

From: "Dickson, Lawrence" <dicksonl@aecl.ca>
To: "Jason Schaperow" <JHS1@nrc.gov>
Date: Wed, Jan 12, 2000 8:47 AM
Subject: RE: Effect of air on FPR&T

Jason:
Glad to be of help. Please let me know if you need copies of any of our other papers or if you need clarification of any of the details.
Regards,
Lawrence

> -----
> From: Jason Schaperow[SMTP:JHS1@nrc.gov]
> Sent: Wednesday January 12, 2000 8:32 AM
> To: dicksonl@aecl.ca
> Cc: cgt@nrc.gov
> Subject: Re: Effect of air on FPR&T
>
> Thank you very much for your assistance.
>
> Sincerely,
> Jason Schaperow
>
> >>> "Dickson, Lawrence" <dicksonl@aecl.ca> 01/11 4:20 PM >>>
> Jason:
> The attached file (fpr_air.pdf) includes copies of 3 presentations I made
> at
> some recent PHEBUS meetings that are relevant to your concerns about the
> effect of high oxygen concentrations on FPR&T.
>
> <<FPR_air.pdf>>
>
> The attached file (Fuel conference ...pdf) is a copy of the paper
> describing
> the results of the HCE3 hot-cell fission-product release and transport
> experiment that we presented at the 6th International Conference on CANDU
> Fuel at Niagara Falls, Canada in 1999 September. It also contains some
> results relevant to your quest.
>
> <<Fuel Conference Paper_99_r4.PDF>>
>
> Please let me know if you have any questions or comments.
> Best regards,
> Lawrence
>
> !
>
>
>

G/2



***Effect of Oxygen on Fission-Product
Chemistry and Release from Fuel***

L.W. Dickson, J. McFarlane & R.S. Dickson, AECL

PHEBUS FP BIC Meeting

Arona, Italy

1999 April 12 & 13



Presentation Outline

- **Characteristics of “air ingress” scenario**
- **Summary of proposal for PHEBUS FPT5 “air ingress” test**
- **Effect of oxygen on FP chemistry and release from fuel:**
 - ◆ **chemical thermodynamics**
 - ◆ **FP release experimental results**
- **Conclusions**



Characteristics of an air ingress scenario

- **Initial stages are a transient in steam**
 - ◆ **either:**
 - **early phase of a severe accident, or**
 - **coolant boil-off for an open vessel**
 - ◆ **removes control rod material**
 - ◆ **remaining fuel is partially oxidized**
- **Air ingress phase**
 - ◆ **air mixes with residual steam in vessel, oxidizing remaining fuel at higher oxygen partial pressure (proposed mixture ~10% air)**



Proposed Objectives for FPT5

- **Bundle**
 - ◆ **investigate behaviour of partly oxidized fuel and FPs when exposed to steam/air mixture**
- **Circuit**
 - ◆ **investigate differences in FP behaviour in steam/H₂ and steam/air environments**
- **Containment**
 - ◆ **address uncertainties in iodine behaviour**
 - ◆ **investigate behaviour of FPs released by air oxidation**

Chemical Thermodynamics (1)

- **Oxidation effects on FP volatility are important**
 - ◆ **for some elements the oxides are more volatile than the metals**
 - e.g., Ru, Mo
 - ◆ **for others the metals are more volatile than the oxides**
 - e.g., Sr, Ba

Chemical Thermodynamics (2)

- **Chemical equilibrium calculations**
 - ◆ **volatilization of Ru & Mo increases in oxidizing environments due to formation of volatile oxides**
 - e.g., RuO, RuO₂, RuO₃, RuO₄, MoO₂, MoO₃
 - ◆ **Ru speciation in a 50/50 steam/air mixture**
 - T < 1200 K dominant form is RuO₂(s)
 - T < 1300 K dominant vapour form is RuO₄(g)
 - T > 1350 K vapour phase dominates mainly as RuO₃(g)
 - T > 1900 K dominant vapour form is RuO₂(g)

Chemical Thermodynamics (3)

- **Knudsen cell mass spectrometry and matrix isolation FTIR spectroscopy experiments**
 - ◆ **in oxidizing atmospheres MoO_2 affects circuit iodine chemistry by reaction with CsI to form Cs_2MoO_4**
 - iodine is released as I atom, I_2 or HI
 - ◆ **reaction of Mo metal with CsI to form Cs_2MoO_4 is not complete in the presence of stoichiometric UO_2 or slightly oxidized UO_{2+x} ($x \leq 0.01$)**



Chemical Thermodynamics - Conclusions

- **Oxidation affects FP volatility**
 - ◆ **oxides can be more or less volatile than metals**
- **Volatilization of Ru and Mo increases in oxidizing environments**
 - ◆ **volatile oxides**
- **In addition, MoO_2 reacts with CsI in oxidizing environments to form Cs_2MoO_4 and volatile iodine**

FP Release Experiments (1)

- **Measurements of FP release from irradiated fuel samples**
- **Fuel temperatures 1110 to 1930°C**
- **Environments**
 - ◆ **Ar / 2% H₂**
 - ◆ **Ar**
 - ◆ **steam**
 - ◆ **air**
- **Kr, Xe, Cs, I, Te, Ru, Nb, Ba, Sb, Zr, La, Ce, Pr, Eu, Y**
- **Releases of volatile FPs (Cs, I, Te) increase in oxidizing environments because diffusion coefficients increase with increasing stoichiometric deviation (x) in UO_{2+x}**



FP Release Experiments (2)

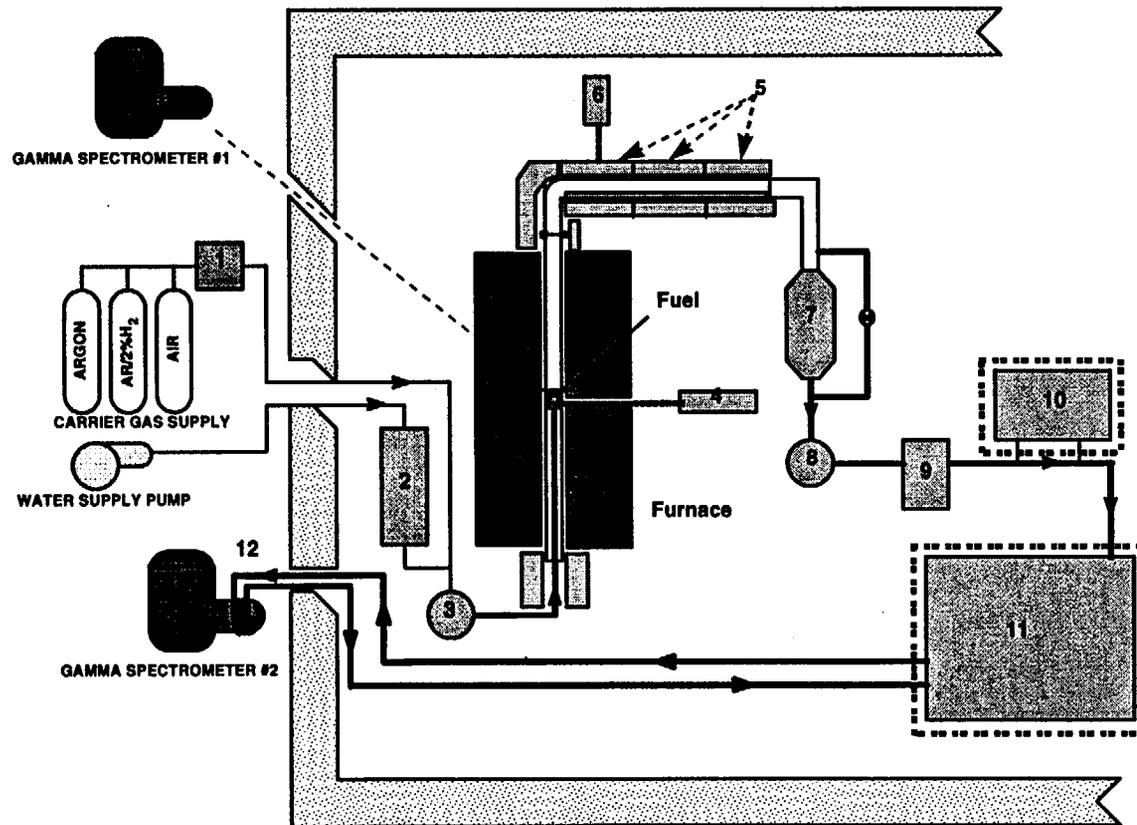
- **Experiments on Ru release from UO₂ fuel fragments show**
 - ◆ **no release in truly inert atmosphere up to 2200 K**
 - ◆ **slow releases observed at 2273 and 2373 K in atmospheres with low oxygen partial pressures ($\sim 2 \times 10^{-5}$ and $\sim 2 \times 10^{-3}$ atm)**
 - ◆ **releases in steam increase with increasing temperature**
 - **releases observed for $T > 1673$ K**
 - **release rate about the same as air at $T \approx 2300$ K**
 - ◆ **release rates in air faster at higher temperature**
 - **releases observed for $T > 1000$ K**
 - **release rate $> 1\%$ /minute at $T = 1400$ K**
 - **release rate $\sim 10\%$ /minute at $T > \sim 1800$ K**



FP Release Experiments (3)

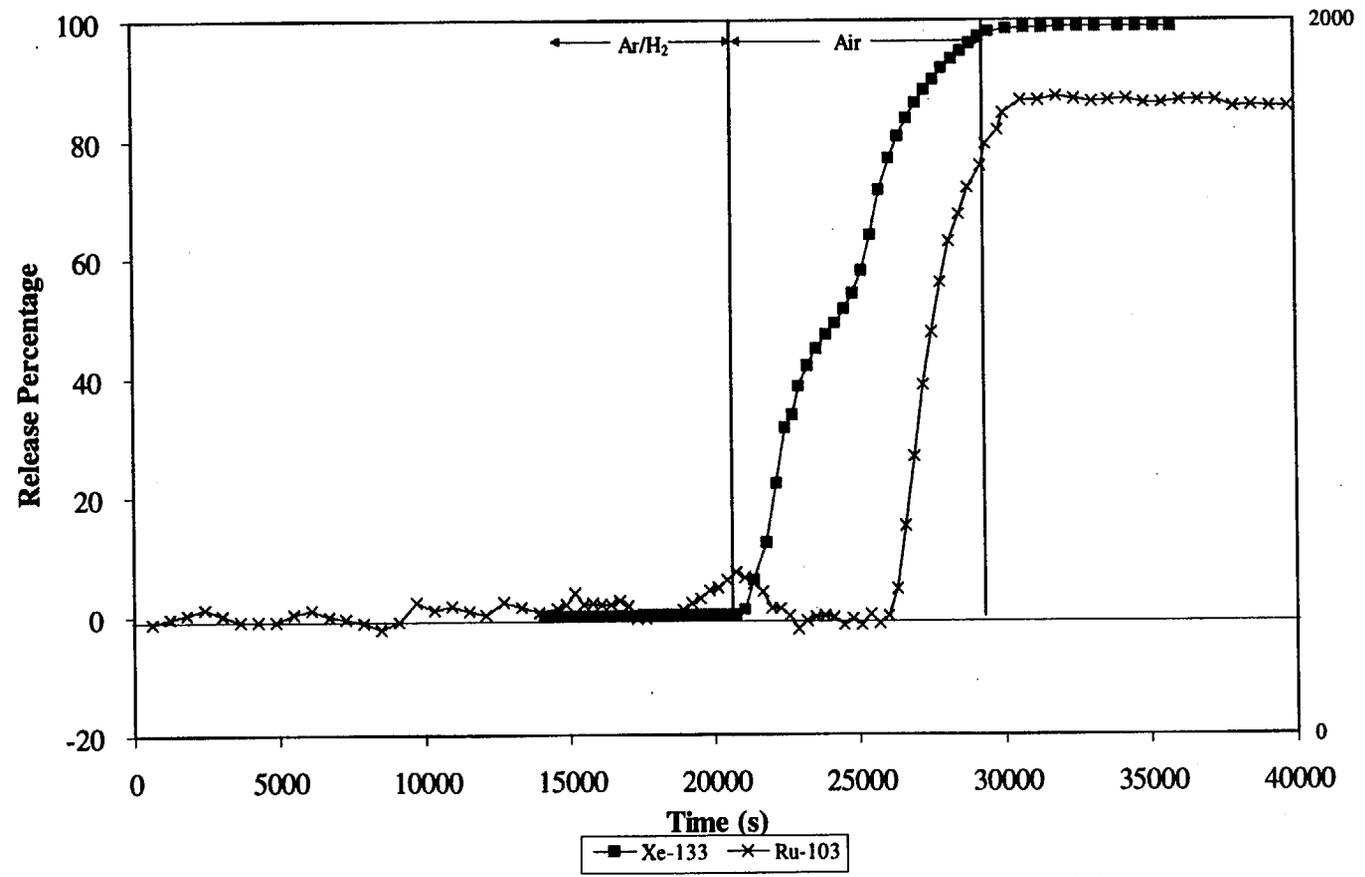
- **Experiments on Ru release from clad UO₂ fuel**
 - ◆ **enhanced Cs release observed after clad oxidation completed**
 - ◆ **enhanced Ru release observed 1000-3000 s after enhanced Cs release**
 - ◆ **~0.02% Ru release in inert atmosphere**
 - **2100 K, Ar/3.5%H₂O/0.5%H₂, ~4500 s**
 - ◆ **~4% Ru release in steam**
 - **2110 and 2200 K, slightly oxygen-rich steam, ~4000 s**
 - ◆ **~86% Ru release in air**
 - **2160 K, air, ~3700 s**

FP Release Experiments (4) - HCE3 Apparatus





FP Release Experiments (5)



Xenon and ruthenium release percentages as functions of time for HCE3 test H02 (sheathed Darlington fuel, air, 1890°C).



FP Release Experiments - Conclusions

- **Releases of volatile FPs (Cs, I, Te) increase in oxidizing environments because diffusion coefficients increase with increasing stoichiometric deviation (x) in UO_{2+x}**
- **Increases in oxygen potential have been observed to affect release behaviour of other FPs**
 - ◆ **Ru releases are more rapid, and more total Ru is released, with increases in oxygen potential**



***Experimental Observations and VICTORIA
Simulations of the Effect of Oxygen on Fission-
Product Transport in the Circuit***

L.W. Dickson and R.S. Dickson, AECL-CRL

PHEBUS FP CACIC Meeting

Arona, Italy

1999 April 14



Presentation Outline

- **Characteristics of “air ingress” scenario**
- **Summary of proposal for PHEBUS FPT5 “air ingress” test**
- **Effect of oxygen on FP transport:**
 - ◆ **FP transport experimental results**
 - ◆ **circuit FP transport calculations**
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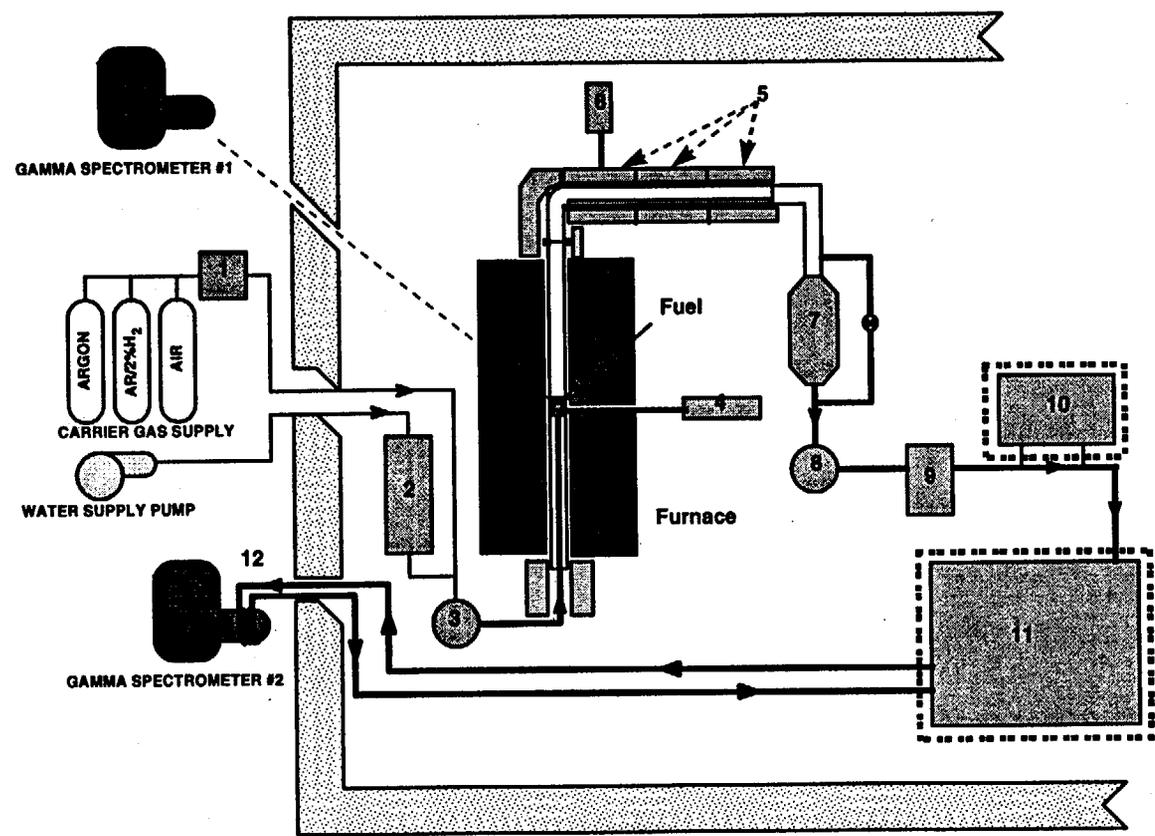


FP Transport Experiments (1)

- **Measurements of FP release from irradiated fuel samples**
- **Fuel temperatures 1110 to 1930°C**
- **Environments**
 - ◆ **Ar / 2% H₂**
 - ◆ **Ar**
 - ◆ **steam**
 - ◆ **air**
- **Kr, Xe, Cs, I, Te, Ru, Nb, Ba, Sb, Zr, La, Ce, Pr, Eu, Y**
- **Deposition and aerosol measurements made in some tests**



FP Transport Experiments (2) - HCE3 Apparatus





FP Transport Experiments (3)

- **Experimental results on transport of I and Ru released from clad UO₂ fuel in thermal gradient tubes**
 - ◆ **Iodine deposition**
 - 66% of released I deposited at 680 K in steam/H₂
 - 50% of released I deposited at 560 K in steam/0.3%O₂
 - 2% of released I deposited at 590 K in air
 - ◆ **Ru deposition**
 - (80±20)% of released Ru deposited at ~1070 K in steam/H₂
 - 95% of released Ru deposited at ~970 K in steam/0.3%O₂
 - 99% of released Ru deposited at ~800 K in air



FP Transport Experiments - Conclusions

- **Increases in oxygen potential have been observed to affect transport behaviour of FP**
 - ◆ **Ru deposition temperature decreases with increases in oxygen potential**
 - ◆ **iodine deposition decreases with increases in oxygen potential (66% in steam, 2% in air in CRL hot-cell FPR&T tests)**



Circuit FP Transport Calculations (1)

- **VICTORIA simulations of PHEBUS FP circuit transport**
 - ◆ **VICTORIA - because it simulates oxidative revaporization**
 - ◆ **13 node VICTORIA model based on 19 node SOPHAEROS model with FPT1 releases and thermalhydraulics**
 - ◆ **4 calculations performed**
 - **AIC - releases as with SOPHAEROS (including Ag/In/Cd)**
 - **AIC-OX - restart of AIC with steam/2%O₂ for 1000 s**
 - **NOAIC - releases as with SOPHAEROS (no Ag/In/Cd)**
 - **NOAIC-OX - restart of NOAIC with steam/2%O₂ for 1000 s**
 - ◆ **Deposition in following table presented as % of amount released from the bundle**

Circuit FP Transport Calculations (2)

- **Comparison of AIC and NOAIC simulation results**
 - ◆ **deposition similar but slightly lower in case without AIC**
- **Comparison of AIC and AIC-OX simulation results**
 - ◆ **about half of Mo deposit revaporizes (Cs_2MoO_4 remains)**
 - ◆ **~2% of Cs, I and Ru revaporization (mainly from plenum)**
 - ◆ **in low temperature zone (150°C) CsI is converted to AgI**
- **Comparison of NOAIC and NOAIC-OX simulation results**
 - ◆ **about half of Mo deposit revaporizes (Cs_2MoO_4 remains)**
 - ◆ **all of I is revaporized**
 - ◆ **~2% of Cs and Ru revaporization (mainly from plenum)**



Circuit FP Transport Calculations (3)

VICTORIA Simulation	Cs Deposit (%)	I Deposit (%)	Mo Deposit (%)	Ru Deposit (%)
AIC	39	36	42	40
AIC-OX	37	35	23	37
NOAIC	34	32	40	37
NOAIC-OX	33	0	16	34



FP Transport Calculations - Conclusions

- **VICTORIA simulations of PHEBUS FP circuit**
- **deposition results similar with and without control rod materials**
- **A period of 1000 s in steam/10%air results in**
 - ◆ **With control rod material present**
 - **revaporization of about 1/2 of Mo deposit**
 - **conversion of CsI to AgI at low temperatures (~150°C)**
 - **~2% Cs, I and Ru revaporization**
 - ◆ **Control rod material absent**
 - **similar revaporization of Mo (~1/2 of deposit)**
 - **100% revaporization of iodine**
 - **~2% Cs and Ru revaporization**



Effect of Oxygen on Mo and Ru Volatility

L.W. Dickson, AECL-CRL
PHEBUS FP SAWG Meeting
Arona, Italy
1999 April 15



Presentation Outline

- **Characteristics of “air ingress” scenario**
- **Summary of proposal for PHEBUS FPT5 “air ingress” test**
- **Effect of oxygen on Mo and Ru volatility:**
 - ◆ **chemical thermodynamics**
 - ◆ **FP release and transport experimental results**
 - ◆ **circuit FP transport calculations**
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- **Oxidation affects FP volatility**
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- **Volatilization of Ru and Mo increases in oxidizing environments**
 - ◆ **volatile oxides**
- **In addition, MoO_2 reacts with CsI in oxidizing environments to form Cs_2MoO_4 and volatile iodine**



FPR&T Experiments

- **Releases of volatile FPs (Cs, I, Te) increase in oxidizing environments because diffusion coefficients increase with increasing stoichiometric deviation (x) in UO_{2+x}**
- **Increases in oxygen potential have been observed to affect release and transport behaviour of other FPs**
 - ◆ **Ru releases are more rapid, and more total Ru is released, with increases in oxygen potential**
 - ◆ **iodine deposition decreases with increases in oxygen potential (66% in steam, 2% in air in CRL hot-cell FPR&T tests)**



FP Transport Calculations

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Conclusions

- **FPT5 test, as proposed, should:**
 - ◆ **confirm our understanding of phenomena relevant to “air ingress” scenarios**
 - **effects of air on fuel behaviour**
 - **changes in FP behaviour in air**
 - **address uncertainties in iodine behaviour**
 - ◆ **provide integrated effects demonstration of differences in phenomena between steam/H₂ and steam/air by comparison with FPT0, FPT1 and FPT2 results**

RELEASE OF FISSION PRODUCTS FROM CANDU FUEL IN AIR, STEAM AND ARGON ATMOSPHERES AT 1500-1900°C: THE HCE3 EXPERIMENT

BARRAND, R.D., DICKSON, R.S., LIU, Z. AND SEMENIUK, D.D.

Fuel Safety Branch
Reactor Safety Division
AECL, Chalk River Laboratories,
Chalk River, Ontario, CANADA K0J 1J0

ABSTRACT

Fission-product release from Zircaloy-sheathed segments of CANDU fuel was measured in the six HCE3 tests in argon, steam and air environments at peak temperatures between 1510 and 1930°C. On-line γ -spectrometry showed significant fractional releases for Xe, Kr, I, Cs, Te and Ru. Post-test deposition measurements also detected releases of Sn, Sb, Ag, Ba, Zr and Nb. The release rates of Kr, Xe, I and Cs were very low before complete sheath oxidation; their peak release rates in steam after complete sheath oxidation were not very temperature-dependent. Ru release was delayed by more than 2000 s after oxidative releases of the volatile fission products.

INTRODUCTION

This paper describes the results of a fission-product release experiment conducted in Universal Cell #3 at Chalk River Laboratories (CRL). The experiment, designated HCE3 (Hot-Cell Experiment 3), was sponsored by the CANDU Owners Group (COG) as part of its program to determine fission-product release, deposition and transport from fuel under accident conditions. The experiment was intended to measure fission-product release and transport from sheathed fuel samples at higher temperatures than previous experiments; the highest temperature previously used in sheathed-fuel tests at CRL was 1650°C [1].

Zircaloy-sheathed segments of CANDU fuel were heated to peak temperatures in the range 1500-1930°C. Air, steam and/or argon atmospheres were flowed past the heated fuel, through a deposition tube into an aerosol collection apparatus. Fission-product release data were collected using direct-viewing and gas-monitoring γ -spectrometry, and post-test γ -scanning of the apparatus.

EXPERIMENTAL APPARATUS & FUEL SAMPLES

A general schematic of the experimental apparatus is shown in Figure 1. A specialized furnace made by Kanthal Artcor Inc. employing zirconia resistive heating elements was used. Chromel pre-heaters

were used to heat the zirconia heating elements to $>600^{\circ}\text{C}$, at which temperature the zirconia heating elements conduct sufficient current to take over the heating task.

The fuel sample was positioned in the centre of the hot zone of the furnace on a thoria pedestal. The gas was flowed upward through the zirconia furnace tube. The vertical furnace tube was surrounded by a segmented zirconia guide tube. The annulus between the furnace tube and the guide tube was flooded with flowing argon or argon/hydrogen mixtures to retard the permeation of oxygen into the furnace tube at high temperatures. The furnace tube, deposition tube and aerosol collection filters were replaced after each test to allow γ -scanning measurement of released fission products.

The direct-viewing γ -spectrometer (Figure 1) monitored the fractional retention of fission products in the sample during 300 s intervals in the test, while the gas-monitoring γ -spectrometer measured the noble-gas activity in the gas stream downstream of the condenser during 300 s intervals. Zirconia oxygen sensors were used to monitor the oxygen partial pressure of the test atmosphere upstream of the furnace tube and downstream of the aerosol collection filters. Sample temperatures were measured using a pyrometer and thermocouples in the laboratory. Unfortunately, the pyrometer failed on placing the apparatus in the hot cell, so sample temperatures for hot-cell tests were inferred from measurements at a thermocouple remote from the furnace tube using sample temperature behaviour measured in laboratory tests with similar temperature histories.

All irradiated fuel specimens used in this experiment were taken from an outer element of a Darlington fuel bundle (peak linear power 46 kW/m, burnup 234 MWh/kgU, 0.6% measured noble-gas gap inventory). Specimens were obtained by cutting the fuel element transversely, giving ~25-mm-long segments of fuel element. Zircaloy end-caps were press-fitted onto the segments.

TEST CONDITIONS

In each test, the sample was heated to a temperature of 1500°C or above in flowing $\text{Ar}/2\%\text{H}_2$ (tests H01, H02 and H03) or $\text{Ar}/4\%\text{H}_2$ (tests H04, H05 and H06) to prevent premature oxidation of the sample during the gradual heating of the furnace. The oxidizing atmosphere was added at a temperature T_{ox} for an interval of time t_{ox} ; these values for each test are given in Table 1, together with the peak temperature and test atmosphere used in each test. Steam was added to the Ar/H_2 carrier gas stream in tests H01, H02 and H06 at a low enough temperature to prevent excessive formation of molten material, which might have compromised furnace tube integrity. Air was substituted for Ar/H_2 carrier gas in tests H02 and H05 at temperatures above 1500°C to prevent excessive sample degradation by U_3O_8 formation. In each test, the sample was cooled from test temperature to room temperature in flowing Ar/H_2 .

In test H01, the furnace tube broke on reaching peak sample temperature. The steam flow was shut off and the flow rates of inert gas inside and outside the furnace tube were increased to maintain the inertness of the atmosphere at the sample. The furnace temperature was then reduced.

In test H04, the atmosphere was effectively argon containing variable amounts of hydrogen and steam; oxygen entered into the annulus surrounding the furnace tube via the breaks in the segmented

zirconia guide tube, then permeated through the zirconia furnace tube. Calculations using the oxygen sensor readings indicate that the $H_2O:H_2$ ratio in the outlet gas was 1:1 at a sample temperature of 1500°C and about 5:1 while the sample temperature was above 1800°C.

DATA ANALYSIS

In this experiment, the direct-viewing γ -spectrometer (Figure 1) detected Y-91, Zr-95, Nb-95, Ru-103, Ru-106, Te-129m, I-131, Cs-134, Cs-137, Ba-140, La-140, Pr-144 and Eu-156. To improve the counting statistics for I and Cs isotopes, spectra accumulated over 300 s periods were added together to give total spectrum collection times of ~1200 s. Release percentages were normalized using selected γ -ray peaks of La-140 and Zr-95 to correct for attenuation and sample geometry changes [1]. The release percentages for Te-129m were calculated from the count rate changes of the 696 keV peak, which contained contributions from Te-129 and Pr-144.

The gas-monitoring γ -spectrometer detected peaks from Kr-85, Xe-131m and Xe-133. The initial calculated percentage releases of the noble-gas isotopes varied widely, and were not very similar to the Cs-134 percentage release, probably due to systematic errors in γ -spectrometer efficiency calibration. Since noble gases and cesium showed similar release behaviour and have shown similar release percentages in previous experiments [1,2], the noble-gas releases in each test were scaled to give the same final cumulative percentage release as Cs-134.

RESULTS

Ceramography of the fuel samples after high-temperature testing showed full oxidation of the sheath in all the tests including H04, which was conducted in nominally-inert atmosphere. Most of the sheath oxidation was from the outside. The only remaining metallic Zircaloy was a small island in the middle of an end-cap on the H06 sample.

The fission-product release behaviours of several isotopes are shown for tests H01 through H06 in Figures 2 through 7, respectively. The fractional release behaviours of Kr and the two Xe isotopes were almost identical to the behaviour of Cs. The beginning of rapid Xe-133 release in each test was within 300 s (one spectrum interval) of the beginning of rapid Cs-134 release. The I-131 releases exhibited most of the same characteristics as the Cs-134 releases, taking into account the poorer counting statistics of I-131 in these samples.

The release curves for the 696 keV peak (Figures 2, 3 and 4), which includes a contribution from Te-129m, have not been processed to remove the Pr-144 contribution; the curves are included only to show the timing of Te-129m releases. Most of the tellurium was probably released in tests H01 and H02 (Table 1), and statistically significant releases occurred during tests H03 and H06. Test H04, conducted in a less-oxidizing environment, and test H05, during which the oxidizing atmosphere was applied for a short time, showed less Te release. The beginning of Te release was approximately coincident with the beginning of rapid Cs release. The calculated final Te release (Table 1) in most of these tests was larger than expected (significantly greater than 100% in one case), indicating that the

inventory calculations or the 696 keV γ -yield may be inaccurate for one or both of Pr-144 and Te-129m.

In the two high-temperature steam tests H01 and H03, Ru-103 showed 4% release. A large fraction of the Ru was released in the high-temperature air test H02. The very small releases of Ru in the "inert" atmosphere test H04, and the lower-temperature air tests could only be observed by deposition measurements.

Of the fission products detected by direct-viewing γ -spectrometry, releases of Y-91, Pr-144 and Eu-156 could not be observed by direct-viewing or scanning γ -spectrometry for any of the test conditions used. The releases of Zr-95 and Nb-95, and Ce-144 were <1% (Table 1). The releases of Ba were between 1% and 7% in all of the high-temperature tests (H01 through H04).

DISCUSSION

The release behaviour of iodine, cesium and noble gases exhibited considerable similarity at elevated temperatures. No significant releases of Cs were observed below $\sim 1500^\circ\text{C}$ in inert environment in the HCE3 experiment, consistent with the low gap inventory of the fuel. In previous experiments, releases of Cs in inert atmosphere at temperatures below 1400°C have been tentatively attributed to gap inventory [2]; the fuels used in those experiments had noble-gas gap inventories ranging from 4 to 24%, indicating high Cs gap inventories. The lower Cs release during inert heating may also be due to the lower heating rates in the temperature range 1000 to 1500°C (0.1 to 0.3°C/s), compared to $0.8\text{--}6^\circ\text{C/s}$ in previous tests.

Rapid Cs release was delayed after the addition or ingress (e.g., test H04) of oxidizing atmosphere until oxidation of the sheath was complete. The oxidative cesium release rates in steam from the HCE3 and UCE12 experiments [2] were not very temperature-dependent compared to the CORSOR-M fission-product-release correlation [5], which is used in light-water-reactor safety analysis (Figure 8). Our experimental data would be better simulated using a pair of correlations to assign different release rates before and after full sheath oxidation.

The timing of Te-129m release indicates that complete sheath oxidation is required for Te release. The final fractional releases of Te were probably similar to those of Cs, allowing for inaccuracies related to inventory calculations.

The solid fraction of fission-product Ru would have been present almost entirely in the metallic state at the temperatures and $P(\text{O}_2)$ values in the HCE3 experiment, and release probably occurred by oxidation to RuO_x gaseous species [6]. As observed previously for fragment samples [2], release of Ru in test H02 (and probably also in tests H01 and H03) began a significant length of time ($>200\text{s}$) after oxidative release of Cs began. This delay was due to the competition among Zircaloy, UO₂ and Ru for oxygen. Test H02 showed the largest percentage release of Ru from any complete Zircaloy-sheathed sample tested at CRL, due to the high test temperature and the comparatively long exposure to air.

Several fission products (Y, Zr, Nb, Ce, and Eu) that showed release by matrix stripping in fragment tests under these conditions [3,4] showed low (<1%) or no fractional release in the HCE3 sheathed-fuel tests. The fraction of the UO₂ volatilized and consequent fractional release by matrix stripping was much less in this experiment than in the fragment tests. The oxidized Zircaloy sheathing would also tend to decrease the amount of UO₂ volatilized by limiting the mass transport of steam and fission-product compounds.

CONCLUSIONS

In each test, the release behaviour of Cs, I and noble gases were very similar to each other. Very little Cs was released before complete oxidation of the sheath, probably because this fuel had a small Cs gap inventory; the measured gap inventory of noble gases was <1% after irradiation. These results suggest that the releases observed in inert atmosphere below 1400°C or before complete sheath oxidation in other experiments were releases of gap inventory. The release rates during UO₂ oxidation in steam showed very little temperature dependence. Release of Te began at about the same time as oxidative release of Cs. Two high-temperature steam tests (H01, 1930°C and H03, 1840°C) showed a release of ~4% of Ru. An air test at 1890°C (H02) showed the greatest release of Ru for a sheathed sample with two end-caps. Ruthenium release began well after oxidative release of volatile fission products (delay of >2000 s).

The high level of oxygen permeation into the furnace tube during the nominally-inert test H04 was probably due to the gaps in the segmented guide tube. Future apparatus should employ a singlepiece guide tube to maintain the inertness of test atmospheres at high temperatures.

REFERENCES

1. COX, D.S., LIU, Z., ELDER, P.H., HUNT, C.E.L. and ARIMESCU, V.I., 'Fission-Product Release Kinetics from CANDU and LWR Fuel during High-Temperature Steam Oxidation Experiments,' Fission Gas Release and Fuel Rod Chemistry Related to Extended Burnup, IAEA-TECDOC-697 (1993).
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TABLE 1: EXPERIMENT CONDITIONS AND FISSION-PRODUCT RELEASES (PERCENTAGE OF INVENTORY) IN HCE3 TESTS.

Test	H01	H02	H03	H04	H05	H06
Peak T (°C)	1930	1890	1840	1830	1510	1540
T _{ox} (°C)	1500	1520	1750	(1)	1510	1540
t _{ox} (s)	8500	8740	5360	(1)	2900	8500
Oxidizing Atmosphere	90%steam/ 10% Ar/ 0.2% H ₂	air	87%steam/ 13% Ar/ 0.3% H ₂	steam/ 96% Ar/ H ₂	air	86%steam/ 13% Ar/ 0.6% H ₂
Zr-95	0.01 ⁽²⁾	0.36 ⁽²⁾	(3)	(3)	(3)	(3)
Nb-95	0.25 ⁽²⁾	0.81 ⁽²⁾	0.48 ⁽²⁾	0.02 ⁽²⁾	(3)	(3)
Ru-103	4.2	86.	3.5 ⁽²⁾	0.02 ⁽²⁾	0.02 ⁽²⁾	(3)
Ru-106	6	89.	3.7 ⁽²⁾	(3)	(3)	(3)
Ag-110m	81. ⁽²⁾	14. ⁽²⁾	50. ⁽²⁾	13. ⁽²⁾	9.5 ⁽²⁾	37. ⁽²⁾
Sn-113	35. ⁽²⁾	(3)	27. ⁽²⁾	(3)	27. ⁽²⁾	44. ⁽²⁾
Sb-125	41. ⁽²⁾	30 ⁽²⁾	45. ⁽²⁾	(3)	6.2 ⁽²⁾	(3)
Te-129m	153 ± 31	198 ± 41	88 ± 29	30. ⁽²⁾	13. ⁽²⁾	64. ⁽²⁾
I-131	86.	100.	77.	36.	14.	56.
Cs-134	85.	98.9	74.	29.	14.	56.
Cs-137	104.	98.6	73.	27.	13.	54.
Ba-140	5.1 ⁽²⁾	3.6 ⁽⁴⁾	2.2 ⁽²⁾	1.9 ⁽²⁾	(3)	0.02 ⁽²⁾
Ce-141	0.02 ⁽²⁾	(3)	0.01 ⁽²⁾	0.01 ⁽²⁾	(3)	(3)
Ce-144	0.06 ⁽²⁾	-	0.01 ⁽²⁾	0.02 ⁽²⁾	-	-

¹ Oxidizing atmosphere was not added deliberately; significant oxygen permeation occurred at >1500°C.

² This value was calculated from deposition and aerosol collector observations.

³ Deposition measurements either did not detect the isotope, or found <0.01% of the sample inventory of the isotope.

⁴ The value of Ba-140 release for this test was inferred from La-140 measurements.

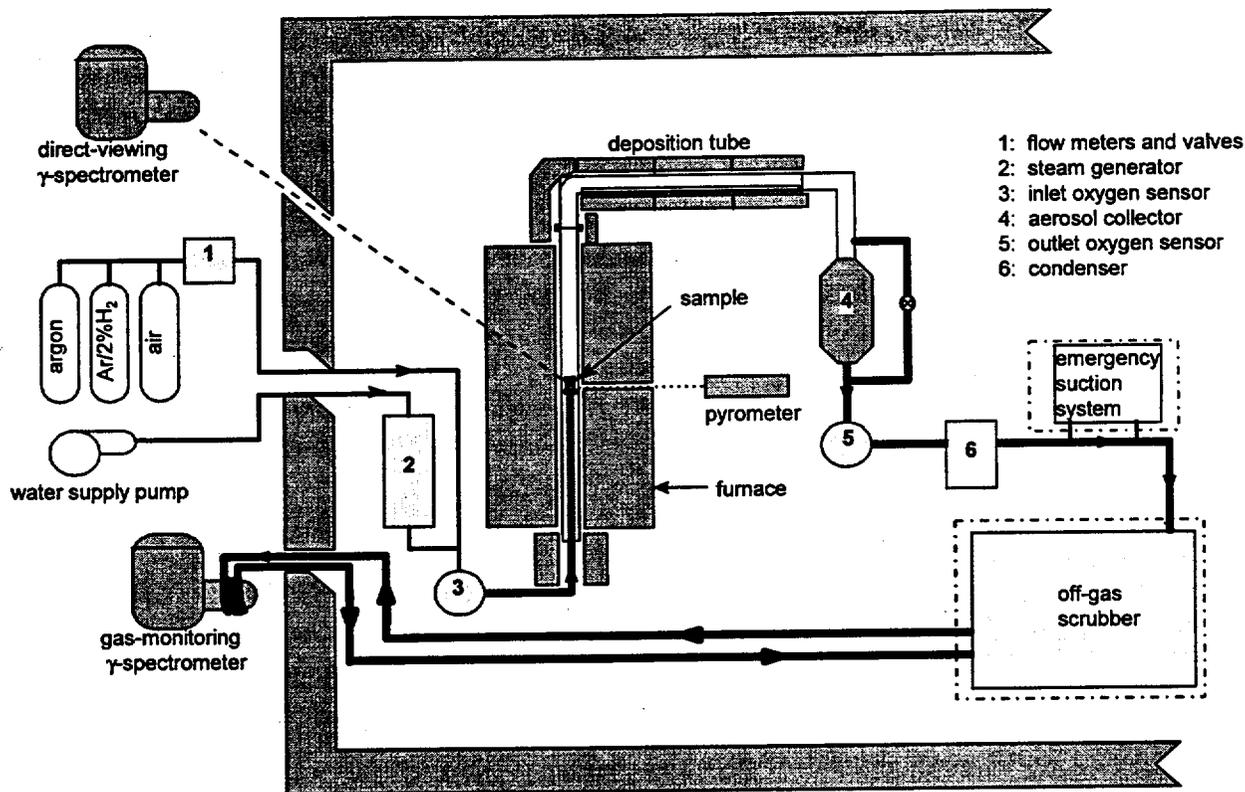


FIGURE 1. SCHEMATIC OF HCE3 HOT-CELL APPARATUS.

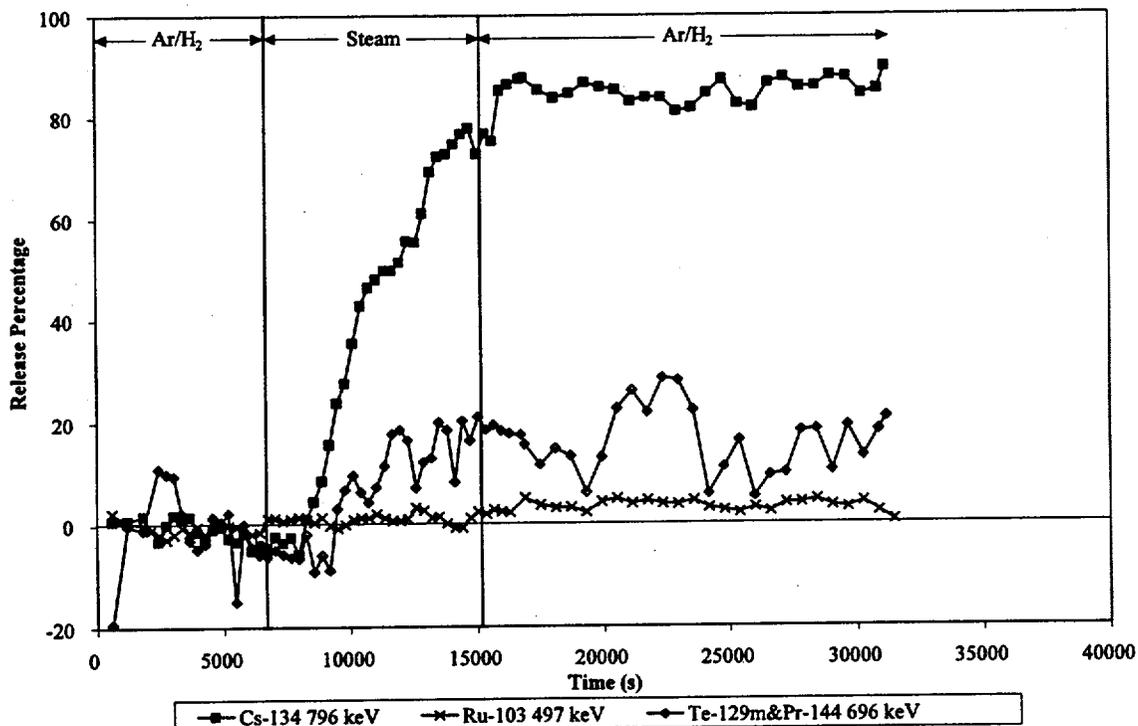


FIGURE 2: CESIUM AND RUTHENIUM RELEASE PERCENTAGES AND 696 keV "RELEASE PERCENTAGE" (Te-129m + Pr-144) FOR TEST H01 (STEAM, 1930°C).

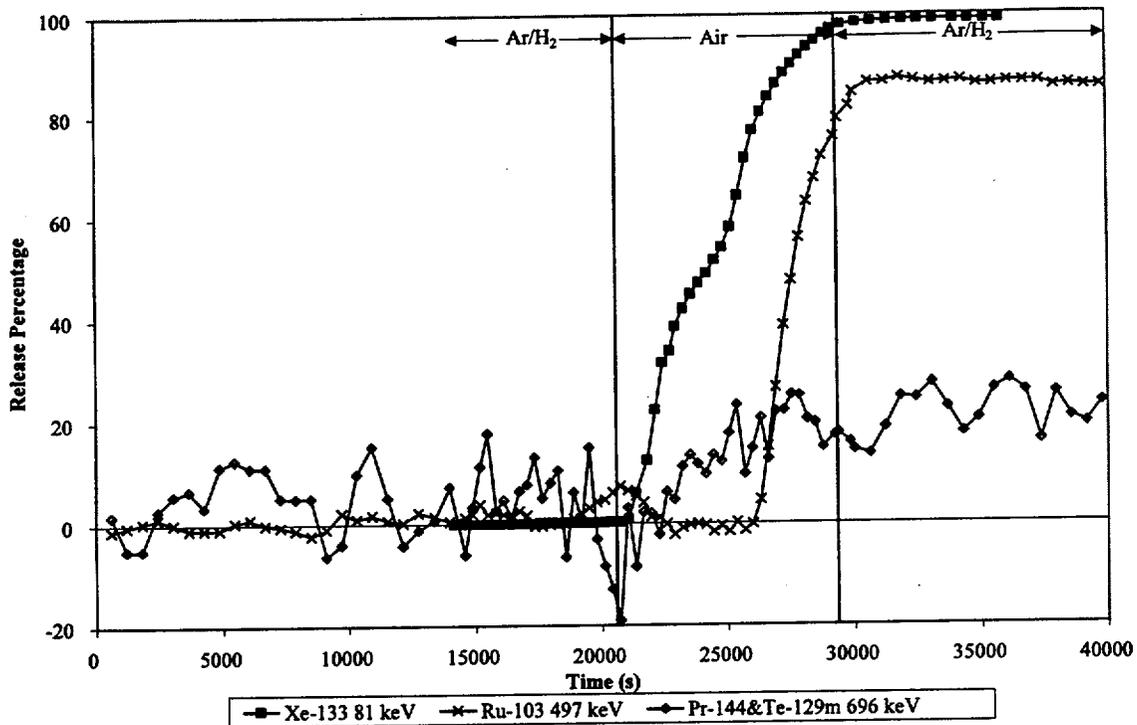


FIGURE 3: XENON AND RUTHENIUM RELEASE PERCENTAGES AND 696 keV "RELEASE PERCENTAGE" (Te-129m + Pr-144) FOR TEST H02 (AIR, 1890°C).

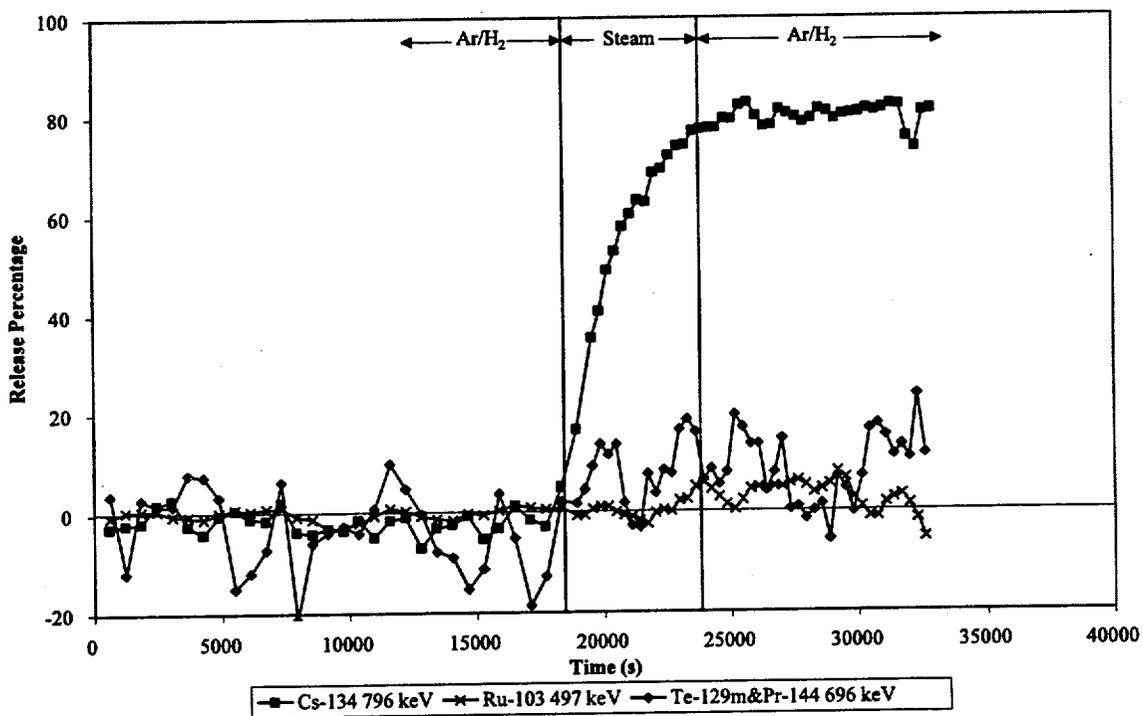


FIGURE 4: CESIUM AND RUTHENIUM RELEASE PERCENTAGES AND 696 keV "RELEASE PERCENTAGE" (Te-129m + Pr-144) FOR TEST H03 (STEAM, 1840°C).

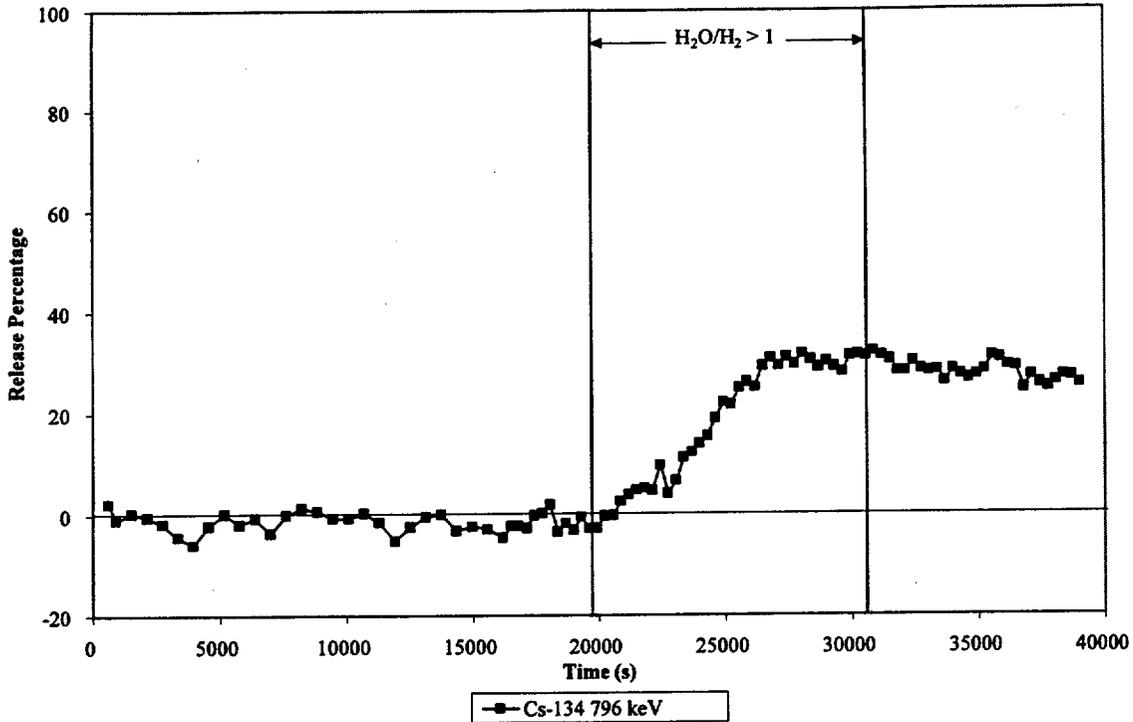


FIGURE 5: CESIUM RELEASE PERCENTAGE AS A FUNCTION OF TIME FOR HCE3 TEST H04 (SHEATHED DARLINGTON FUEL, $Ar/H_2/STEAM$, 1830°C).

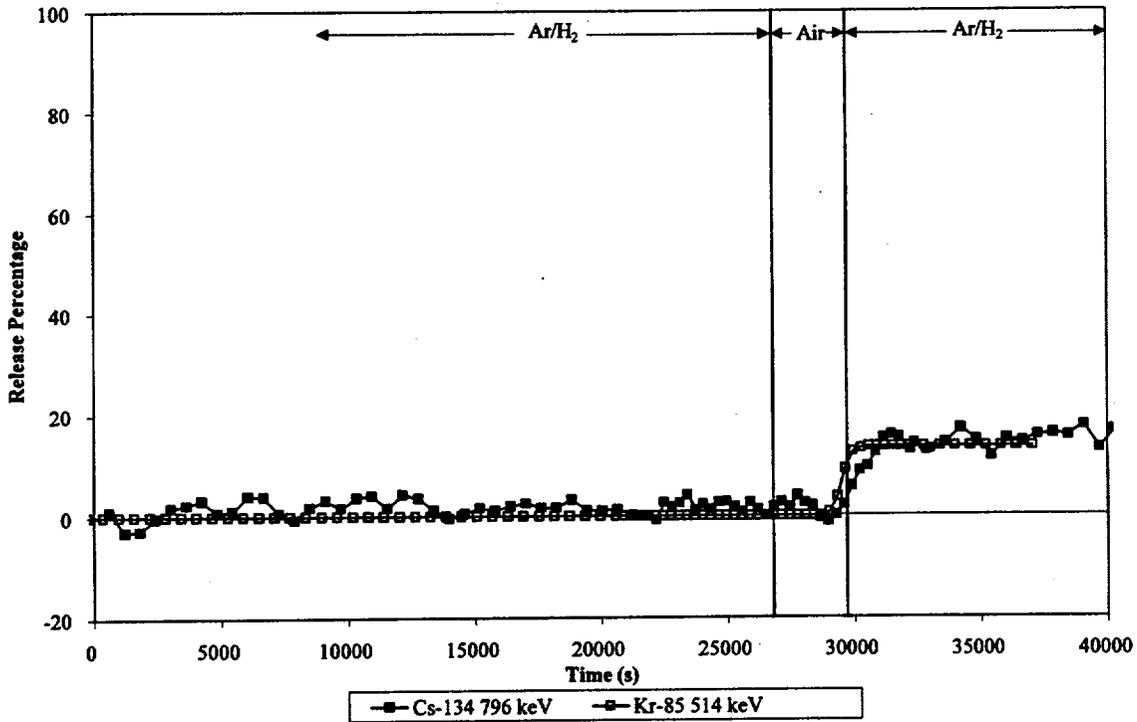


FIGURE 6: CESIUM AND KRYPTON RELEASE PERCENTAGES AS FUNCTIONS OF TIME FOR HCE3 TEST H05 (SHEATHED DARLINGTON FUEL, AIR, 1510°C).

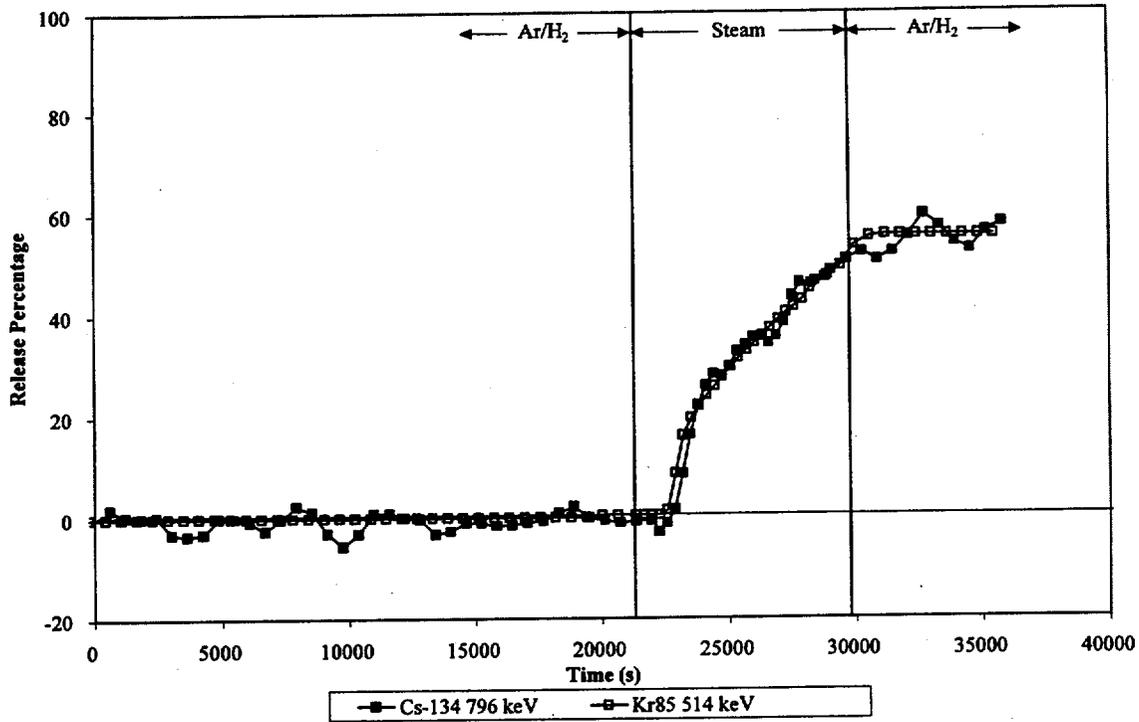


FIGURE 7: CESIUM AND KRYPTON RELEASE PERCENTAGES AS FUNCTIONS OF TIME FOR HCE3 TEST H06 (SHEATHED DARLINGTON FUEL, STEAM, 1540°C).

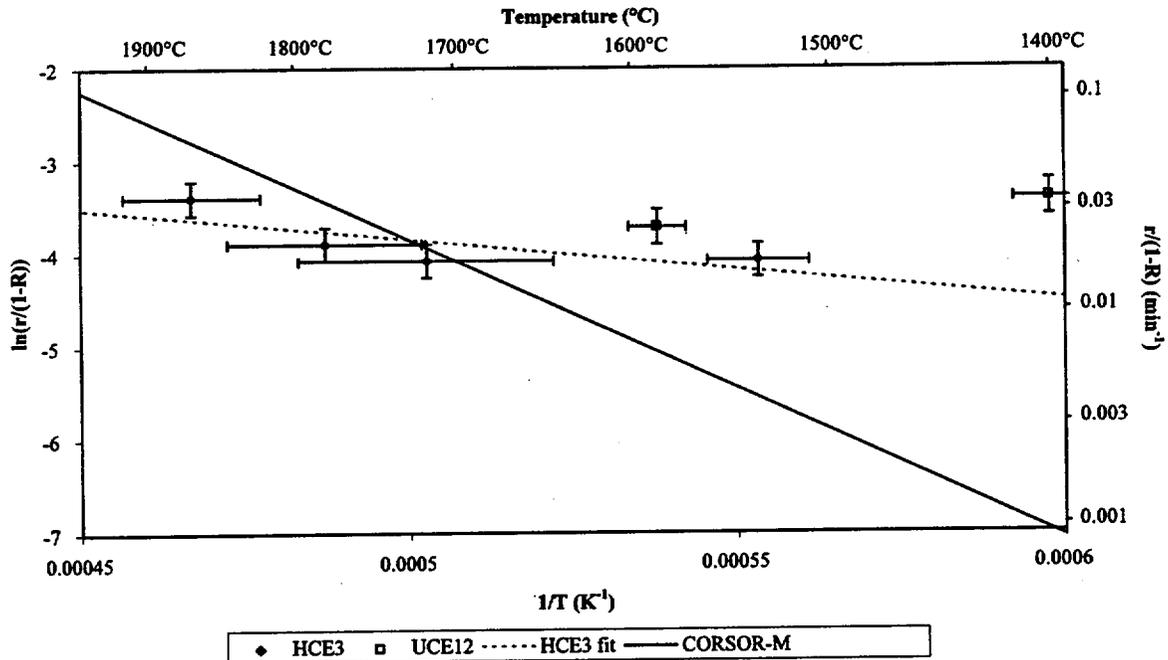


FIGURE 8: ARRHENIUS PLOT OF Cs RELEASE RATE AFTER FULL SHEATH OXIDATION (r) DIVIDED BY REMAINING FRACTION OF Cs INVENTORY ($1-R$) FOR SHEATHED-FUEL STEAM TESTS (HCE3 and UCE12 EXPERIMENTS)