THE EFFECT OF HEATING RATE ON FISSION-PRODUCT RELEASE: THE HRE1 EXPERIMENT

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ABSTRACT

Small samples of irradiated UO_2 were heated at controlled rates of 1, 5, 10, and 50 K/s to 1920 K in a flow of Ar/2%H_2, in order to investigate the effect of heating rate on fission-product release from CANDU® fuel. Observed fractional releases of Kr-85 were higher than those of the Xe-133 generated by trace-reirradiation, indicating that the grain-boundary inventory contributed significantly to releases under the test conditions. At 5 K/s, the Kr, Xe and Cs releases for heating to 1920 K were significantly higher than for heating to 1620 K, indicating that a threshold temperature must also be exceeded.

INTRODUCTION

Fission-product release during transient heating of UO_2 fuel is a phenomenon of interest for reactor operation and safety analysis. The fractional releases of volatile fission products and fission gases increase when UO_2 fuel is heated at rates higher than 1 K/s [1,2,3,4,5]. This experimental study was undertaken to investigate the heating-rate dependence of fission-product releases from CANDU® fuel.

Studies [1,2] on transient heating of civilian advanced gas-cooled reactor fuel (with a burnup of 432 MWh/kgU) to 1970 K or 2270 K at rates between 1 and 50 K/s indicated maximum releases around 10 K/s. The reduction in fractional release by increasing the heating rate from 10 to 50 K/s was ascribed to the change in the rate-controlling step of Kr release from bubble interlinkage to intergranular microcracking [3]. In annealing boiling-water reactor fuel samples (with burnups of 552 and 672 MWh/kgU) to 2070 K with heating rates of 0.03-10 K/s, the fractional Kr-85 releases were measured to be 20-30% and almost independent of the heating rate [4], although the release rate increased with increasing heating rate. A burst release of Kr was detected for each sample in the range 1770 K to 2070 K, followed by a slower diffusional release [4,5]. The burst releases were interpreted as being controlled by grain-boundary bubble interconnection and tunnel formation. A fractographic technique determined that no grain-boundary separation was observed at heating rates up to 10 K/s.

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The different fuels and test conditions used in these international programs make it difficult to compare their experimental results, and the effect of heating rate is difficult to isolate from the influence of other conditions, such as fuel characteristics and environment. The fuel samples in these experiments had much higher burnup and lower operating powers than CANDU fuel. Also, the dependence of the releases on the Grain-Boundary Inventory (GBI) of the samples had not been investigated. Accordingly, it was decided to investigate the dependence of fission-product release fraction on heating rate for CANDU fuel, in order to supply data for model development and validation.

EXPERIMENTAL APPARATUS, TEST CONDITIONS AND FUEL SAMPLES

A schematic diagram of the apparatus is given in Figure 1. A horizontal tube furnace with $MoSi_2$ heating elements (manufactured by Lindberg) was maintained at a temperature of 1920 K during the test. The alumina furnace tube was attached to a borosilicate glass tube containing a Type R (Pt-13%Rh vs. Pt) thermocouple (0.5 mm wire diameter) insulated using high-purity alumina twin-bore tubing. A small alumina sample boat (about 0.4 g) was also attached to the tip of the alumina twin-bore tubing. The thermocouple junction was within 1.5 mm of the sample. The thermocouple signal was passed out of the end of the borosilicate glass tube using a coil of Type R thermocouple extension wire. The out-furnace end of the alumina twin-bore tubing was surrounded by a shaped magnetic armature. A magnet outside the glass tube was driven by a slide table and stepping motor to couple to the magnetic armature and insert the sample into the furnace at controlled speeds (depending on the sample position) to attain the desired heating rates. The input parameters of the computer program controlling the stepping motor were adjusted in laboratory commissioning tests to deliver the desired heating rates. During the whole process, the sample temperature was continuously monitored at intervals of 1 s. The sample was held above 1900 K for about 600 s, then cooled by withdrawing the sample from the furnace. A typical temperature history is shown in Figure 2.

Zirconia oxygen sensors [6] monitored the oxygen partial pressure of the gas flow near the inlet and outlet of the furnace tube. The oxygen-sensor heaters were operated at about 970 K. A third oxygen sensor was used during laboratory commissioning tests to measure the gas transit time from the sample position to the middle of the delay coil. Inlet gas flows of Ar/2%H_2 (100 mL/min) were measured using a rotameter. After the gas left the furnace tube, it passed through high-efficiency particulate filters and a bubbler filled with NaOH solution to scrub vapour-phase iodine compounds before passing into the delay coil for γ-spectrometric monitoring.

Three γ-spectrometers were used to measure fission-product release (Figure 1). A direct-viewing γ-spectrometer measured the sample and background count rates for fission products other than noble gases before and after the test. Two γ-spectrometers monitored the Xe-133 and Kr-85 count rates in the carrier gas in two shielded delay coils of different γ-ray counting efficiency connected serially outside of the hot cell. The counting time for each γ-spectrum was 200 s for the delay-coil γ-spectrometers, and 300 s for the direct-viewing γ-spectrometer.

Most of the irradiated UO_2 samples were cut from central, mid-radial and peripheral locations in epoxy-impregnated segments of two CANDU power reactor fuel elements, employing the sample cutting method used for GBI test samples [7]. The samples were rectangular prisms 2.2 mm × 2.2 mm across the cross-section of the fuel, and approximately 5 mm long. The fuel element irradiated in Darlington Unit #2 had a peak linear power rating of 43 kW/m and a burnup of 219 MWh/kgU, while the fuel element irradiated in Bruce A Unit #3 had a peak linear power rating of 58 kW/m and a burnup of 539 MWh/kgU. The two elements had very different Kr-85 GBI fractions [7], with the measured GBI fraction for the central region of the Bruce fuel ranging up to ~90%, while the central region of the Darlington fuel only reached GBI fractions of ~40%. Most of the fuel samples were trace-reirradiated in the NRU reactor at low power to build up inventories of short-lived fission products (*e.g.*, Xe-133 and I-131). After the trace-reirradiation, the short-lived fission products were primarily resident in the UO_2 grains. The first tests were conducted 15 days after the end of the trace-reirradiation. Two irregular fragment samples were selected from a non-epoxied segment of Bruce fuel, and used in tests MC and MD.

DATA ANALYSIS

The γ-count rates of Kr-85 ($t_{1/2}$ = 10.85 years) were decay-corrected to 0800 h on the first test date, and the count rates of Xe-133 ($t_{1/2}$ = 5.25 d) were decay-corrected to 0800 h on the date of each test. The decay-corrected count rates (I) were used to calculate the released noble-gas activities during each counting period [8]:

$$\dot{A} = \frac{I\,\upsilon}{60\,E} \tag{1}$$

where $\dot{A}$ is the average release rate (in Bq/s) of the noble gas in this γ-counting period, υ is the flow rate of the carrier gas in mL/min through the delay coil, and E is the efficiency coefficient of the γ-spectrometer with the delay coil in units of cps/(Bq/mL). The timing of all the noble-gas data was corrected to account for the transit time between the fuel and the delay coils. The release rate and the cumulative release of the noble gases as a percentage of their inventories were determined by normalizing the measured activities to the calculated inventories in the sample.

Isotopes other than noble gases were γ-counted before and after the tests using the direct-viewing γ-spectrometer. These isotopes included long-lived isotopes created from the original power-reactor irradiation (Ru-106, Cs-134, and Cs-137). The low-temperature reirradiation created short-lived isotopes (Zr-95, Nb-95, Ru-103, I-131, Cs-134, La-140, and Ce-141). Because the pre- and post-test γ-countings of the sample were finished within less than three hours, and the half-lives of the isotopes listed above ranged from 8 days to 30 years, the count rates were not decay-corrected. This gives rise to an error of less than 1% for I-131, and much lower errors for the other isotopes.

The count rates of the isotopes retained in the sample after the test (I_t) were compared with the pre-test count rate (I_s) to obtain the fractional release (R) of the isotope. In some cases, changes in the counting geometry made a significant contribution to the change in the count rate. To account

for sample geometry effects, the measured post-test net count rate was normalized [9,10] by modifying the count rate by a factor k:

$$R = 1 - \frac{I_t - B_t}{k(I_s - B_s)} \qquad (2)$$

where B_s and B_t are the background count rates in the pre-test and post-test measurements, respectively. The factor k was obtained from an isotope of an element that was assumed not to be released (*i.e.*, $R = 0$) and the change in the count rate of this isotope was assumed to be entirely due to the counting geometry change. In trace-reirradiated samples, the I-131 releases (364 keV) were normalized using La-140 count rates at 328 or 487 keV, and Cs-137 count rates (662 keV) were normalized using La-140 count rates at 487 keV or Zr-95 count rates at 757 keV. The Cs-137 releases from as-received samples were normalized to the 512 keV count rate of Rh-106 (a short-lived daughter of Ru-106). No significant amounts of La, Zr or Ru would have been released from the fuel samples under these experimental conditions [9,10]. The k values for most tests were in the range 1.3 to 0.85, with values of about 0.5 for test BG (in which some of the sample was lost), and a value of 1.419 in test GG (for which no experimental reason could be discerned). In some tests, after removal from the furnace, part of the sample fell into the spill tray; normalization may not fully correct for the effect of such large geometry changes, because the distributions of different isotopes may not be identical.

The Kr-85 distribution varied in the fuel-pellet radial direction due to the migration of isotopes during the power-reactor irradiation. For the Darlington fuel, the distribution along the radial direction was fairly uniform, due to the comparatively low peak linear power of 43 kW/m, corroborated by its low fission-gas release of 0.3% [7,10]. The Bruce fuel had a high peak linear power of 58 kW/m, and the average noble-gas release of the fuel elements in the outer ring of the bundle during irradiation was 23.6% [7,11,12]. Grain-size measurements [12] and GBI test results [7,11] provided evidence of a steep change of Kr concentration in the mid-radial region of the Bruce fuel. The inventories of Kr in the samples were estimated from the calculated inventories and the estimated radial distribution [7]. The Xe inventories were nearly uniform along the radial direction, because the reirradiation was performed at low temperature (linear power about 2.5 kW/m).

The isotopes Ru-106, Cs-137 and Cs-134 were produced mainly during the power-reactor irradiation. The count rates of Ru-106 from different locations of the fuel did not show significant variation. For the Darlington fuel, the Cs-137 distribution (count rate per unit sample mass) was quite uniform along the radial direction. In the Bruce fuel, however, the count rates indicated depletion of Cs-137 at the centre, and much higher concentration at the mid-radius [11], in agreement with the GBI experiment results [7,11]. Due to the uncertainties of the sample radial location and the probable neutron-flux tilt during power-reactor irradiation, the variations in the Cs count rate in Bruce mid-radial samples were large. The rest of the observed isotopes (Zr-95, Nb-95, Ru-103, I-131, La-140, and Ce-141) were created during the trace-reirradiation, and were distributed nearly uniformly from the centre to the periphery of the fuels.

RESULTS

Twenty-five tests were performed on Darlington fuel samples and twelve tests were performed on Bruce fuel samples under inert-atmosphere (Ar/2%H_2) test conditions. The nominal heating rate and fractional releases of the fission products Kr-85, Xe-133, I-131 and Cs-137 are shown in Table 1. An example of the noble-gas releases during a test for a re-irradiated Darlington central sample is shown in Figure 2. Of the other isotopes observed, no statistically significant releases of Zr-95, Nb-95, Ru-103, Ru-106, La-140 or Ce-141 were observed under these test conditions.

Test MC (5 K/s, Bruce mid-radial sample) was performed on a non-epoxied sample, for comparison with tests HC and HK (5 K/s, epoxied Bruce mid-radial samples). The Kr-85, Xe-133 and I-131 fractional releases for test MC were between the releases for HC and HK, and the Cs-137 release was slightly lower for test MC.

Some tests were conducted under conditions different from those described above. In test BE, a reirradiated Darlington central sample was heated to 1900 K, but cooled to 1830 K within 50 s. The release results from this test were similar to those of other central-region Darlington samples. In test MD, a reirradiated Bruce mid-radial sample was heated to an unplanned low temperature of 1630 K due to failure of the insertion mechanism. The releases in this test were much lower than releases from other Bruce central and mid-radial samples. In test CG, a reirradiated Darlington peripheral sample was heated to a thermocouple temperature of 1850 K at 50 K/s and cooled immediately without the 600 s dwell at high temperature. The maximum actual sample temperature was estimated to be 1720 K. No noble-gas release was detected in test CG, so the test results are not reported in Table 1.

DISCUSSION

The technique of rapid sample heating by insertion into a hot furnace seems to have been appropriate for studying the releases of Kr, Xe, I and Cs isotopes from small fuel samples. No obvious influence of epoxying on the release of noble gases and Cs was detected in comparing test MC (5 K/s, non-epoxied Bruce mid-radial sample) with tests HC and HK (5 K/s, epoxied Bruce mid-radial samples). The differences in releases for tests MC, HC and HK are probably due to variations in GBI and other release-related characteristics, as manifested by variability in Cs-137 count rates observed for Bruce mid-radial samples. In most of the HRE1 tests, there was no evidence for release during cooling because of the low count rates of Kr during the cooling period. The apparent persistence of releases during cooling in some tests may have been due to gas mixing in the furnace tube and oxygen sensor gas volumes, contrary to the plug flow assumption in the gas release analysis.

The power and temperature histories of the fuel samples had a strong effect on fission-product release measured in the HRE1 experiment. The Kr-85 and Cs-137 fractional releases from the central and mid-radial Bruce samples were higher than releases from Darlington samples by a factor of two or more under similar test conditions, and the Kr-85 releases from central Darlington samples were higher than the releases from mid-radial and peripheral Darlington samples. The releases of

Xe-133 and I-131 from all types of fuel samples were similar. The higher transient release from fuels with higher peak power or higher burnup was related to the GBI, because Kr-85 and Cs-137 had significant GBI's, while very little of the Xe-133 and I-131 created during the low-power trace-reirradiation was resident on the grain boundaries [7,11]. The fractional releases of Kr-85 did not usually exceed the GBI fraction of Kr-85 expected for the sample [7].

For the Darlington fuel, the Kr release at 50 K/s was lower than at lower heating rates, but this was not the case for the Bruce fuel. For heating rates of 10 K/s or lower, the fractional releases of Kr-85 from Darlington samples were highest for central samples, intermediate for mid-radial samples and lowest for peripheral samples. At the heating rate of 50 K/s, the release of Kr-85 from Darlington samples from different radial locations did not show a significant difference. This seemed to be true also for the Bruce fuel, although no peripheral samples were tested. This is also in agreement with the hypothesis that the releases were largely due to GBI.

The effect of the heating rate on Cs-137 and I-131 releases was not as clear as it was on the Kr-85 release due to large sample geometry changes, which gave rise to significant additional measurement uncertainty in the fractional releases. The trend of the data, however, showed that the Cs-137 and I-131 releases were not significantly influenced by the heating rate. The releases of Cs-137 were mostly comparable with the release of Kr-85. Releases of Xe-133 were not sensitive to the radial location or heating rate for the Darlington fuel. For the Bruce samples, less Xe-133 seems to have been released from the central samples than from the mid-radial samples, probably because of the larger grain size in the central zone of the fuel.

It was observed in other experimental programs [5,13] that the critical temperature for the onset of burst-release during transient heating decreased with increasing burnup from 2070 K for fuel with burnup below 180 MWh/kgU, to 1870 K for burnups in the range 240 to 380 MWh/kgU, to 1770 K for burnups above 550 MWh/kgU. The burst-release temperatures in these experiments were measured with a resolution of 100 K. The normal HRE1 test temperature of 1920 K was about 50 K above the burst-release temperature measured for LWR fuel with the same burnup as the Darlington fuel. The Bruce fuel seems to have approached (but not exceeded) its burst-release temperature in test MD (conducted at 1630 K), while the Darlington fuel did not exceed its burst-release temperature in test CG (maximum temperature 1720 K). For heating rates of about 0.1 K/s in the HCE2 experiment, the Kr and Cs burst-release temperatures for high-burnup Bruce fuel were in the vicinity of 1620 K [14], and the burst-release temperature for LWR fuel with a burnup of 1375 MWh/kgU was about 1470 K [15], indicating that the burst-release temperature is probably lower at lower heating rates. The fuel microstructure must change during the heating to permit burst release, and a lower heating rate would give more time for microstructural changes, allowing the burst release to occur at lower temperature.

The release rate during the transient heating and soon after the 1920 K dwell started was higher than the release rate observed later in the dwell (Figure 2). This probably indicates that for the heating rates (1-50 K/s) in the HRE1 experiment, the main release mechanism was not diffusion, which should increase with temperature [16]. The mechanisms for release of GBI in transient heating may include grain-boundary separation and grain-boundary bubble growth with interlinkage to form grain edge tunnels. The mechanisms for release of grain inventory include single atom

diffusion and intragranular bubble migration. Fuel fragmentation may also release noble gases and Cs from grain boundaries, and in smaller portions from grains. Ceramographic examination is needed to determine the mechanisms and impact of the parameters on the observed releases.

CONCLUSIONS

An apparatus and technology were developed to study the effect of heating rate on fission-product release from irradiated UO_2 fuel samples at peak temperatures up to 1920 K at heating rates from 1 to 50 K/s. Releases of fission products from a typical discharged Darlington fuel element and a high-burnup Bruce fuel element were measured as a function of heating rate, temperature and sample radial location in the fuel element. Some of the samples were trace-reirradiated at low power, so that the Xe-133 and I-131 generated in the fuel samples had low grain-boundary inventory (GBI), while the Kr-85 and Cs-137 from the power reactor irradiations had a wide range of GBI values. The average heating rates produced in the HRE1 experiment were within ±20% of the nominal heating rates. The releases of noble gases during cooling were insignificant.

Releases of Kr-85 from the typical discharged Darlington fuel had the same dependence on heating rate as observed in previous studies [1,2]; the Kr-85 release at 50 K/s was lower than at lower heating rates. The highest fractional releases of Kr-85 from the Darlington fuel were in the tests with heating rates of 10 K/s. On the other hand, releases of Kr-85 from the high-burnup Bruce fuel were not dependent on heating rate. Observed fractional releases of Kr-85 were higher than those of Xe-133, indicating that the GBI contributed significantly to releases under the test conditions. The fractional releases of Kr-85 did not exceed the GBI fraction of Kr-85 expected for the sample. At the heating rate of 5 K/s, the Kr-85, Xe-133 and Cs-137 releases for heating to 1920 K were significantly higher than for heating to 1620 K, indicating that rapid heating alone does not cause significant fission-product releases, unless a threshold temperature is also exceeded.

For the heating rates (1-50 K/s) used in the HRE1 experiment, the main release mechanism seemed to be grain-boundary bubble interlinkage, which contributed to the burst-type releases observed. Grain-boundary separation may also have contributed to the releases observed at the higher heating rates. The diffusional release from the grain interiors to the grain boundaries was a secondary release mechanism, and may be the rate-controlling process after the burst release. Ceramographic examination is needed to determine the release mechanisms and impact of the parameters on the observed releases.

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Table 1: Percentage Releases (With 1σ Uncertainties) of Fission Products in HRE1 Tests

Test	Fuel	Radial Location	Nominal heating rate (K/s)	Xe-133		Kr-85		I-131		Cs-137	
				R%	±R%	R%	±R%	R%	±R%	R%	±R%
AA	Darlington (peak linear power 43 kW/m, burnup 219 MWh/kgU)	C	1	5.8%	2.2%	30.7%	4.0%	10.8%	8.2%	13.2%	5.5%
AC		C	5	6.3%	2.5%	26.9%	3.7%	12.1%	9.6%	13.0%	5.9%
AE		C	10	4.2%	1.6%	32.4%	4.2%	19.3%	4.7%	6.5%	4.9%
AG		C	50	5.0%	2.0%	9.6%	1.2%	3.1%	5.5%	2.3%	5.6%
BA		M	1	3.4%	1.3%	4.5%◊	0.6%	-1.0%n	9.7%	5.1%	3.6%
BC		M	5	5.3%	2.1%	18.0%	2.3%	6.5%	5.5%	10.9%	3.3%
BE↓		M	10	3.1%	1.2%	16.1%	2.1%	-8.0%n	13.6%	6.4%	5.0%
BG		M	50	4.7%	1.9%	5.8%	0.7%	15.2%	9.0%	2.7%	14.1%
BI		M	1	8.2%	3.1%	6.7%	0.8%	9.8%	5.7%	8.8%	4.2%
BM		M	10	9.3%	3.6%	9.6%	1.2%	20.1%	4.6%	3.9%	5.4%
BO		M	50	6.8%	2.6%	12.3%	1.6%	5.7%	6.2%	17.1%	4.0%
BY		M	10	6.4%	2.5%	4.8%◊	0.6%	NM	NM	NM	NM
CC		P	10	7.8%	3.1%	6.7%	0.9%	-4.2%n	10.1%	19.4%	6.0%
CH		P	50	2.9%	1.1%	1.8%◊	0.3%	16.5%	7.8%	5.2%	5.2%
DA		C	1			34.1%	4.5%	ND	ND	4.0%	4.0%
DC		C	5			4.5%◊	0.6%	ND	ND	6.1%	5.4%
DE		C	10			30.2%	3.9%	ND	ND	0.5%	4.4%
DG		C	50			6.0%◊	0.8%	ND	ND	5.1%	5.5%
EA		M	1			9.6%	1.2%	ND	ND	5.2%	3.5%
EC		M	5			3.4%◊	0.5%	ND	ND	7.7%	3.5%
EE		M	10			13.3%	1.7%	ND	ND	10.1%	3.6%
EG		M	50			31.2%	4.1%	ND	ND	-2.0%n	5.2%
FC		P	5			0.5%◊	0.1%	ND	ND	NM	NM
FG		P	50			10.1%	1.3%	ND	ND	8.9%	3.5%
GC	Bruce (peak linear power 58 kW/m, burnup 539 MWh/kgU)	C	5	1.7%	0.7%	50.4%	6.4%	-2.6%n	9.1%	58.0%	3.1%
GG		C	50	1.9%	0.7%	68.7%	8.7%	18.5%	7.8%	49.1%	4.0%
HC		M	5	2.3%	0.9%	29.6%	3.7%	-1.6%n	9.2%	66.6%	4.4%
HG		M	50	5.3%	2.0%	40.5%	5.2%	0.2%	8.9%	58.9%	5.1%
HK		M	5	7.8%	3.0%	42.8%	5.6%	23.4%	7.6%	31.2%	4.6%
HO		M	50	6.6%	2.5%	45.5%	6.0%	23.7%	7.8%	41.9%	4.0%
JC		C	5			78.2%	10.6%	ND	ND	49.9%†	3.8%
JG		C	50			47.7%	6.2%	ND	ND	53.0%†	2.6%
KC		M	5			60.7%	7.8%	ND	ND	79.2%†	3.1%
KG		M	50			72.3%	9.5%	ND	ND	68.6%†	4.2%
MC		M	5	13.0%	5.2%	35.1%	4.5%	19.6%	5.9%	19.7%	3.0%
MD↓		M	5	1.7%	0.7%	5.6%◊	0.8%	16.8%	6.9%	14.5%	5.5%

C, M and P indicate central, mid-radial and peripheral radial locations of the samples in the fuel element, respectively.

↓ Tested at temperatures lower than 1920 K.

◊ The actual Kr releases may have been higher than observed, because of low count rate.

n Negative values of percentage release, although not physically meaningful, are included to show the statistical distribution of the calculated value.

NM Direct-viewing measurements were not taken.

ND Not detected

† Not normalized, because Rh-106 512 keV γ-ray counting statistics were poor for these samples.

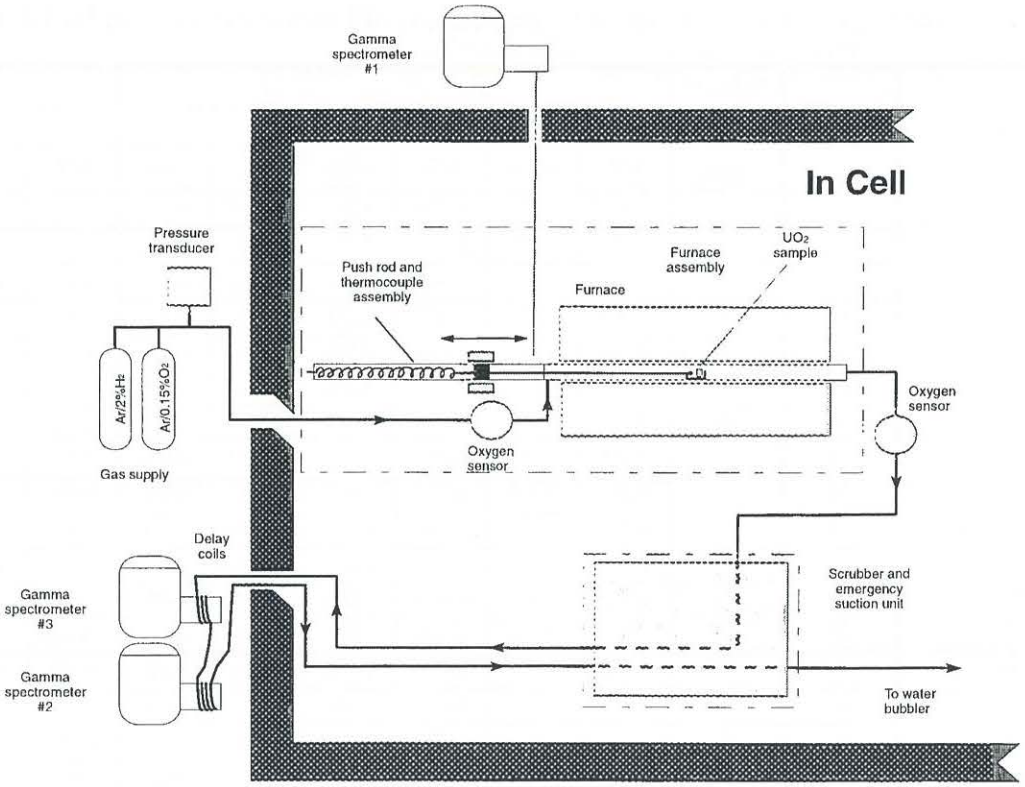

Figure 1: Schematic of the HRE1 experimental apparatus, showing sample fully inserted into the furnace.

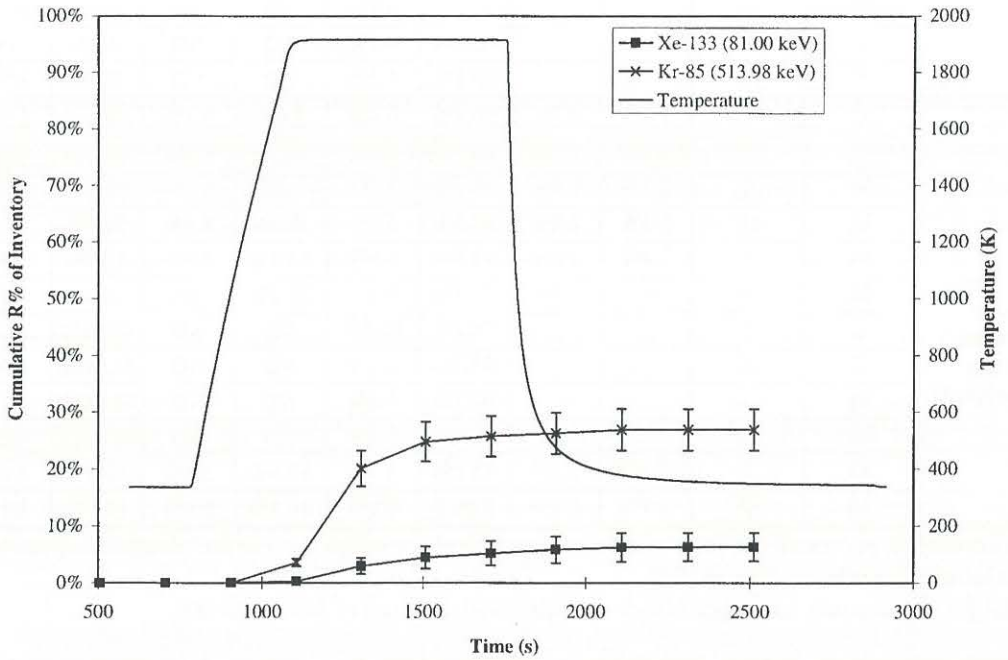

Figure 2: Noble-gas cumulative releases as fractions of inventories calculated from count rates at the 10-turn delay coil in test AC (reirradiated Darlington central sample, 5 K/s).