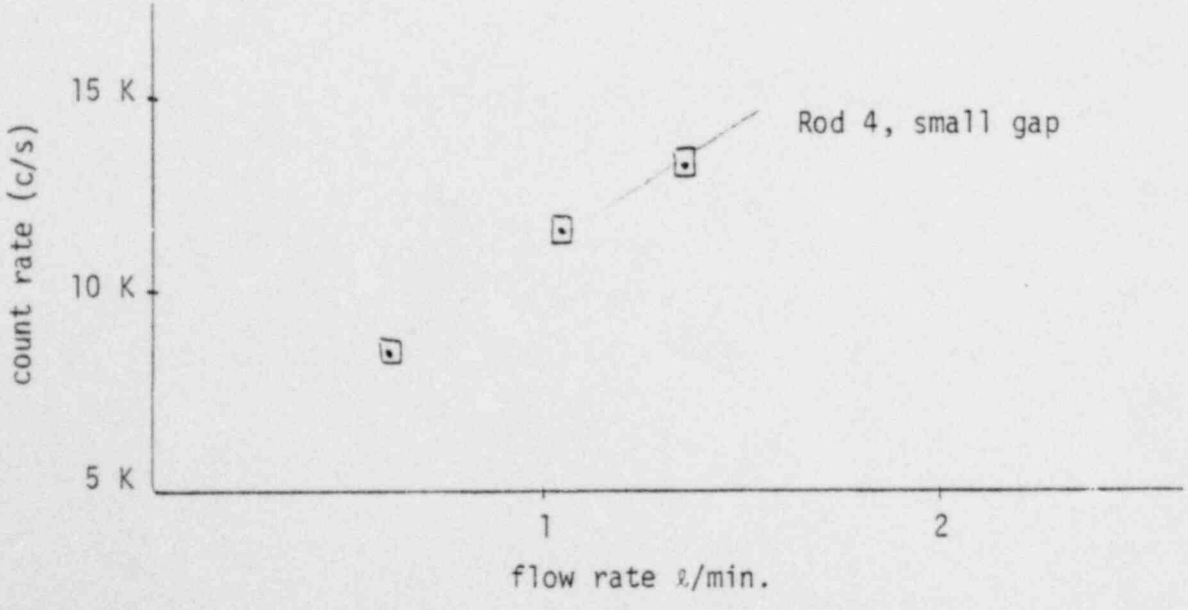

Figure 6 Processed count rate of Ge spectrometer as a function of flow rate at full power for a 3 cm sample pipe geometry.

TABLE 2. ISOTOPES OF Kr AND Xe MEASURABLE WITH IFA-430 FPMS IN CONTINUOUS MODE

Xe	$T_{1/2}$	Kr	$T_{1/2}$
135	9.09 h	85m	4.48 h
135m	15.6 m	87	76.3 m
137	3.84 m	88	2.84 h
138	14.13 m	89	3.16 m
139	40.4 s	90	32.3 s
140	13.6 s	91	8.57 s

required to bring the count rate into the acceptable region. The current flowmetering equipment has a lower bound of ∿0.5 liters per minute thus the 30-cm pipe geometry is considered unacceptable. The results of count rate versus flow rate measurements for a 3-cm pipe geometry are shown in Figure 6, they indicate that this geometry is acceptable for flow rates of ∿1 liter per minute or less. As shown in Figure 5, the count rate when the gas is swept through Rod 2 is significantly less than when Rod 4 is used. This implies that, using the 3-cm geometry, flow rate could be as high as 2 liters per minute when sweeping Rod 2 and still result in acceptable count rates.

The optimum collimation was inherently determined by the results of the flow rate versus count rate tests. A 3-cm length of pipe viewed from 10 cm away provides an acceptable count rate in a flow rate region where the flow measurement error is small. The spectral acquisition time necessary to provide adequate statistics for gamma spectra was 15 min at these conditions (∿1 liter per minute and a 3-cm geometry); this is an acceptable acquisition time for steady state testing.

Objective 3 of Scoping Test 1 was to determine the delay time from the fuel rod to the detection station. Although this measurement would appear to be simple to perform the actual system design and dynamics make it difficult. Figure 7 shows the gas flow system piping schematic; the delay time was determined by measuring the delay time from the exit gas line tee just upstream of the iodine filter to the

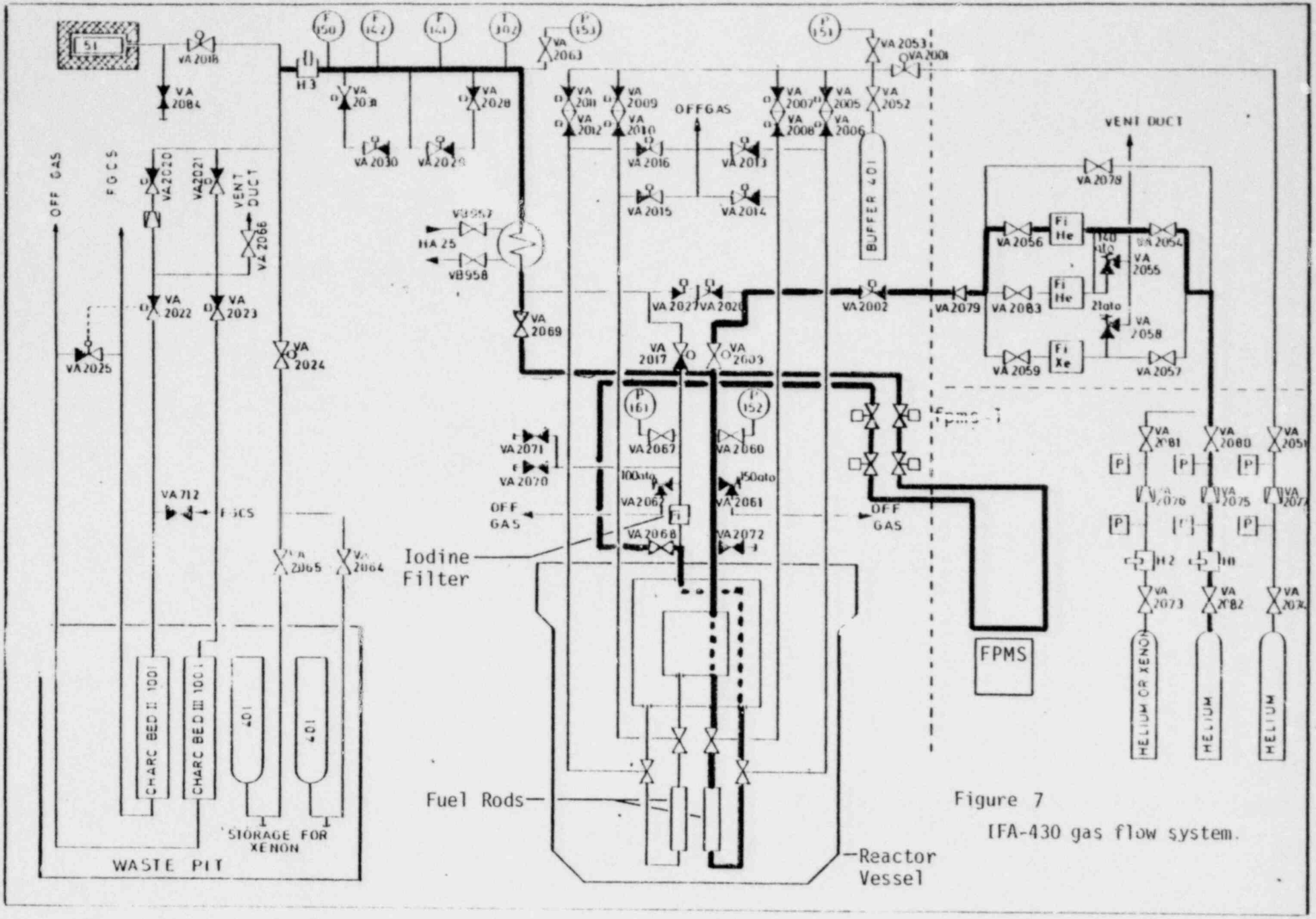

Figure 7

IFA-430 gas flow system.

detector station and calculating the delay time from the fuel rod to the tee using the piping volumes. The measurement of the delay from the tee to the detector station was accomplished by establishing a steady flow rate through the system (Figure 7) with Valve 2017 open and Valve FPMS-1 closed and then simultaneously closing 2017 while opening FPMS-1 and measuring the time for fission products to reach the FPMS detector. The delay time at a flow rate of 0.4 ℓ/min was 144 s which implies the piping volume is 0.96ℓ from the tee to the FPMS detector. The volume of the piping from the bottom of the fuel rod to the tee is 1.13ℓ thus the total system volume between the bottom of the fuel rod and the detector is 2.09 ℓ. This results in a delay time

$$t_d = 2.09\ f^{-1} \qquad (1)$$

where

t_d = delay time (min)
f = flow rate (ℓ/min) at STP.

Since flow rate must be restricted to the region $0.5 \geq f \leq 2.0$, the delay times will range from ∿250 to ∿60 s; Figure 8 is a plot of the delay time for this flow region.

The fourth objective of Scoping Test 1 was to determine any flow-rate-induced effects on volatile fission product measurements and check for consistency in noble gas measurements as a function of flow rate. Cesium was measured but appeared to orginate only as a daughter of the Xe that was transported to the detector station. Since no volatile fission products were detected at the sample station (that were being transported to the detector station as volatiles) there were no observable effects of flow rate on volatile fission product transport in the system. The effect of flow rate on noble gas release rate measurement is in question because of an apparent discontinuity in the release rate versus flow rate data. As the release rate is a main parameter of interest, the method of determining the release rate from the data is presented here; a discussion of the data follows.

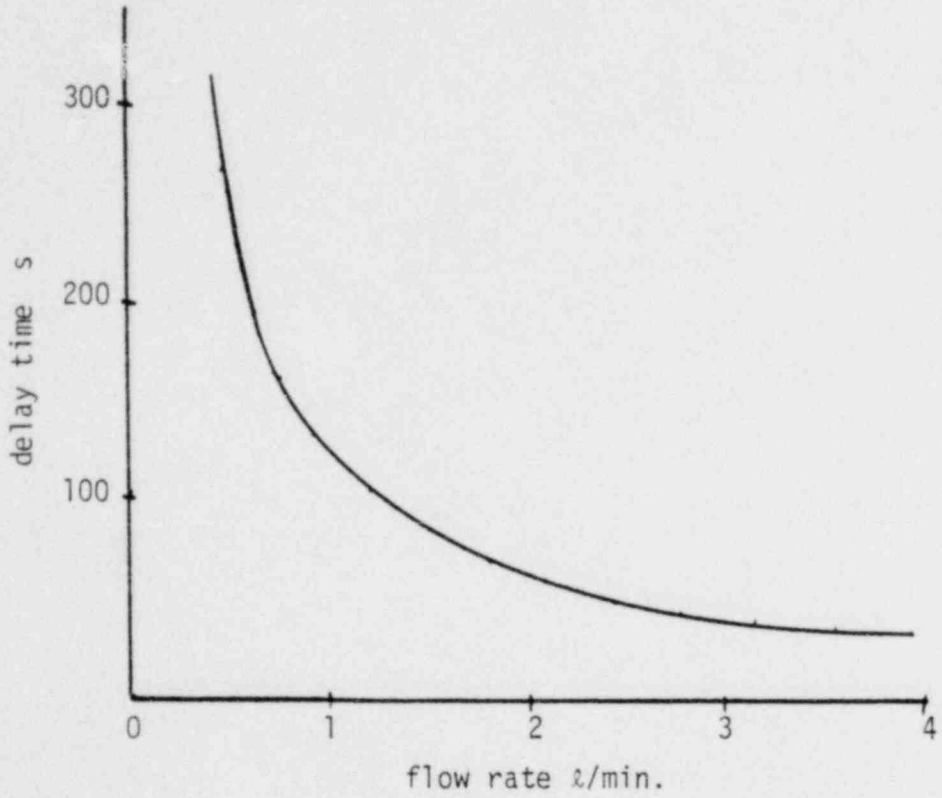

Figure 8 Delay time from fuel rods to detector station as a function of flow rate.

The release rate R of an isotope X from the fuel is defined as

R_x = number of atoms of isotope X released/sec

and the concentration of isotope X in the sweep gas is given by

$$C_x = \frac{R_x\ t}{V} \tag{2}$$

where

C_x = concentration of isotope X (atoms/cc)
t = time gas is in fuel rod (s)
V = volume of gas in fuel rod (cc)

The time that the gas is in the fuel rod, assuming no compression, is

$$t = \frac{V}{f} \tag{3}$$

where

f = flow rate (cc/s)

Thus, the concentration can be expressed as

$$C_x = \frac{R_x}{f} . \tag{4}$$

The actual measurement made with the spectrometer is the specific activity of the gas in the sample pipe, which is

$$A_D = \frac{P}{T_1^{\varepsilon}\ V_p\ B} \tag{5}$$

where

A_D = activity at the detector (disintegrations/s-cc)

P = counts in photopeak corresponding to isotope X

T_1 = live time for spectrum aquisition (s)

ε = efficiency of detector $\left[\frac{\text{gammas measured}}{\text{gammas emitted}}\right]$

V_p = volume of sample pipe viewed by detector (cc)

B = abundance of gamma ray (gammas/decay).

The specific activity of isotope X at the detector sample station is related to the specific activity at the fuel by

$$A_{Fx} = A_{Dx} e^{\lambda t_d} \tag{6}$$

where

A_{Fx} = specific activity of isotope X at fuel rod (disintegrations/s-cc)

λ = decay constant (s^{-1})

t_d = delay time from fuel to detector (s)

The specific activity of isotope X in the gas at the fuel is

$$A_{Fx} = C_x \lambda_x \tag{7}$$

Which, using Equation (4) can be written as

$$A_{Fx} = \frac{R_x \lambda_x}{f} \tag{8}$$

Solving for the release rate from the fuel, R_x, using Equations (8), (6), and (5)

$$R_x = \frac{Pfe^{\lambda t_d}}{T_\ell \varepsilon \quad V_p \quad \lambda B} \cdot \tag{9}$$

The normalized release rates for three isotopes of Xe and one isotope of Kr at three flow rates are plotted in Figure 9. A significantly higher release rate was measured at the highest flow rate. Since there appears to be no physical reason to expect the release rate to be dependent on flow, at least at these moderate flowrates, the problem is suspected to be in the flow measurement. Flow measurements at 1.4 and 2.2 ℓ/min were made with flowmeter F-141 (range 0.5 to 2.5 ℓ/min) while the 3.5 ℓ/min flow was measured with flowmeter F-142 (range 3 to 11 ℓ/min). A second complicating factor is that the data were taken at very high count rates, leading to large errors. Further tests will be conducted to resolve this problem. The consistency between the measurements at 1.4 and 2.2 ℓ/min indicate that small changes in flow rate, for which the same measurement instrumentation is used, do not affect the measured release rate. Measured release rates for several isotopes of Xe and Kr are given in Table 3. It must be noted that due to the extremely high count rates at which the data were acquired the error is estimated to be ±50%. This error will be reduced to the order of 20% during future tests.

The fifth and last objective of Scoping Test 1 was to establish the ability to use He/Xe sweep gas mixtures during fission gas measurement tests. A mixed fill gas test was conducted during installation of the FPMS. The qualitative results were that Xe and Kr isotopes were measurable at low power (~30% of full power) and that activation of the Xe in the sweep gas (10% Xe/90% He) did not grossly disturb the measurements. Since future plans include the use of a mixture of Xe or Ar with the He to increase fuel temperatures, this result is encouraging.

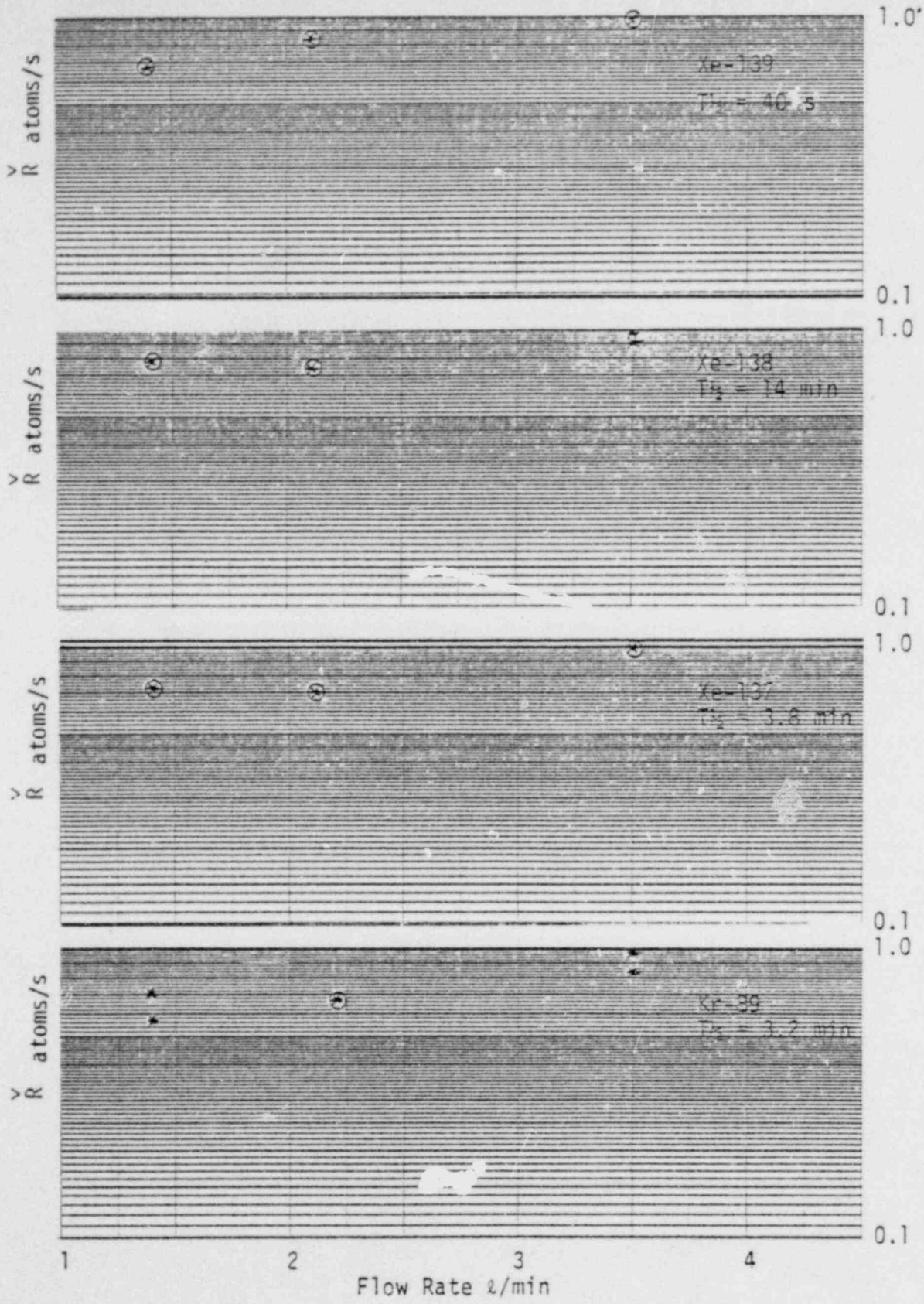

Figure 9 Release rate, $\bar{R}$, as a function of flow rate normalized to release rate at 3.5 ℓ/min flow rate. Circled data points indicate overlaying data.

TABLE 3. RELEASE RATES OF Xe AND Kr ISOTOPES*

Isotope	Release Rate (atoms/s)
^{135m}Xe**	2.4×10^{7}
^{137}Xe	3.8×10^{7}
^{138}Xe	5.6×10^{7}
^{139}Xe	3.3×10^{7}
^{85m}Kr	4.4×10^{7}
^{87}Kr	6.2×10^{7}
^{88}Kr	1.6×10^{8}
^{89}Kr	3.2×10^{7}
^{90}Kr	3.0×10^{6}

* At rod average linear heat rate of 25 kW/m.
** Not corrected for neutron capture.

The objective of Scoping Test 2 was to evaluate the feasibility of measuring the iodine 135 that plates out in the fuel rod and piping system during operation. This measurement, taken after the reactor has been operating at a constant power for a predetermined time and then scrammed, allows the steady state iodine release from the fuel to be estimated.

Measurement of the iodine that has plated out in the system is accomplished by measuring the daughter of ^{135}I, ^{135m}Xe. The production/decay rate of ^{135}I at a constant power reaches 99% of equilibrium value after 6.5 half lives (43 hours) and it is assumed that the release rate also comes to equilibrium during this time period. If all of the ^{135}I released from the fuel plates out in the system the plate out concentration of ^{135}I will reach equilibrium such that the amount decaying is equal to the amount being deposited. Measurements by Bannister et al[4] have shown that for temperature gradients similar to those in the IFA-430 assembly, essentially all of the iodine will be trapped in the piping or fuel rod; this is in agreement with the on-line measurements which show no iodine present in the gas stream at the detector station. During steady state operation, the equilibrium ^{135}I decay rate (from plated out ^{135}I) results in an effective ^{135m}Xe production rate, which, combined with the ^{135m}Xe released from the fuel as a gas, makes up the total ^{135m}Xe measured release rate. Scramming the reactor stops the production of I and Xe from fission and the rapid cooling of the fuel effectively halts any further diffusion of I and Xe out of the fuel. Following scram, the ^{135m}Xe production comes only from decay of the ^{135}I plated out in the system; thus, measurement of the ^{135m}Xe present after scram provides an indirect measurement of the equilibrium ^{135}I release.

To determine if this measurement was possible the reactor was scrammed from 98% of full power and the ^{135m}Xe in the sweep gas stream was measured for seven hours following the scram. Figure 10 shows the normalized ^{135m}Xe release rate as a function of time after

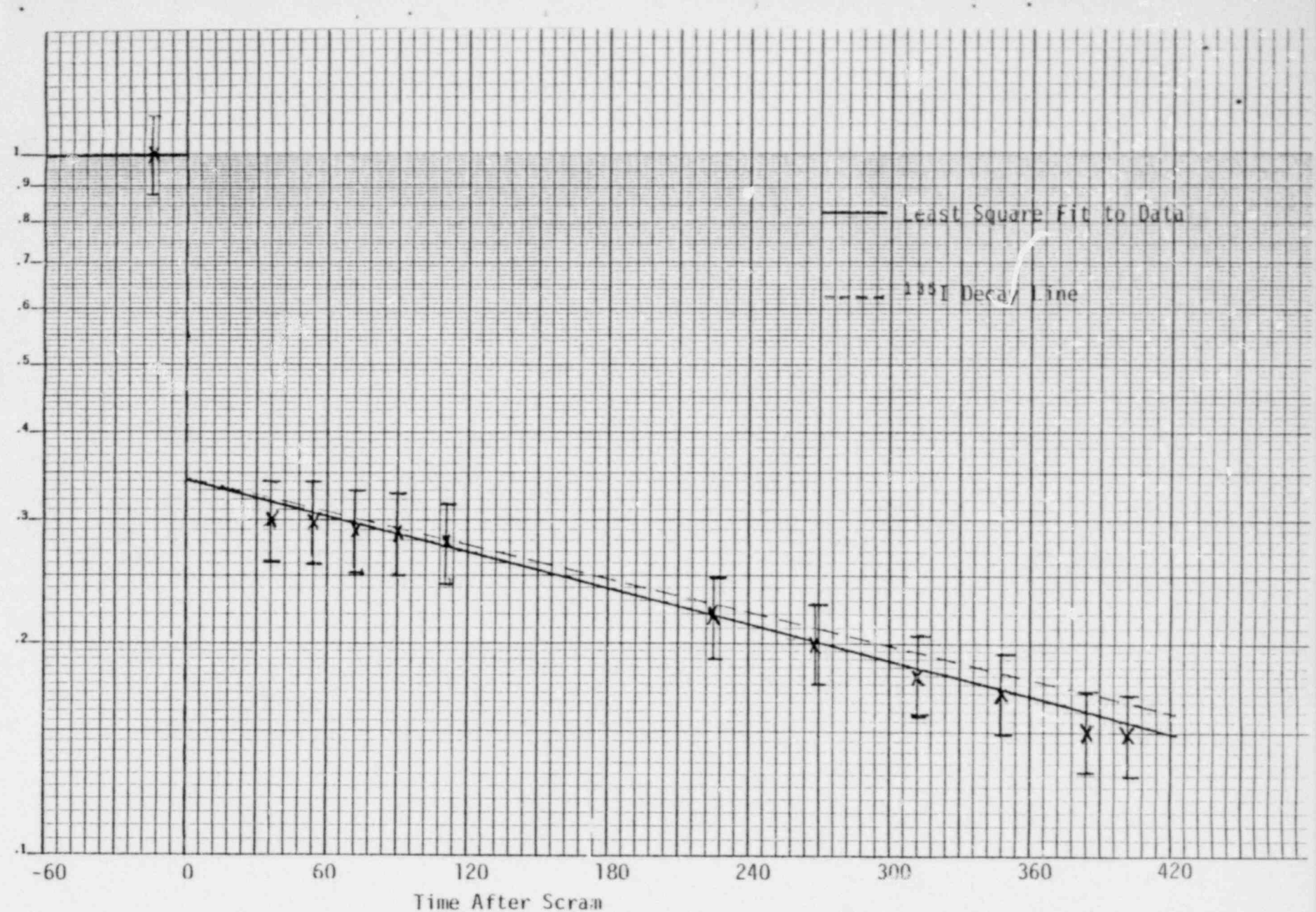

Figure 10 Post scram release of ^{135m}Xe resulting from the decay of ^{135}I plated out during full power operation. The data have been normalized to the steady state ^{135m}Xe release rate just prior to reactor scram. The least square fit line of the data is compared to the decay line expected for ^{135}I. Extrapolation to time zero shows ~ 35% of ^{135m}Xe measured prior to scram was due to ^{135}I decay and that ~ 65% was actually released from the fuel as Xe gas. (Note: data are only qualitative since ^{135}I had not reached equilibrium prior to scram.)

reactor scram. Since the reactor had not been operating at constant power for a long enough period prior to scram to achieve an equilibrium ^{135}I plate out condition the data are only qualitative and have been normalized to the ^{135m}Xe release rate at steady state conditions prior to scram. Plotted with the data are a least square fit line for the data, and the calculated decay line for ^{135}I. To determine the percent of the ^{135m}Xe being released from the fuel as a gas and the percent due to the decay of ^{135}I during full power operation the least square fit line is extrapolated to time zero (scram). For the scoping test data this shows that 35% of the ^{135m}Xe being measured at full power prior to scram was due to the decay of ^{135}I plated out in the system and 65% of the ^{135m}Xe was being released from the fuel as Xe gas. Thus the ^{135}I release rate at full power is ~35% of the measured ^{135m}Xe release rate. It must be emphasized that these fractions are not absolute numbers and apply only to this particular set of pre-scram conditions. The data set was collected during a period when the spectrometer was operating in an uncalibrated mode due to problems with the detector. The presentation of the analysis is included only for illustrative purposes to show that the ^{135}I release rate can be measured using this technique.

The scoping test results indicate that measurement of ^{85}Kr and ^{133}Xe, the fuel-rod-to-detector-station delay time, and a procedure for measurement during power-ramps should be explored during future tests. A brief discussion of each of these follows.

Two isotopes which are of interest in the analysis of fission gas release but are not measurable, in the on-line mode, are ^{133}Xe and ^{85}Kr. These isotopes provide information on the diffusion characteristics of Xe and Kr since their long half-lives permit them ample time to diffuse through the fuel. The long half-lives (5 days for ^{133}Xe, 10 years for ^{85}Kr) of these isotopes result in their specific activity (gammas emitted/cc-s) being very low in comparison to shorter half-lived isotopes. The short-lived isotopes (those in Table 2) dominate the spectra and mask the long lived isotopes. To

measure the ^{133}Xe and ^{85}Kr release rate a liquid nitrogen cooled charcoal filter could be installed to trap and concentrate the Xe and Kr gases. This filter would then be removed and stored for a period to allow the short lived isotopes to decay away thus making the long lived isotopes measurable. The complexity of modifying the system to make these measurements prohibits this technique at the present. Another alternative would be to store the gas in high pressure cylinders (to allow for storage of a large number of moles of gas) and then, following decay of the short lived isotopes, flow the gas back through the FPMS to measure the ^{85}Kr and ^{133}Xe. This would appear to be a viable alternative and will be explored further by the program in the future.

Actual measurement of the delay time from the fuel rods to the detector station is desirable. One possible method for measurement may be to cycle the outlet valve of the small-gap gas flow rod while holding the outlet valve of the large-gap gas flow rod open. The major part of the total gas flow would be through the large-gap rod and should not change significantly as the small-gap rod valve is cycled; thus, if the gas stream fission gas concentrations change due to the small-gap rod valve cycling the delay time can be measured.

A potential problem noted during the scoping tests was that small changes in power significantly affect the flow rate through the rods. This is due to the change in fuel diameter and thus hydraulic diameter as the temperature of the fuel changes. This implies that online measurement of fission gas release during power ramps will be very difficult. The operational procedures for such tests will have to be developed through actual testing experience.

IV. EVALUATION OF RELEASE-TO-BIRTH RATIOS AND RELEASE MECHANISMS

This section provides a brief discussion of the results of the scoping tests and provides a comparison of the data with results of other experiments. A discussion of release-to-birth ratios and mechanisms of release (recoil and diffusion) are included. Although a detailed error analysis has not been performed the error of the release measurements is estimated to be large (±50%) and is due to the high counting rates and to the modification of the operating voltage of the Ge detector necessary to compensate for equipment malfunctions that occurred during the tests. For future tests these problems will be eliminated and the data will be compared quantitatively with the results of previous experiments.

Determining the release to birth (R/B) ratios for the isotopes measured is a primary objective of the fission gas release tests. A model of the two fuel rods is being developed so that the birth rates can be determined using the ORIGEN[5] computer code. To obtain an estimate of the R/B ratios the birth rates for ^{138}Xe and ^{88}Kr were calculated using

$$B_x = Y_x f \tag{10}$$

where

B_x = birth rate of isotope x (#x/sec)
Y_x = cumulative fission yield for isotope x
f = fission rate

The R/B for ^{88}Kr, at an average rod linear heat rate of ~25 kW/m for the large gap rod, was 4.6×10^{-6} and for ^{138}Xe was 8.9×10^{-7}. The average fuel surface temperature for these measurements was ~750 K at which temperature recoil is expected to be the predominant mode of release.[6] Friskney and Turnbull[7,8] reported R/B for 1.2-mm diameter spheres of 8.6×10^{-4} for ^{88}Kr,

and 3.6×10^{-5} for ^{138}Xe at fuel temperatures of ~1050 K. Turnbull's data also showed that the R/B for these two isotopes is relatively linear for fuel temperatures in the range of 1300 K to 1050 K. Performing a least square fit to Turnbull's data and extrapolating to 750 K the expected R/B for a small sphere should be 3.4×10^{-5} for ^{138}Xe and 4.7×10^{-4} for ^{88}Kr. As expected, the R/B for the fuel pellets was smaller by a factor of ~50 than the ratio for the 1.2-mm spheres, which is due in part to the difference in the surface-to-volume (S/V) ratios. The S/V for a 1.2-mm sphere is 5 while for an intact IFA-430 fuel pellet (r = 5.4 mm L = 12.7 mm) S/V is 0.16. The R/B ratios normalized by the S/V for fuel pellets and spheres are shown in Table 4. The estimated error in the R/B for the spheres is ±20% and for the fuel pellets is ±50%.

TABLE 4. R/B RATIOS NORMALIZED BY S/V RATIOS FOR 1.2-mm SPHERES AND IFA-430 FUEL PELLETS

Isotope	$\left(\frac{R/B}{S/V}\right)$ Sphere	$\left(\frac{R/B}{S/V}\right)$ Pellet
^{138}Xe	7×10^{-6}	5×10^{-6}
^{88}Kr	9×10^{-5}	4×10^{-5}

This highly simplified comparison shows that the release rate per unit surface area from the IFA-430 fuel pellets is of the same order of magnitude as for the small spheres.

Of particular interest in the fission gas release measurements is determining the predominant mechanism of release. The three mechanisms for release of fission gases from UO_2 are diffusion, knockout, and recoil. Carrol[9,10] has shown that the predominant release mechanism can be determined from the ratio of two isotopes which have the same diffusion coefficient. Recoil release is proportional to the surface area and fission rate; thus, the ratio of the release rates for two isotopes is proportional to the ratio of the cumulative yields of the two isotopes. Diffusion release is

proportional to the fission yield of the isotopes and inversly proportional to the square root of their decay constants (for short half-lived parents). That is,

$$\text{Ratio of release rates for Recoil Release} \approx \frac{Y_x}{Y_y}$$

and

$$\text{Ratio of release rates for diffusion release} \approx \frac{Y_x}{Y_y}\sqrt{\frac{\lambda_y}{\lambda_x}}$$

where

Y_x = fission yield of isotope x
Y_y = fission yield of isotope y
λ_x = decay constant of isotope x
λ_y = decay constant of isotope y.

By comparing the measured release rate ratios with the ratios expected for diffusion or recoil release, the predominant release mechanism can be determined. Such a comparison is shown in Table 5 for release measurements made at ∿25 kW/m average power for the IFA-430 large gap fuel rod. The measured values of the release rates were determined from data that were taken at a very high count rate therefore having a large uncertainty, (∿±50%).

This preliminary analysis shows that Xe release is predominantly due to recoil while Kr release shows no conclusive trend. The diffusion coefficient[7] of Br, the precursor of Kr, at these temperatures is ∿10^3 times that of I, the Xe precursor; this may be a cause of the apparent difference in the behavior of these two gases.

TABLE 5 CALCULATED ISOTOPIC RELEASE RATIOS FOR DIFFUSION AND RECOIL RELEASE COMPARED WITH MEASURED RELEASE RATIOS

Isotopes	Measured	Recoil	Diffusion
$\frac{^{88}Kr}{^{89}Kr}$	5.0	0.8	5.6
$\frac{^{88}Kr}{^{87}Kr}$	2.6	1.4	3.1
$\frac{^{87}Kr}{^{85m}Kr}$	1.4	1.9	1.0
$\frac{^{88}Kr}{^{85m}Kr}$	3.7	2.7	0.3
$\frac{^{138}Xe}{^{139}Xe}$	1.7	1.3	5.0
$\frac{^{138}Xe}{^{137}Xe}$	1.5	1.0	2.0
$\frac{^{137}Xe}{^{139}Xe}$	1.2	1.2	2.9

More detailed models for fission gas release in a fuel element have been developed[6]; however, comparison of the preliminary test data with these models is beyond the scope of this report.

V. CONCLUSIONS

A fission product measurement system has been installed at the Halden Reactor to provide data on the release of xenon, krypton and iodine in the IFA-430 fuel rods. Two scoping tests have been performed which were designed to determine (a) the operating limits and optimum parameter setpoints for fission gas measurement during nuclear operation and (b) the capability to measure iodine plateout.

The predominant short-lived isotopes of Xe and Kr are measurable on line at steady state nuclear operating conditions. ^{85}Kr and ^{133}Xe are not measurable on line, but possible alternate techniques have been suggested and will be explored.

The Ge detector count rate can be held within acceptable limits using a 3-cm collimator with a 10-cm source-to-detector spacing. For this geometry, and at gas stream flow rates from 0.5 to 2.0 ℓ/min (delay times of 250 to 60 s), spectra can be acquired at ∿15-minute intervals.

There appears to be either a flow-rate-induced effect on fission gas measurements or a faulty flowmeter. The measured release rate of all isotopes changed by ∿30% when flow was increased from 2.2 to 3.5 ℓ/min; however, the flow measurements were made with a different flowmeter for the 2.2-ℓ/min flow than for the 3.5-ℓ/min flow. Checkout of the flowmeters will be performed and further tests conducted to identify the cause of the problem.

Spectra aquired when Xe/He sweep gas was used indicate that measurements are possible and that the Xe activation does not completely mask the spectrum.

Measurement of the iodine release rate can be made by measuring the ^{135m}Xe released in the decay of ^{135}I following reactor scram. The measurements made in Scoping Test 2 showed that the technique can

be used for the IFA-430 system and that the release rate of ^{135}I is of the same magni de as ^{135m}Xe and ^{138}Xe. This technique also allows the ^{135m}Xe release rate to be decoupled from the ^{135}I precursor contribution to the measured ^{135m}Xe release rate at power.

Measurement of release rate during power ramps will be very difficult because the flow rate is very sensitive to the changes in the fuel rod hydraulic diameter, which in turn is sensitive to the fuel rod power. An accurate measurement of the flow rate is needed to determine the release rates since the flowrate is an integral part of the release rate calculation.

Estimates of the release-to-birth ratio have been made using a highly simplified model for the birth term. The measured data compare well with data of other researchers when the data are normalized to the surface-to-volume ratio of the fuel.

The predominant mode of release is expected to be recoil and knockout rather than diffusion for the fuel temperatures of the scoping tests. The data for the Xe isotopes confirms this; however, the Kr isotope data indicates that diffusion may be significant. It is suggested that the high mobility of the Kr precursor, Br, may be the cause of this inconsistency.

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