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FISSION GAS RELEASE IN LWR FUEL MEASURED DURING NUCLEAR OPERATION[a]

MASTER

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ABSTRACT

A series of fuel behavior experiments are being conducted in the Heavy Boiling Water Reactor In Halden, Norway, to measure the release of Xe, Kr, and I fission products from typical light water reactor design fuel pellets. Helium gas is used to sweep the Xe and Kr fission gases out of two of the Instrumented Fuel Assembly 430 fuel rods and to a gamma spectrometer. The measurements of Xe and Kr are made during nuclear operation at steady state power, and for ^{135}I following reactor scram.

The first experiments were conducted at a burnup of 3000 MWd/t UO_2, at bulk average fuel temperatures of ~850 K and ~23 kW/m rod power. The measured release-to-birth ratios (R/B) of Xe and Kr are of the same magnitude as those observed in small UO_2 specimen experiments, when normalized to the estimated fuel surface-to-volume ratio. Preliminary analysis indicates that the release-to-birth ratios can be calculated, using diffusion coefficients determined from small specimen data, to within a factor of ~2 for the IFA-430 fuel. The release rate of ^{135}I is shown to be approximately equal to that of ^{135}Xe.

INTRODUCTION

Measurement of the release of fission product gases and volatiles from UO_2 during nuclear operation provides data which can be used in the development and assessment of models for predicting fission gas release, in the assessment of possible contributions of fission product volatiles (iodine) to stress corrosion cracking induced fuel rod failure, and in establishing the inventory available for release in the event of a breach in the fuel cladding.

Detailed and extensive studies of the release of fission gases and volatiles from small specimens of UO_2 have been performed both in the U. S.[1,2] and in the U. K.[3,4] From these studies the release to birth ratios (R/B) and diffusion coeffecients for Xe, Kr, and I have been determined and, as a consequence, the release of fission gases and iodine from small samples of UO_2 is predictable.

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b. OECD Halden Reactor Engineer

DISCLAIMER

The release of fission product gases and volatiles from full size UO_2 fuel pellets in typical LWR design fuel rods is being investigated by EG&G-Idaho using the Instrumented Fuel Assembly 430 (IFA-430) in the Halden Reactor located in Halden, Norway. This work is being performed as part of the U.S. Nuclear Regulatory Commission's Reactor Safety Research Program. This paper presents the preliminary results and analysis of experiments performed with IFA-430 to characterize the release of Xe, Kr, and I fission products during steady state nuclear operation at peak fuel centerline temperatures below 1250 K. The release to birth ratios (R/B) are presented, the possible release mechanisms discussed and the results are compared with data from small sample UO_2 experiments.

EXPERIMENT DESIGN AND CONDUCT

IFA-430 contains four fuel rods (1.28-m-long) with 10% enriched UO_2 fuel pellets of 95% theoretical density. Two rods, termed gas flow rods, each have a centerline thermocouple and three axially distributed pressure transducers mounted directly to the cladding to measure internal gas pressure. These two fuel rods are connected to a gas flow system, shown in Figure 1, which allows the released fission gases to be swept out of the fuel rods to a gamma spectrometer where they are quantitatively measured. The other two rods are equipped with two centerline and three off-center thermocouples and are sealed and pressurized with helium to 0.48 MPa.

The IFA-430 began irradiation in November 1978; the gamma spectrometer was installed in 1979 and the data for this analysis were obtained at an average burnup of ~ 3000 MWd /t UO_2. The fuel pellets are pressed and sintered UO_2, the grain size ranging from ~ 20μm at the outer radius to ~70μm in the middle of the pellet. The fuel-cladding gap sizes are 0.1 mm and 0.23 mm. The maximum peak centerline fuel temperatures over the life of the rods have been below 1560 K; the peak fuel centerline temperatures during the fission gas release tests were ~1250 K, the bulk average fuel temperature ~ 850 K. The average linear heat ratings of the rods were 22 to 23.5 Kw/m, with peak to average of 1.2.

The isotopic release rates for Xe and Kr were measured at a steady state rod power of 26 to 28 Kw (~25 w/gm) on three different occasions during a one week period; the reactor had been at constant power for sufficient time to allow all of the Xe and Kr isotopes used in the analysis to come to equilibrium prior to the fission gas release tests. The steady state release rates were determined by flowing a constant stream of He(~1 ℓ/min), through each fuel rod and acquiring 4 to 5 measurements of the content of the gas stream using on line gamma spectroscopy.

The technique described by Carroll[1] was used to measure the iodine release rate. The ^{135}Xe release measured at steady state power consists of ^{135}Xe released from the UO_2 in its gaseous state and ^{135}Xe coming from the decay of ^{135}I plated out on the surface of the cladding and piping. A stable ^{135}Xe release rate indicates that both the release of ^{135}Xe from the fuel as a gas and the release of ^{135}Xe as a result of ^{135}I decay are at equilibrium. At this stage the amount of ^{135}I being released from the fuel and plating out is equal to that decaying to ^{135}Xe. If the reactor is scrammed the production and release of ^{135}Xe and ^{135}I from the UO_2 essentially stops, and the ^{135}Xe measured after scram is a result of the decay of the plated out ^{135}I. Thus, by measuring the release of ^{135}Xe after scram the equilibrium ^{135}I release rate can be determined.

NOBLE GAS RELEASE

The Xe and Kr fission gases swept out of the fuel rods were measured in a continuous manner by gamma-spectrometry. The release rate, R, was calculated for each isotope and used with the calculated birth rate, B, to determine the release-to-birth ratio $(R/B)_o$. The birth rates were calculated with the ORIGEN isotope generation and depletion code.[5] The mean values of $(R/B)_o$ (observed release to birth ratio) along with the standard deviation for each isotope are presented in Table 1 for each fuel rod. The difference in $(R/B)_o$ between the two rods is not presently understood.

TABLE 1 MEASURED (R/B)o AND STANDARD DEVIATION (s) FOR Xe AND Kr ISOTOPES

	0.1-mm-Gap Rod		0.23-mm-Gap Rod		Mean
Isotope	$(R/B)o \cdot 10^5$	$s \cdot 10^5$	$(R/B)o \cdot 10^5$	$s \cdot 10^5$	$(R/B)m \cdot 10^5$
^{139}Xe	5.1	0.3	2.8	0.1	4.0
^{137}Xe	8.4	0.1	2.8	0.1	5.6
^{138}Xe	10.4	0.2	3.5	0.1	7.0
^{135m}Xe	15.4	0.7	9.9	0.6	12.7
^{135}Xe	45.4	5.0	30.7	6.0	38.1
^{90}Kr	5.5	0.4	3.5	0.1	4.5
^{89}Kr	9.5	0.3	3.2	0.1	6.4
^{87}Kr	22.4	1.2	9.9	0.7	16.2
^{88}Kr	43.3	2.0	16.2	1.7	29.8
^{85m}Kr	28.8	3.2	14.2	1.1	21.5

To compare the R/B ratios from the full size fuel pellets with previously published results, the R/B ratios must be normalized by the specimen surface-to-volume (S/V) ratio. The true surface area of the spheres has been determined[6] to be up to three times the geometric surface area, and that of the pellets up to 10 times the geometric surface area.[6] The 0.1-mm-gap rod IFA-430 fuel pellets are 12.7 mm long with a radius of 5.405 mm resulting in a geometric surface-to-volume ratio of 0.53 mm^{-1}. In Figure 2 the IFA-430 results are compared to Friskney and Turnbull's[4] data on 1.2-mm-diameter spheres. Figure 2 shows that the R/B ratios for the small spheres and the IFA-430 pellets are of the same order of magnitude, the data for the spheres generally falling within the range of the pellet data.

The mechanisms for release of the fission gases from the UO_2 are recoil, knockout, and diffusion. Recoil and knockout are usually the dominant mode of release at temperatures below 1000 K, and diffusion begins to dominate at temperatures above 1000 K. Because the fuel temperature range in IFA-430 is from ~750 K (at the fuel outer surface) up to ~1250 K (at the centerline), the release of fission gases is expected to be a combination of all three mechanisms. First the knockout and recoil mechanisms will be discussed and then, using experimentally determined diffusion coeffecients, the possibility of describing the release as a diffusion process will be discussed.

The parameter most often used to investigate release mechanisms is the decay constant (λ) dependence of the R/B ratios. The mean values of $(R/B)_o$

for the 0.1-mm-gap rod are plotted as a function of the decay constant in Figure 3. The solid line drawn through the data is the least-square-fit line, which has a slope of -0.33. Olander[7] has shown that theoretically the R/B ratio for recoil and knockout release can be expressed as

$$\frac{R}{B} = \frac{1}{4}\,\eta\,\frac{Sg}{V}\,\mu \;+\; \frac{1}{4}\,\frac{St}{V}\,\frac{\mu\alpha F}{N\lambda} \qquad (1)$$

where

η	= fraction of direct recoils not embedded in fuel or cladding		
St	= total surface area of fuel	α	= knock-on ejection yield
Sg	= geometric surface area of fuel	V	= fuel volume
μ	= fission fragment range	λ	= decay constant
N	= uranium atom density	F	= fission density

The first term on the right is the recoil contribution, and the second term is the knockout contribution. Equation (1) indicates that if knockout is the dominant mechanism R/B should show a λ^{-1} dependence and if recoil is the dominant mechanism R/B should show no λ dependence; however, the measured data show a $\lambda^{-0.33}$ dependence suggesting that there is a contribution from both recoil and knockout. These results are in agreement with the results of Soulhier[8], which show a $\lambda^{-0.2}$ to $\lambda^{-0.3}$ dependence for 95% dense sintered pellets at temperatures of ~500 K (note that knockout and recoil are independent of temperature). However, since the temperature range for IFA-430 is 750 K to 1250 K there may also be some diffusion release.

At fuel temperatures in the range 1000 to 1200 K Friskney and Turnbull's [4] data show only a very weak dependence of R/B (and thus the diffusion coefficient D) on temperature. The R/B ratio can be expressed as[4]

$$R/B = 3\left[\frac{\coth y - 1/y}{y(1 - y^2/x^2)} + \frac{\coth x - 1/x}{x\,(1 - x^2/y^2)}\right], \qquad (2)$$

where $x = a\sqrt{\lambda_1 D_1}$, $y = a\sqrt{\lambda_2 D_2}$, a = effective sphere size; D_1, D_2 and λ_1, λ_2 are the diffusion coefficients and decay constants for the precursor and the noble gas, respectively. This expression takes into account diffusion of the noble gas precursor. Noting the dependence of Equation (2) on λ, for fuel average bulk temperatures of ~850 K, if the measured R/B shows an approximate $\lambda^{-0.5}$ dependence then the release should be describable as a diffusion process. Thus, from the present data, the dominant release mechanism cannot be conclusively determined, and is expected to be a combination of recoil, knockout, and diffusion mechanisms.

Applying Equation (2) and Friskney's diffusion coefficients, D_1 and D_2, extrapolated to the average IFA-430 fuel temperatures, the calculated release-to-birth ratios (R/B)c, are compared to the measured, (R/B)m, ratios in Table 2. Note that these diffusion coeffecients are experimentally determined and implicitely contain the recoil and knockout contribution to R/B. An effective sphere radius of 100 μm was used, based on a = 3V/St; St being the total surface area for diffusion as given by Belle.[9] Table 2 shows that apparently the release of Xe and Kr from full size UO_2 pellets can be estimated using Equation (2). This conclusion is based on only the IFA-430 data which is in a relatively low temperature range (750-1250 K) and may not be applicable to high temperature (>1250 K) release. This is a

TABLE 2. CALCULATED, (R/B)c, AND MEASURED, (R/B)[a]m, RELEASE TO BIRTH RATIOS

Isotope	^{138}Xe	^{135m}Xe	^{135}Xe	^{87}Kr	^{88}Kr	^{85m}Kr
(R/B)m	7.0 E-5	1.3 E-4	3.8 E-4	1.6 E-4	3.0 E-4	2.2 E-4
(R/B)c	3.6 E-5	1.1 E-4	1.6 E-4	1.1 E-4	1.1 E-4	2.0 E-4
$\frac{(R/B)m}{(R/B)c}$	1.9	1.2	2.4	1.5	2.7	1.1

a. See Table 1.

preliminary analysis and further investigation is needed to understand the differences between the calculated and measured R/B for ^{138}Xe, ^{135}Xe, and ^{88}Kr.

IODINE RELEASE

The release rate of ^{135}I was determined by scramming the reactor and measuring the ^{135}Xe daughter of ^{135}I for 20 hours following scram. Figure 4 shows the normalized release rate of ^{135}Xe prior to and following the reactor scram. The ^{135}Xe release rate was essentially constant for the three days prior to scram indicating that the ^{135}Xe and ^{135}I release rate had come to equilibrium. The ^{135}Xe release rate after scram drops off with the 6.6 hour half-life of its ^{135}I precursor; by extrapolating this decay back to reactor scram the equilibrium ^{135}I release rate at power is determined. Figure 4 shows that, at equilibrium conditions just prior to scram the total ^{135}Xe release rate consisted of 52% from the decay of plated out ^{135}I and 48% from direct release of ^{135}Xe as a gas from the UO_2. Thus, the ^{135}I release rate is 0.52 times the measured ^{135}Xe total release rate, which results in a R/B rate for ^{135}I of 2.52×10^{-4}. The diffusion coefficients for iodine given by Friskney and Turnbull[4] only go down to temperatures of ~1000 K and, as they are nonlinearly dependent on temperature, cannot be extrapolated to the fuel temperatures in IFA-430 (~850 K bulk average). Thus, a meaningful comparison of calculated and measured results cannot be made at present.

CONCLUSIONS

The preliminary results from the IFA-430 experiment, have shown that measurement of the release of short lived Xe, Kr and I fission products during nuclear operation of LWR type fuel is possible. The release at bulk average fuel temperatures of ~850 K appears to be due to a combination of recoil, knockout, and diffusion mechanisms. The R/B ratios are in the range observed in small sphere specimen experiments, when corrected for the specimen S/V ratio. The diffusion equations that take precursor mobility into account, coupled with the diffusion coefficients determined by Friskney and Turnbull from small specimen experiments, appear to predict, within a factor of about two, the release of Xe and Kr from the IFA-430 pellet fuel.

The strong temperature dependence of the available Iodine diffusion coefficients, and the current absence of reliable diffusion coefficients in the IFA-430 fuel temperature range have precluded comparison of the IFA-430 iodine release data with calculated results. However, the release of ^{135}I has been shown to be the same order as the ^{135}Xe release.

ACKNOWLEDGMENT

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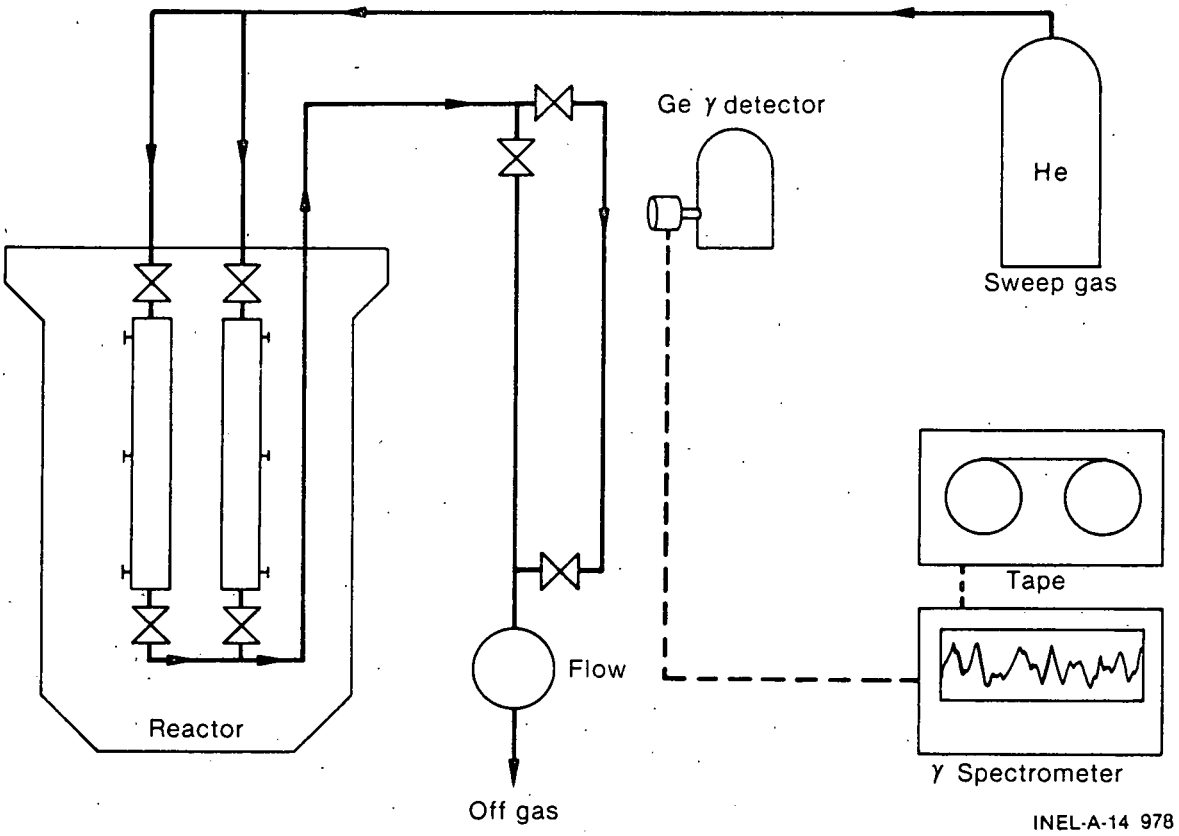

Figure 1. IFA-430 sweep gas and fission product measurement system.

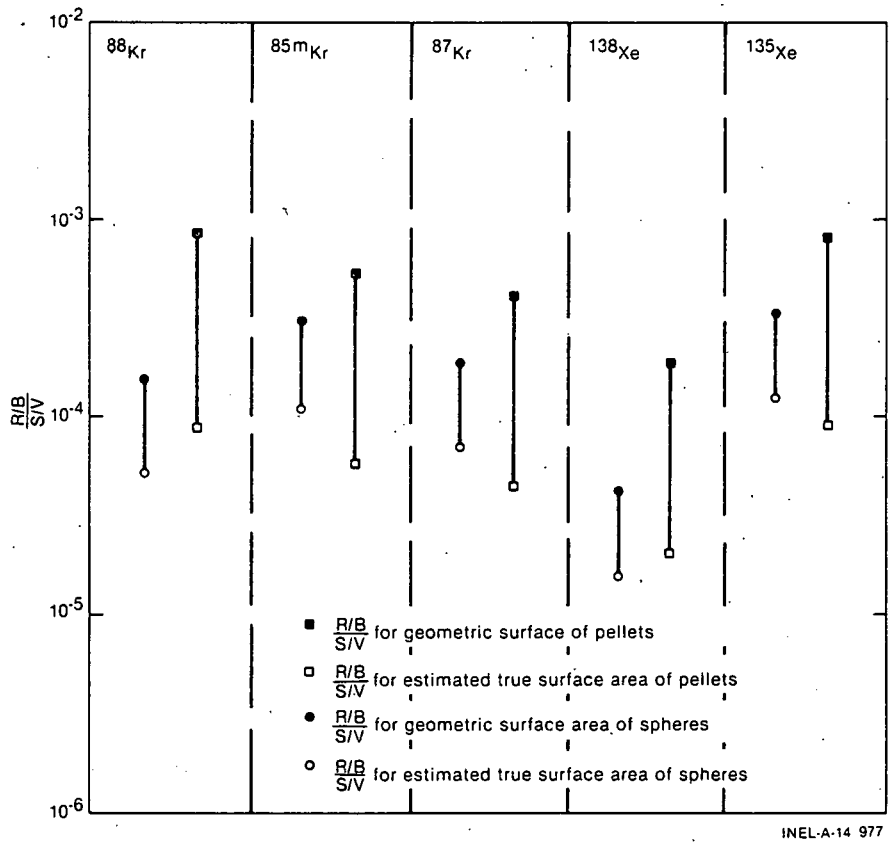

Figure 2. Comparison of the Release-to-Birth ratio, normalized by the surface-to-volume ratio, of the IFA-430 fuel pellets (■) and 1.2 mm diameter spheres[4] (●).

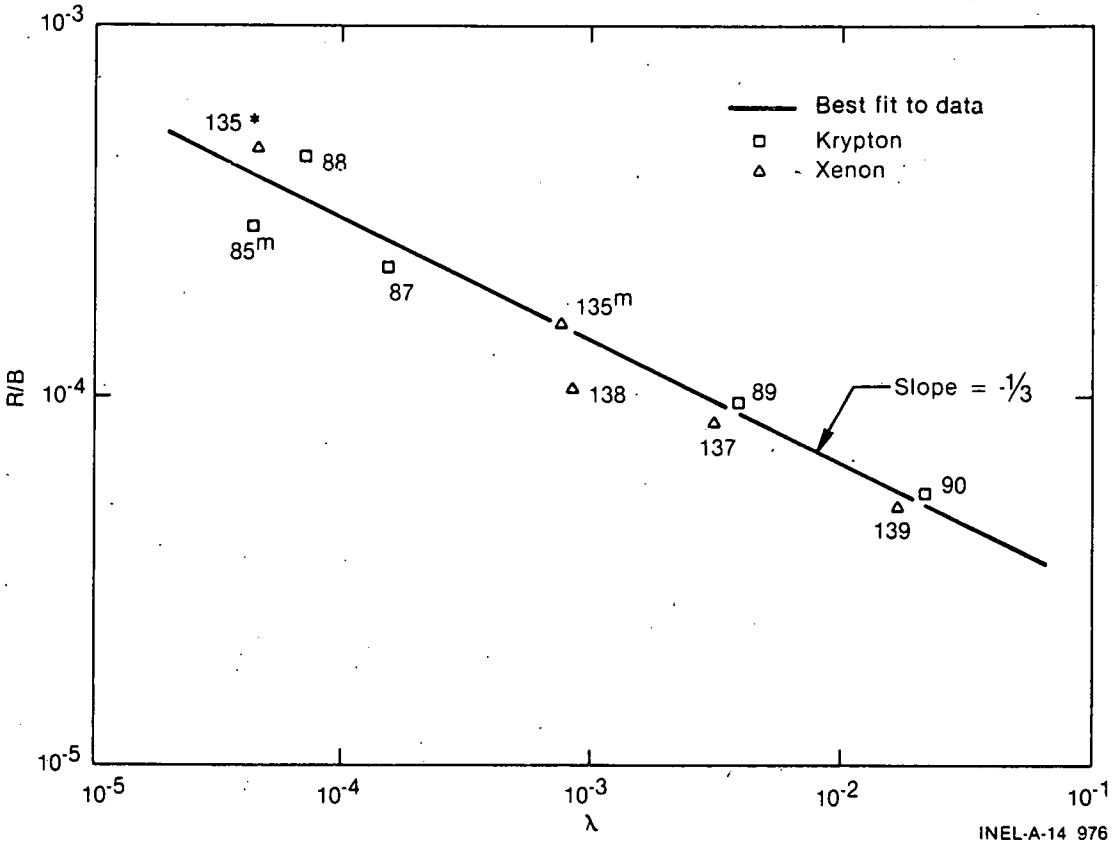

Figure 3. Measured R/B ratios as a function of decay constant (*^{135}Xe corrected for neutron capture).

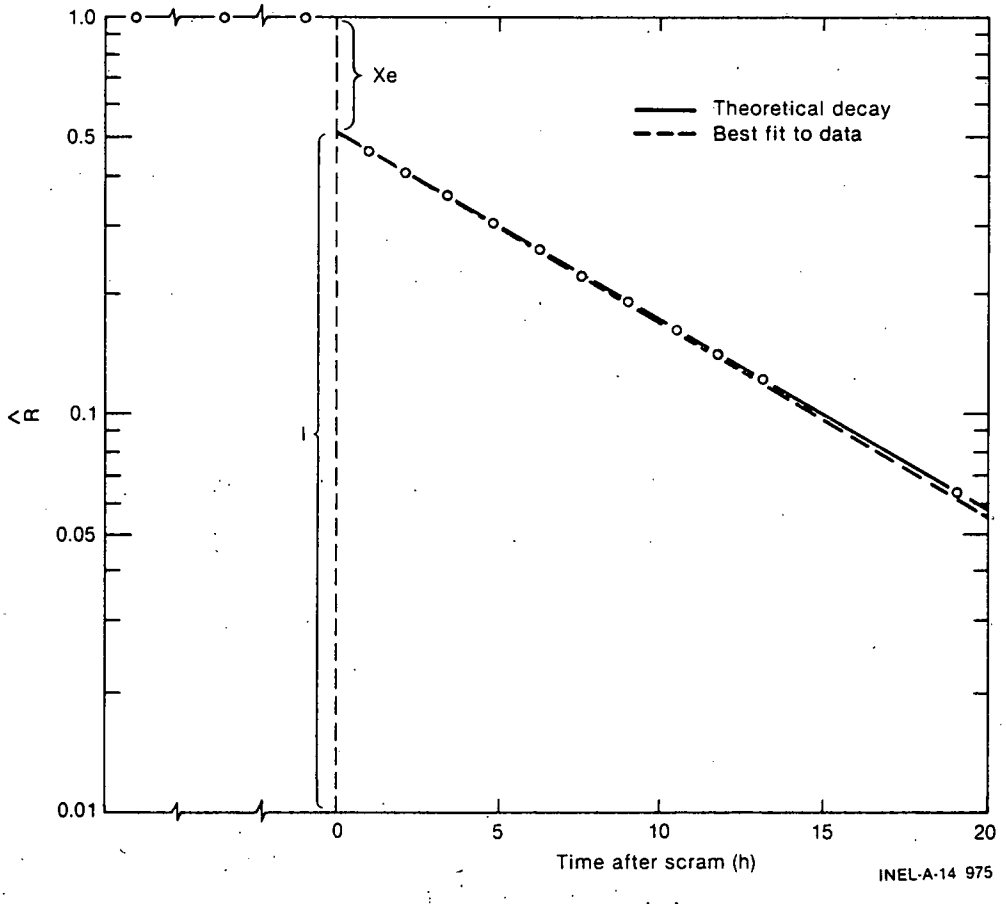

Figure 4. Normalized 135 Xe release rate (R) as a function of time for three days prior to scram and following scram indicating ^{135}I release rate is ∿ 50% of ^{135}Xe release rate.

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μ	= fission fragment range	λ	= decay constant
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The first term on the right is the recoil contribution, and the second term is the knockout contribution. Equation (1) indicates that if knockout is the dominant mechanism R/B should show a λ^{-1} dependence and if recoil is the dominant mechanism R/B should show no λ dependence; however, the measured data show a $\lambda^{-0.33}$ dependence suggesting that there is a contribution from both recoil and knockout. These results are in agreement with the results of Soulhier[8], which show a $\lambda^{-0.2}$ to $\lambda^{-0.3}$ dependence for 95% dense sintered pellets at temperatures of ~500 K (note that knockout and recoil are independent of temperature). However, since the temperature range for IFA-430 is 750 K to 1250 K there may also be some diffusion release.

At fuel temperatures in the range 1000 to 1200 K Friskney and Turnbull's [4] data show only a very weak dependence of R/B (and thus the diffusion coefficient D) on temperature. The R/B ratio can be expressed as[4]

$$R/B = 3\left[\frac{\coth y - 1/y}{y(1 - y^2/x^2)} + \frac{\coth x - 1/x}{x\,(1 - x^2/y^2)}\right], \qquad (2)$$

where $x = a\sqrt{\lambda_1 D_1}$, $y = a\sqrt{\lambda_2 D_2}$, a = effective sphere size; D_1, D_2 and λ_1, λ_2 are the diffusion coefficients and decay constants for the precursor and the noble gas, respectively. This expression takes into account diffusion of the noble gas precursor. Noting the dependence of Equation (2) on λ, for fuel average bulk temperatures of ~850 K, if the measured R/B shows an approximate $\lambda^{-0.5}$ dependence then the release should be describable as a diffusion process. Thus, from the present data, the dominant release mechanism cannot be conclusively determined, and is expected to be a combination of recoil, knockout, and diffusion mechanisms.

Applying Equation (2) and Friskney's diffusion coefficients, D_1 and D_2, extrapolated to the average IFA-430 fuel temperatures, the calculated release-to-birth ratios (R/B)c, are compared to the measured, (R/B)m, ratios in Table 2. Note that these diffusion coeffecients are experimentally determined and implicitely contain the recoil and knockout contribution to R/B. An effective sphere radius of 100 μm was used, based on a = 3V/St; St being the total surface area for diffusion as given by Belle.[9] Table 2 shows that apparently the release of Xe and Kr from full size UO_2 pellets can be estimated using Equation (2). This conclusion is based on only the IFA-430 data which is in a relatively low temperature range (750-1250 K) and may not be applicable to high temperature (>1250 K) release. This is a

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Isotope	^{138}Xe	^{135m}Xe	^{135}Xe	^{87}Kr	^{88}Kr	^{85m}Kr
(R/B)m	7.0 E-5	1.3 E-4	3.8 E-4	1.6 E-4	3.0 E-4	2.2 E-4
(R/B)c	3.6 E-5	1.1 E-4	1.6 E-4	1.1 E-4	1.1 E-4	2.0 E-4
$\frac{(R/B)m}{(R/B)c}$	1.9	1.2	2.4	1.5	2.7	1.1

a. See Table 1.

preliminary analysis and further investigation is needed to understand the differences between the calculated and measured R/B for ^{138}Xe, ^{135}Xe, and ^{88}Kr.

IODINE RELEASE

The release rate of ^{135}I was determined by scramming the reactor and measuring the ^{135}Xe daughter of ^{135}I for 20 hours following scram. Figure 4 shows the normalized release rate of ^{135}Xe prior to and following the reactor scram. The ^{135}Xe release rate was essentially constant for the three days prior to scram indicating that the ^{135}Xe and ^{135}I release rate had come to equilibrium. The ^{135}Xe release rate after scram drops off with the 6.6 hour half-life of its ^{135}I precursor; by extrapolating this decay back to reactor scram the equilibrium ^{135}I release rate at power is determined. Figure 4 shows that, at equilibrium conditions just prior to scram the total ^{135}Xe release rate consisted of 52% from the decay of plated out ^{135}I and 48% from direct release of ^{135}Xe as a gas from the UO_2. Thus, the ^{135}I release rate is 0.52 times the measured ^{135}Xe total release rate, which results in a R/B rate for ^{135}I of 2.52×10^{-4}. The diffusion coefficients for iodine given by Friskney and Turnbull[4] only go down to temperatures of ~1000 K and, as they are nonlinearly dependent on temperature, cannot be extrapolated to the fuel temperatures in IFA-430 (~850 K bulk average). Thus, a meaningful comparison of calculated and measured results cannot be made at present.

CONCLUSIONS

The preliminary results from the IFA-430 experiment, have shown that measurement of the release of short lived Xe, Kr and I fission products during nuclear operation of LWR type fuel is possible. The release at bulk average fuel temperatures of ~850 K appears to be due to a combination of recoil, knockout, and diffusion mechanisms. The R/B ratios are in the range observed in small sphere specimen experiments, when corrected for the specimen S/V ratio. The diffusion equations that take precursor mobility into account, coupled with the diffusion coefficients determined by Friskney and Turnbull from small specimen experiments, appear to predict, within a factor of about two, the release of Xe and Kr from the IFA-430 pellet fuel.

The strong temperature dependence of the available Iodine diffusion coefficients, and the current absence of reliable diffusion coefficients in the IFA-430 fuel temperature range have precluded comparison of the IFA-430 iodine release data with calculated results. However, the release of ^{135}I has been shown to be the same order as the ^{135}Xe release.

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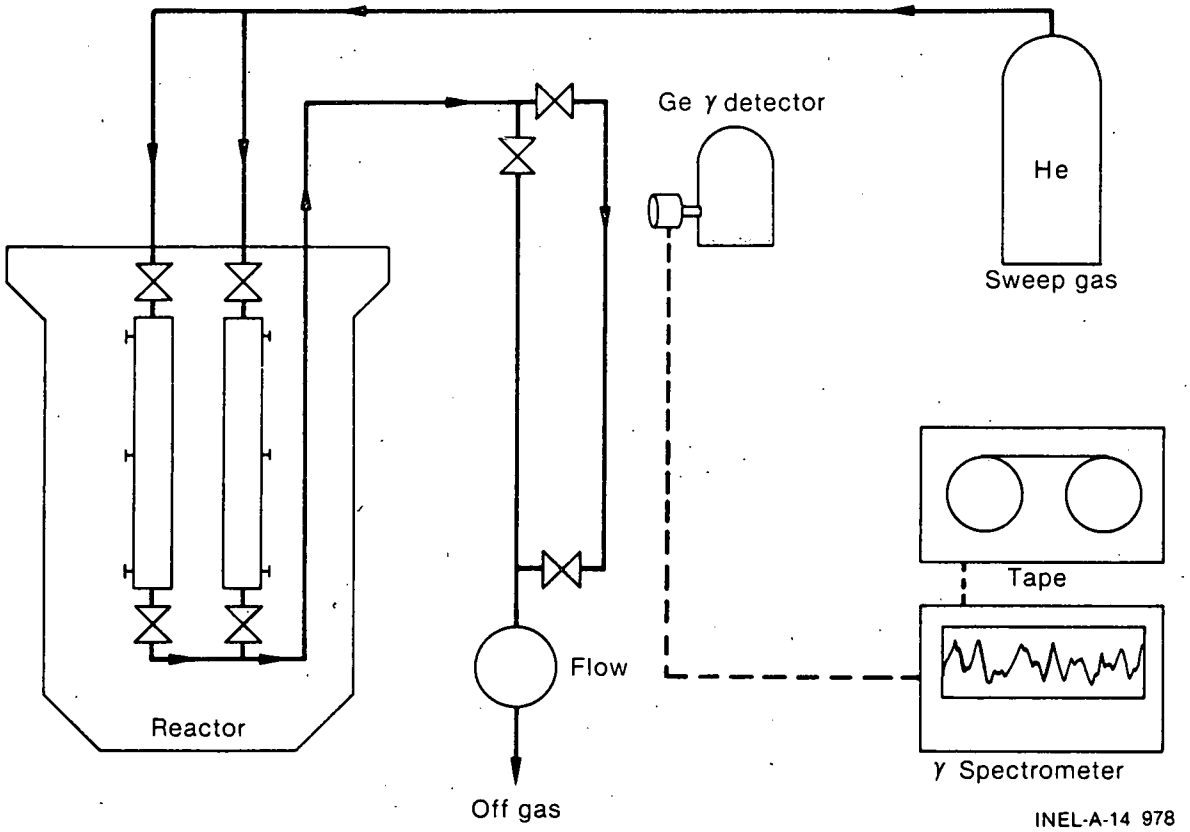

Figure 1. IFA-430 sweep gas and fission product measurement system.

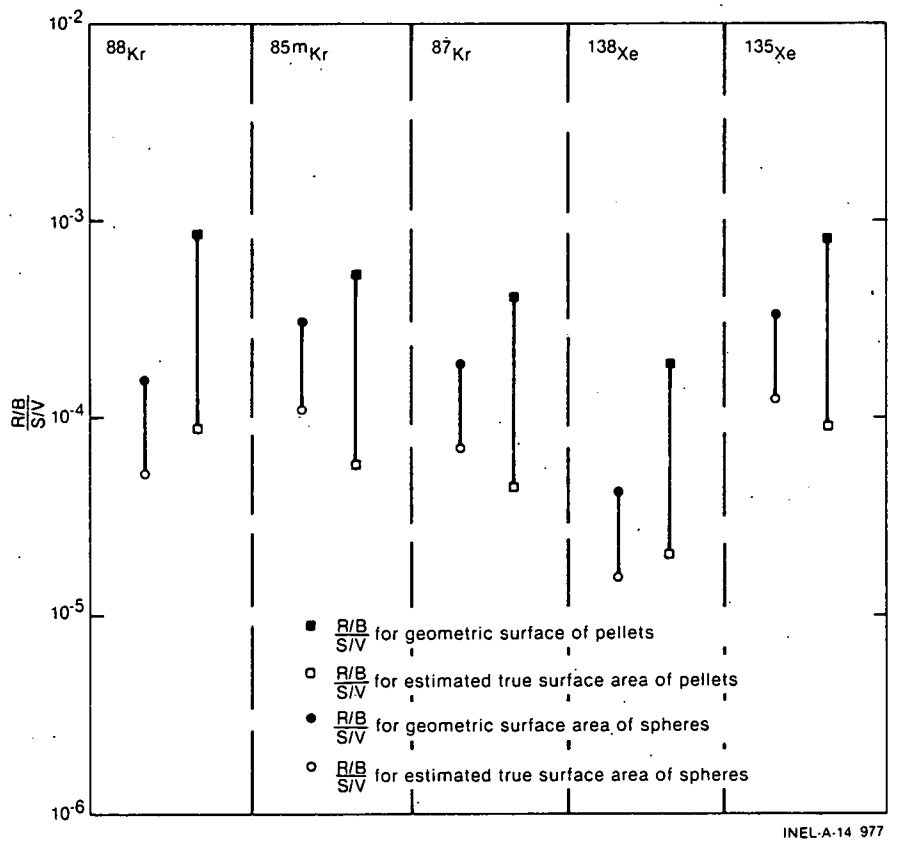

Figure 2. Comparison of the Release-to-Birth ratio, normalized by the surface-to-volume ratio, of the IFA-430 fuel pellets (■) and 1.2 mm diameter spheres[4] (●).

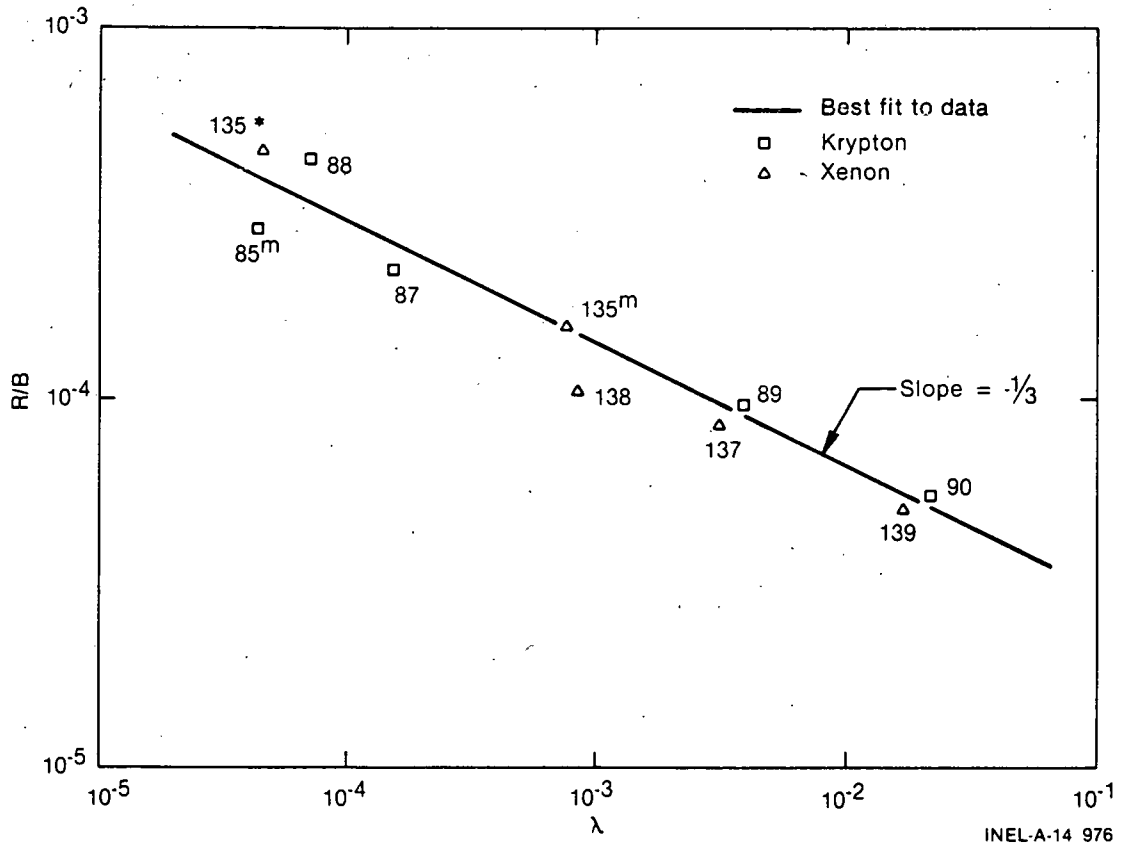

Figure 3. Measured R/B ratios as a function of decay constant (*^{135}Xe corrected for neutron capture).

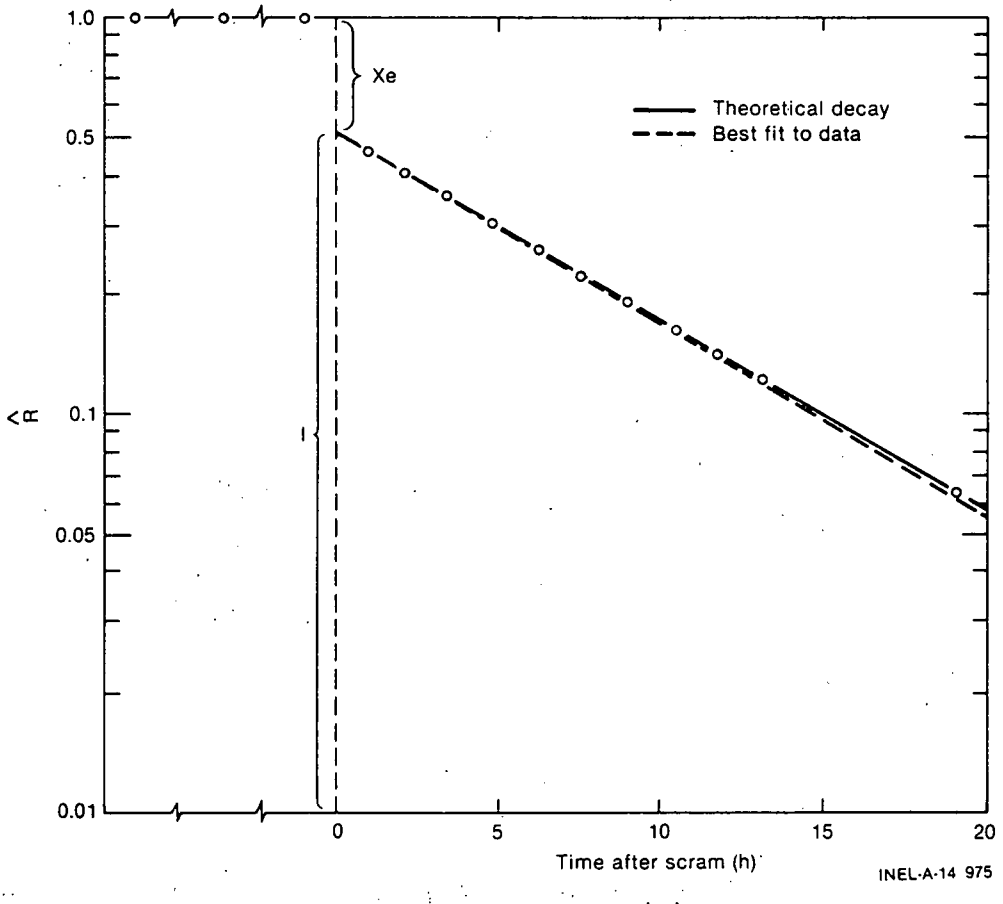

Figure 4. Normalized 135 Xe release rate (R) as a function of time for three days prior to scram and following scram indicating ^{135}I release rate is ∿ 50% of ^{135}Xe release rate.