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Data Summary Report for Fission Product Release Test VI-5

Prepared by
M. F. Osborne, R. A. Lorenz, J. R. Travis,
C. S. Webster, J. L. Collins

Oak Ridge National Laboratory

Prepared for
U.S. Nuclear Regulatory Commission

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M. F. Osborne, R. A. Lorenz, J. R. Travis,
C. S. Webster, J. L. Collins

Oak Ridge National Laboratory
Operated by Martin Marietta Energy Systems, Inc.

Oak Ridge National Laboratory
Oak Ridge, TN 37831-6285

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ABSTRACT

Test VI-5, the fifth in a series of high-temperature fission product release tests in a vertical test apparatus, was conducted in a flowing mixture of hydrogen and helium. The test specimen was a 15.2-cm-long section of a fuel rod from the BR3 reactor in Belgium, which had been irradiated to a burnup of ~42 MWd/kg. Using a hot cell-mounted test apparatus, the fuel rod was heated in an induction furnace under simulated LWR accident conditions to two test temperatures, 2000 K for 20 min and then 2700 K for an additional 20 min. The released fission products were collected in three sequentially operated collection trains on components designed to measure fission product transport characteristics and facilitate sampling and analysis.

Based on fission product inventories measured in the fuel or calculated by ORIGEN2, analyses of test components showed total releases from the fuel of ~100% for ^{85}Kr , ^{134}Cs , and ^{137}Cs ; 18% for ^{125}Sb ; and 57% for ^{154}Eu . Most (>93%) of the released cesium and antimony escaped from the furnace to the collection system, but a large fraction (55%) of the released europium was deposited on the furnace ceramics. Almost 3% of the iodine was collected in a volatile form. In the hydrogen atmosphere, the Zircaloy cladding melted, ran down, and reacted with the UO_2 and some of the fission products. Gamma ray monitoring of the fuel specimen during and after the test showed that the fuel collapsed at a temperature >2400 K. Including fission products and also fuel and structural materials, the total mass released from the furnace to the collection system was ~0.681 g, which was much less than the mass released from a similar test in steam atmosphere. The results from this test were compared with those obtained in previous tests in this series and with the CORSOR-M and ORNL diffusion release models for fission product release.



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FOREWORD

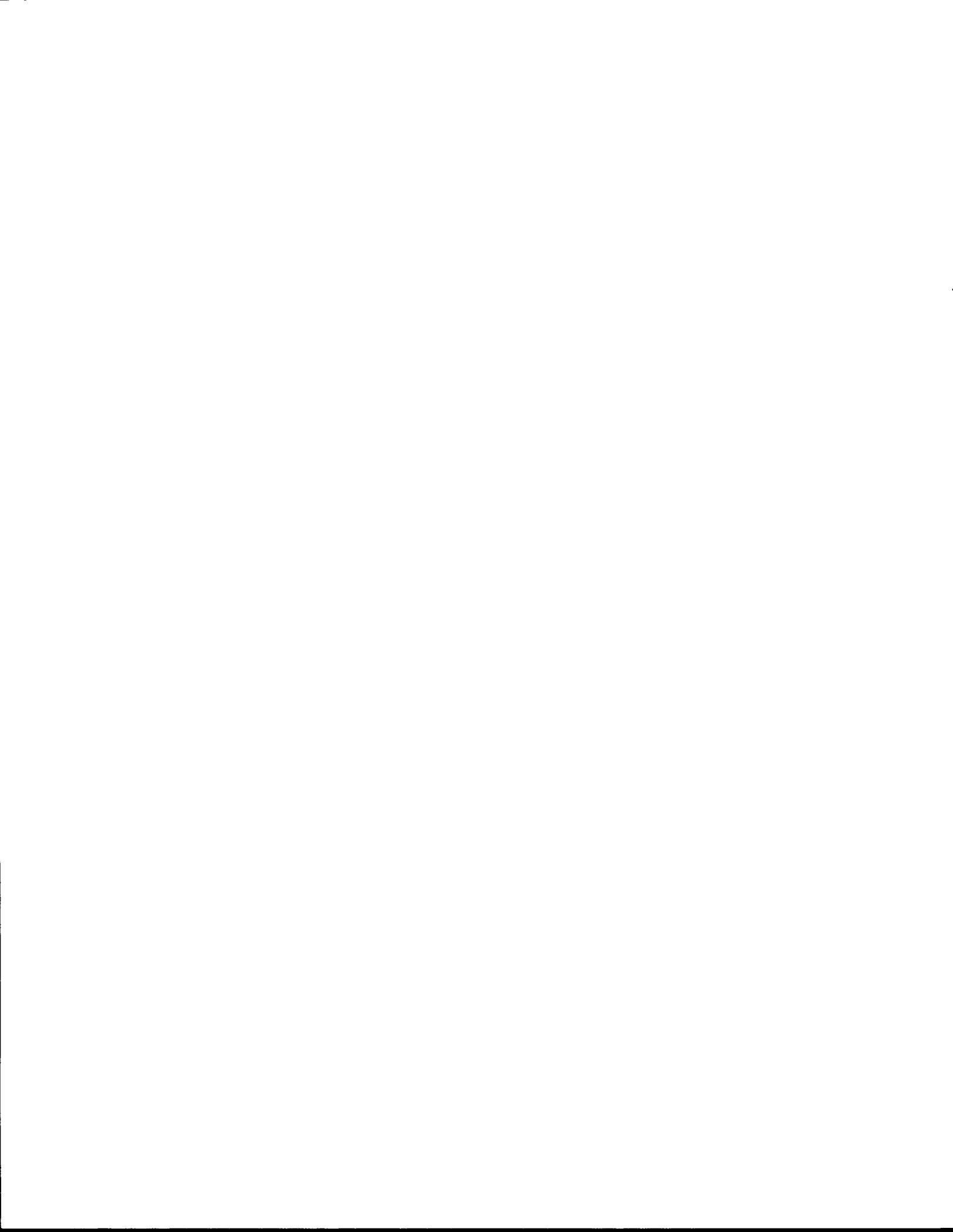
This document describes the tenth in a series of recent fission product release tests of commercial LWR fuel under severe accident conditions. Other reports describing the work conducted in this fission product release project are:

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6. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, *Data Summary Report for Fission Product Release Test HI-6*, NUREG/CR-4043 (ORNL/TM-9943), September 1984.
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DATA SUMMARY REPORT FOR FISSION PRODUCT RELEASE TEST VI-5

M. F. Osborne
R. A. Lorenz
J. R. Travis
C. S. Webster
J. L. Collins

1. EXECUTIVE SUMMARY

The objective of this report is to document as completely as possible the observations and results of fission product release test VI-5, which was performed May 18, 1989. Although all final data have not been obtained, this report presents all currently available results for potential use by other reactor safety researchers. Complete interpretation and correlation of these results with related experiments and with theoretical behavior will be included in a subsequent report, which will consider the results of several tests over a range of test conditions. Similar data summary reports for previous tests in this project, as well as other reports of related project activities, are listed in the Foreword.

The fuel specimen used in this test was cut from fuel rod I-114, which had been irradiated in the BR3 reactor in Belgium from July 15, 1976, to September 26, 1980. The specimen contained 71.25 g uranium and had been irradiated to a burnup of ~42 MWd/kg uranium. The fission product inventories, as measured in the fuel and calculated by ORIGEN2, and a description of the test procedure and conditions are included in Sect. 3. This test in a hydrogen atmosphere experienced the same temperature history as test VI-3 did in steam, allowing a direct comparison of fission product release and behavior in the two different atmospheres. The test results and some preliminary interpretations are presented in Sect. 4, and these results are compared with data from previous tests in Sect. 5. The most important results are:

1. Posttest examination, in agreement with on-line radioactivity measurements, indicated that the Zircaloy cladding had melted during the test and that the 15-cm-long specimen had partially collapsed sometime after the temperature had reached ~2300 K.
2. The values for total fractional release from the fuel specimen, based on ORIGEN2 calculations and gamma ray spectrometry measurements, were ~100% for ^{85}Kr , 18% for ^{125}Sb , ~100% for both ^{134}Cs and ^{137}Cs , and 57% for ^{154}Eu . About 6.7% of the released ^{125}Sb and 55% of the released ^{154}Eu were deposited on the ThO_2 and ZrO_2 ceramics at the outlet end of the furnace, compared to almost complete transport of the released Kr and Cs from the furnace to the collection system. Pretest and posttest gamma ray spectrometry of the fuel was valuable in determining the release fractions as well as the axial distributions of the fission products within the fuel rod and the furnace.

3. Compared to earlier tests in steam, a much larger fraction (~65% vs ~30%) of the released cesium deposited on the thermal gradient tubes (TGTs), where primarily vapors are condensed, rather than on the filters, where the deposits are mostly aerosols. This result confirmed earlier indications from test VI-4 that, in hydrogen atmosphere tests, most of the cesium is transported in vapor forms.
4. The fission product distributions in the TGTs showed very high concentrations of cesium at several locations in the liners from tubes A and B. The main peak in tube A occurred at ~895 K (620°C). Three large cesium peaks were concentrated at deposition temperatures of ~1000, ~1050, and ~800 K in tube B. The peaks or shoulders at ~795 K (520°C) almost certainly contained CsI along with other cesium species, and the higher temperature peaks probably contained more complex cesium compounds. Because TGT C could not be removed from the surrounding Inconel tube, much less detailed data were obtained from it.
5. The total mass of deposits on the TGTs and filters was determined by direct weighing to be 0.681 g, with ~73% of this (0.497 g) being deposited on the filters. The mass release rates were highest during the heatup to the high-temperature (2700 K) test period. The maximum aerosol concentration, averaged over the collection period, was ~36 g/m³ at 423 K (150°C) during test Phase B. It should be noted that the test configuration did not include reactor structural or control materials which could add significant aerosol masses during an accident.
6. Comparison of the release data from this test with the results of previous tests provided clear evidence of the influence of atmosphere (oxidizing and reducing) on some of the fission products, notably antimony and europium. Compared with tests in steam, the release of antimony was strongly reduced and the release of europium was strongly increased in test VI-5 in hydrogen. However, no significant influence of atmosphere on the releases of krypton and cesium was found.
7. In comparison with the release rates calculated by the CORSOR-M model, measured release rates for krypton and cesium were somewhat lower in test VI-5, but not as low as in the earlier test VI-3, in the temperature range of 2200 to 2700 K.
8. Approximately 2.85% of the released iodine was collected in a volatile form. This may be an effect of the hydrogen atmosphere and the unoxidized stainless steel liner in TGT A.

2. INTRODUCTION

This report summarizes data from the fifth test in a vertical test apparatus. This series of tests is designed to investigate fission product release from light-water reactor (LWR) fuel in steam and/or

hydrogen in the temperature range of 2000 to 2700 K. Earlier tests, conducted under similar conditions in a horizontal furnace at temperatures of 1675 to 2275 K (1400 to 2000°C), were reported previously.¹⁻⁶ The purpose of this work, which is sponsored by the U.S. Nuclear Regulatory Commission (NRC), is to obtain the experimental data needed to reliably assess the consequences of a variety of heatup accidents in LWRs.⁷ The specific objectives of this program are:

1. to obtain fission product release and behavior data applicable to the analysis of reactor accidents, and
2. to apply these data to the development of VICTORIA and other release and transport models.

Tests of high-burnup LWR fuel are emphasized in this program. The applicability of simulated fuel (unirradiated UO₂ containing radioactive fission product tracers) has been considered, and several simulant tests have been conducted to provide valuable data about the behavior of specific fission product species.⁸ All tests have been conducted at atmospheric pressure in helium mixed with either steam or hydrogen in a hot cell-mounted test apparatus.

The procedures and techniques used in preparing and conducting the test, as well as in posttest examination and analysis, were very similar to those used in earlier tests. The analytical techniques employed are listed in Table 1. This report provides a brief description of test VI-5 and a compilation of all results obtained to date. Other analyses are still in process but have been delayed by the unavailability of special facilities and a shortage of funding. These results will be reported later. Thorough data evaluation and correlation of all results from the VI test series will be included in subsequent reviews and reports covering this series of fission product release tests at temperatures up to 2700 K.

3. TEST DESCRIPTION

The vertical test apparatus⁹ is operated remotely and is capable of conducting tests at temperatures up to 2700 K for time periods up to 60 min in reactive atmospheres, such as steam and/or hydrogen mixed with helium. Details of the furnace are shown in Fig. 1, and the entire test apparatus installed in the hot cell is illustrated in Fig. 2. Both manual and automatic optical pyrometers are used for temperature measurement, supplemented by thermocouples during the low-temperature heatup phase. The released fission products are collected in three sequentially operated, parallel collection trains, each composed of (1) a 7.6-mm-ID platinum or stainless steel-lined thermal gradient tube (TGT) for vapor condensation; (2) a filter package containing graduated fiberglass filters for aerosol collection and heated charcoal (KI impregnated) for iodine sorption; and (3) cold charcoal traps for rare gas collection. The on-line releases of ⁸⁵Kr and ¹³⁷Cs are monitored by detectors observing these collector components. (In steam atmosphere tests, a hot CuO bed is used to oxidize the hydrogen generated by

Table 1. Analytical techniques for fission product analysis

Technique	Time	Location	Elements
Gamma spectrometry	Pretest, posttest	Fuel specimen	Long-lived, gamma-emitting fission products - Ru, Ag, Sb, Cs, Ce, Eu
	On-line	Thermal gradient tube (TGT), charcoal traps, filters	Cs, Kr
	Posttest	Furnace components, TGT, filters	Ru, Ag, Sb, Cs, Ce, Eu
Neutron activation analysis	Posttest	Charcoal, solution from furnace, TGT, filters	I, Br, (Te, Mo, Ba) ^a
Chemical analysis	Posttest	Furnace, filters	U, Pu
Spark-source mass spectrometry	Posttest	Samples from furnace, TGT, filters	All elements with atomic numbers ≥ 10
Inductively coupled plasma-emission spectrometry	Posttest	Acid solutions from furnace, TGT, or filters	Many cations, especially Mo, Te, Ba, U
Scanning electron microscope with energy dispersive X-ray system	Posttest	TGT, aerosol sampler	All elements with atomic numbers ≥ 10

^aIodine and bromine have been analyzed by neutron activation throughout HI and VI test series; neutron activation analysis techniques for Te, Mo, and Ba are in various stages of development.

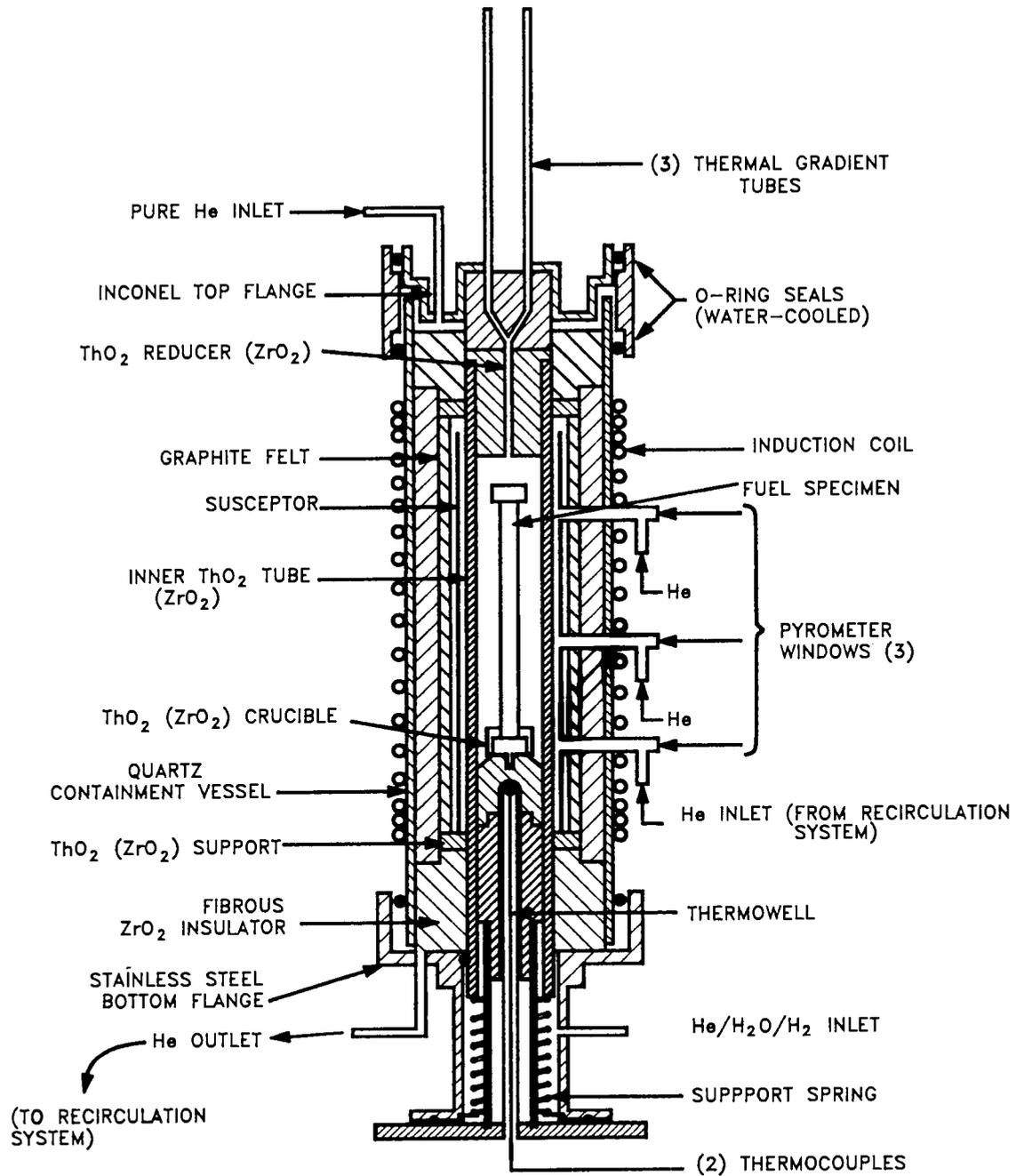


Fig. 1. Vertical fission product release furnace, as used in test VI-5.

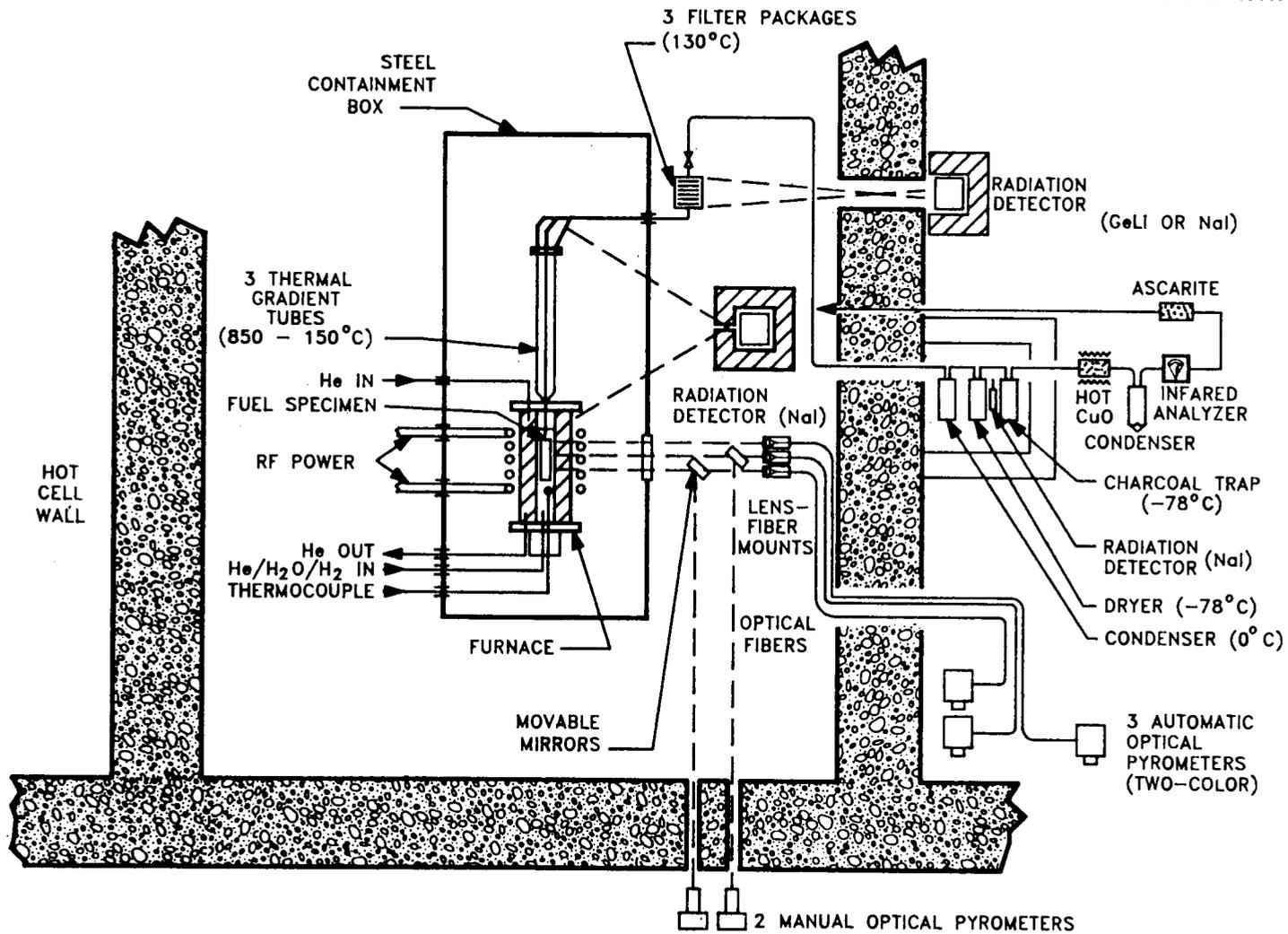


Fig. 2. Vertical fission product release apparatus.

reaction of steam with the Zircaloy cladding. The collection of this water in a condenser is measured continuously by a modified conductivity meter, thereby indicating the oxidation rate of the cladding.) Details of the fission product collection system are shown in Fig. 3. Included in the test VI-5 apparatus, but not shown in Fig. 2, were three radiation detectors that were collimated to monitor the radioactivity (primarily ^{134}Cs and ^{137}Cs) in the top, middle, and bottom regions of the fuel specimen. Data from these instruments detected the movement (collapse) of the fuel.

The tests planned for this vertical apparatus assumed that temperature (2000 to 2700 K) was the dominant variable. The flow rate of reactive gas (0.3 to 1.5 L/min) and time (1 to 60 min) were considered secondary variables. The objectives of this particular test were to obtain release rate data from BR3 fuel heated at temperatures of 2000 and 2700 K in hydrogen. The fuel specimen was maintained at each of the test temperatures for 20 min. Test VI-3 was conducted at the same conditions except in a steam atmosphere.

3.1 FUEL SPECIMEN DATA

The test specimen was a 15.2-cm-long section of rod I-114 from the BR3 reactor in Belgium, as shown in Fig. 4. This fuel was irradiated from July 15, 1976, until September 26, 1980. Details of the irradiation and of the characteristics of this particular specimen were reported by Adams and Dabell* and are listed in Table 2. The fuel in this rod had an initial enrichment of 5.76% ^{235}U , and the VI-5 specimen had attained a burnup of ~42 Mwd/kg during irradiation. Fission product inventories for the specimen were measured by direct gamma spectrometry of the fuel and were calculated with the ORIGEN2 computer program,¹⁰ with adjustments based on direct analyses for ^{137}Cs in the fuel; these data are shown in Tables 3 and 4.

No axial scan of the gamma radioactivity along the intact fuel rod was made before the rod was sectioned. Scans of nearby rods with similar operating histories, however, indicated that rod I-114 had experienced no unusual temperature, power, or burnup histories during irradiation. Reactor operating data indicated that this fuel rod had operated at a maximum linear power of 222 W/cm averaged over the 1-m length, indicating that the fuel in the peak burnup region (near mid-length) would have operated at a peak linear power of ~285 W/cm at that time. These data indicated that the fuel had not experienced high enough operating temperatures to cause significant migration of the fission product cesium during irradiation. The data shown in Fig. 5 were used to estimate fission gas release from the peak burnup region of a fuel rod. Using Fig. 5, our thin-slit gamma scan, which is shown in

*J. P. Adams and B. R. Dabell, Idaho National Engineering Laboratory, Idaho Falls, ID 83415, "Characteristics of $\text{UO}_2\text{-Zr}$ Fuel Rods Irradiated in the BR3 Reactor," private communication, 1986.

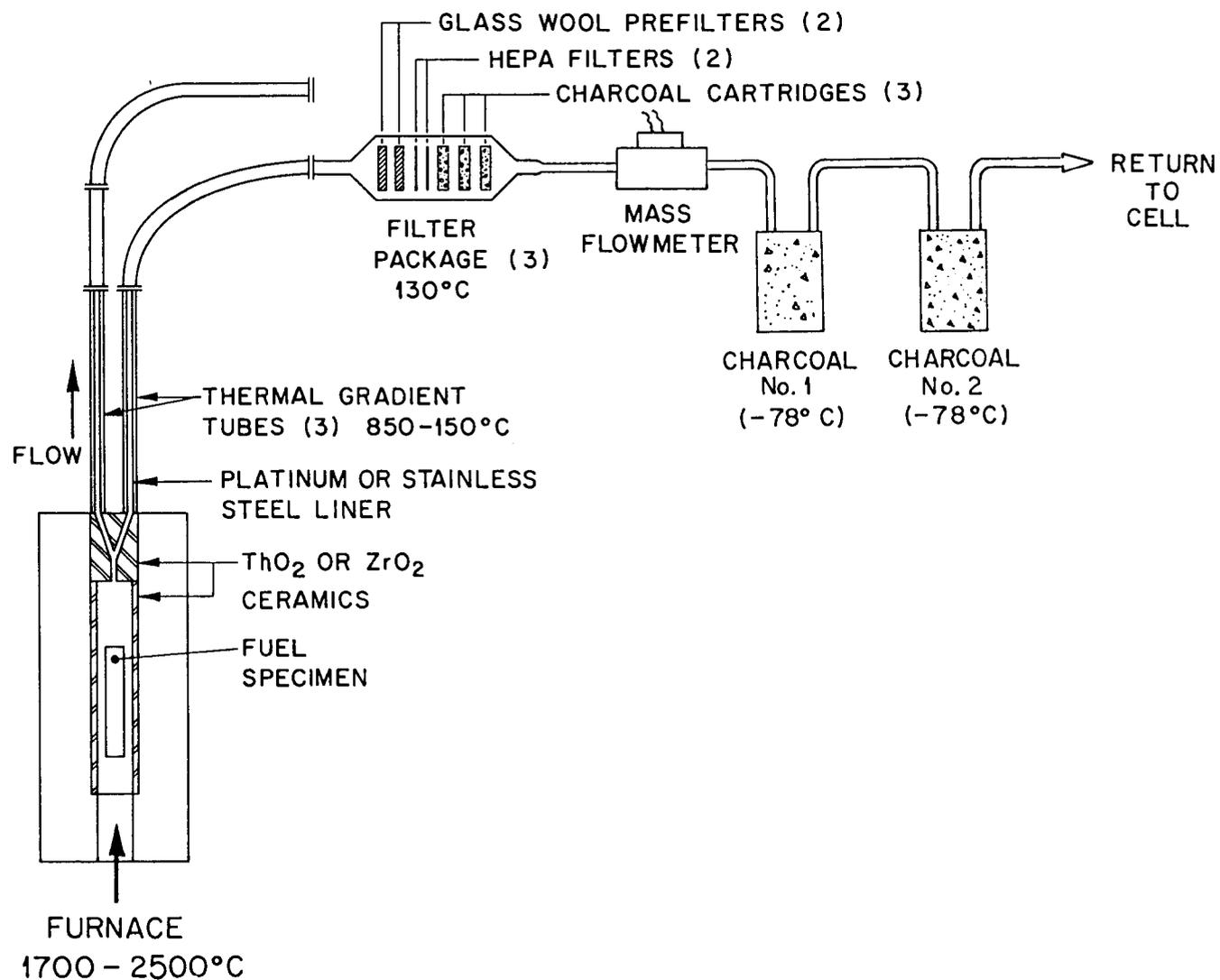


Fig. 3. Components of the fission product collection system.

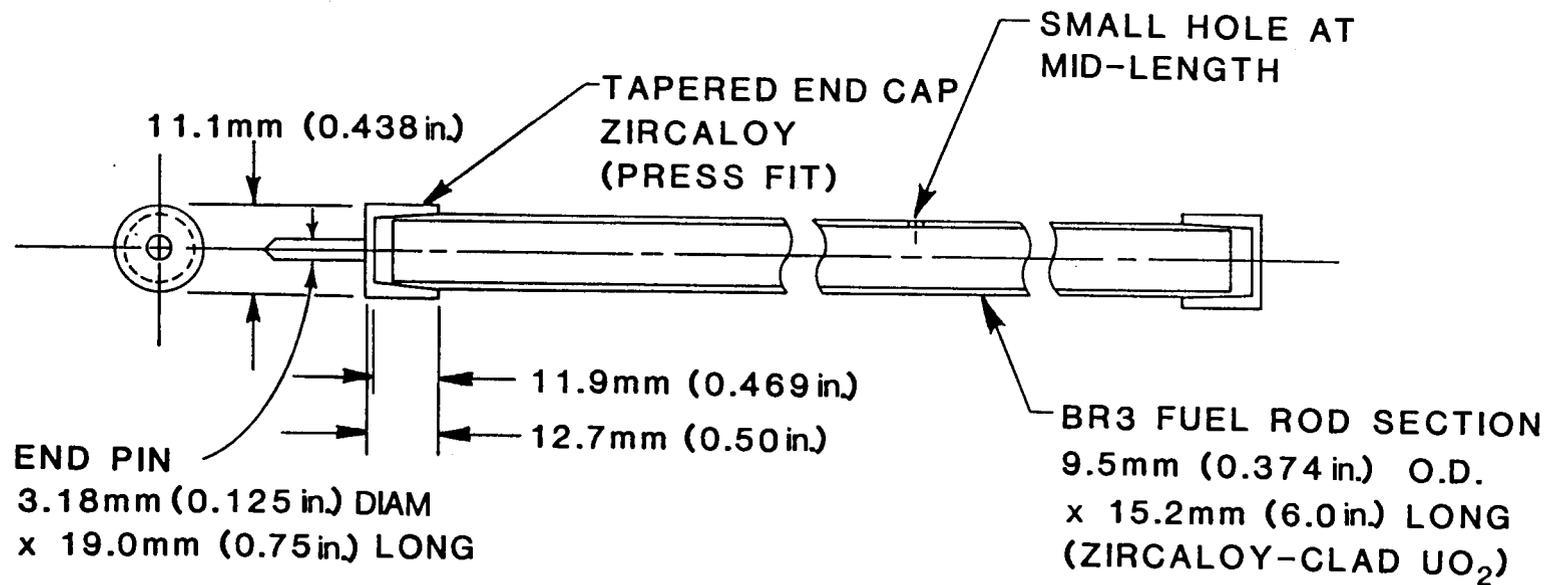


Fig. 4. Details of the fuel specimen heated in test VI-5.

Table 2. Data for fuel specimen used in test VI-5

Fuel specimen identification	Rod I-114 BR3 Reactor (Belgium)
Irradiation data	
Period	July 15, 1976, to Sept. 26, 1980
Fuel specimen burnup	42 MWd/kg U
Maximum heat rating	222 W/cm
Specimen data	
Length	15.2 cm
Fuel loading	80.8 g UO ₂ (71.25 g U)
Initial enrichment	5.76% ²³⁵ U
Total weight	102.0 g
Weight of Zircaloy	21.22 g
Gas release during irradiation	~2% (from specimen)

Table 3. Inventories of principal fission product elements
in test VI-5 fuel^a

Elements	Amounts in fuel (g/MTU)	Amounts in test specimen ^b (mg)
Se	75.70	5.414
Br	29.09	2.080
Kr	511.5	36.58
Rb	503.7	36.02
Sr	1,167	83.43
Y	664.5	47.52
Zr	4,940	353.4
Mo	4,306	307.9
Tc	997.2	71.32
Ru	2,534	181.2
Rh	533.7	38.17
Pd	1,225	87.64
Ag	61.40	4.39
Cd	86.93	6.22
In	2.55	0.182
Sn	88.52	6.33
Sb	20.74	1.483
Te	555.3	39.72
I	260.2	18.61
Xe	6,586	471.0
Cs	3,228	230.8
Ba	2,118	151.5
La	1,583	113.6
Ce	3,099	221.7
Eu	151.4	10.83
Total fission products	4,327	3,094
U	943,800	67,500
Pu	8,126	581.2
Total actinides	953,000	68,160

^aInventories for this BR3 fuel were calculated by ORIGEN2, assuming a burnup of 42,000 MWd/MTU, and were corrected for decay to July 1, 1986.

^bThe test VI-5 specimen contained 71.25 g U (80.8 g UO₂), which is equal to 7.125E-5 MTU before irradiation.

Table 4. Inventories of long-lived radionuclides in test VI-5 fuel^a

Radionuclides	Amounts in fuel		Amounts in test specimen ^b	
	(g/MTU)	(Ci/MTU)	(mg)	(mCi)
<i>Fission products</i>				
⁸⁵ Kr	21.71	8,519	1.55	609.3
⁹⁰ Sr	659.1	89,950	47.14	6,434
¹⁰⁶ Ru	1.78	5,972	0.13	427.2
^{110m} Ag	0.00	6.30	0.00	0.45
¹²⁵ Sb	2.57	2,659	0.18	190.2
¹²⁹ I	202.5	0.04	14.48	0.00
¹³⁴ Cs	16.02	20,730	1.15	1,482
¹³⁷ Cs	1,287	112,000	92.05	8,009
¹⁴⁴ Ce	1.64	5,226	0.12	373.8
¹⁵⁴ Eu	24.53	6,625	1.75	473.8
Total fission products	43,270	497,100	3,094	35,550
<i>Actinide elements</i>				
²³⁴ U	301.6	1.89	21.57	0.13
²³⁵ U	17,760	0.04	1,270	0.00
²³⁶ U	6,962	0.45	498.0	0.03
²³⁸ U	918,800	0.31	65,710	0.02
Total U	943,800	4.58	67,500	0.33
²³⁸ Pu	160.3	2,746	11.46	196.4
²³⁹ Pu	4,840	300.9	346.1	21.53
²⁴⁰ Pu	2,109	480.7	150.8	34.38
²⁴¹ Pu	737.1	75,970	52.72	5,433
²⁴² Pu	280.0	1.07	20.03	0.08
Total Pu	8,126	79,500	581.2	5,685
Total actinides	953,000	81,150	68,160	5,804

^aInventories for this BR3 fuel were calculated by ORIGEN2, assuming 42,000 MWd/MTU, and were corrected for decay to July 1, 1986. The inventory values used to calculate the releases of ¹²⁵Sb (96.32 mCi) and ¹³⁷Cs (8.196 Ci) were measured by gamma spectrometry.

^bVI-3 contained 71.52 g U, which is equal to 7.152E-5 MTU before irradiation.

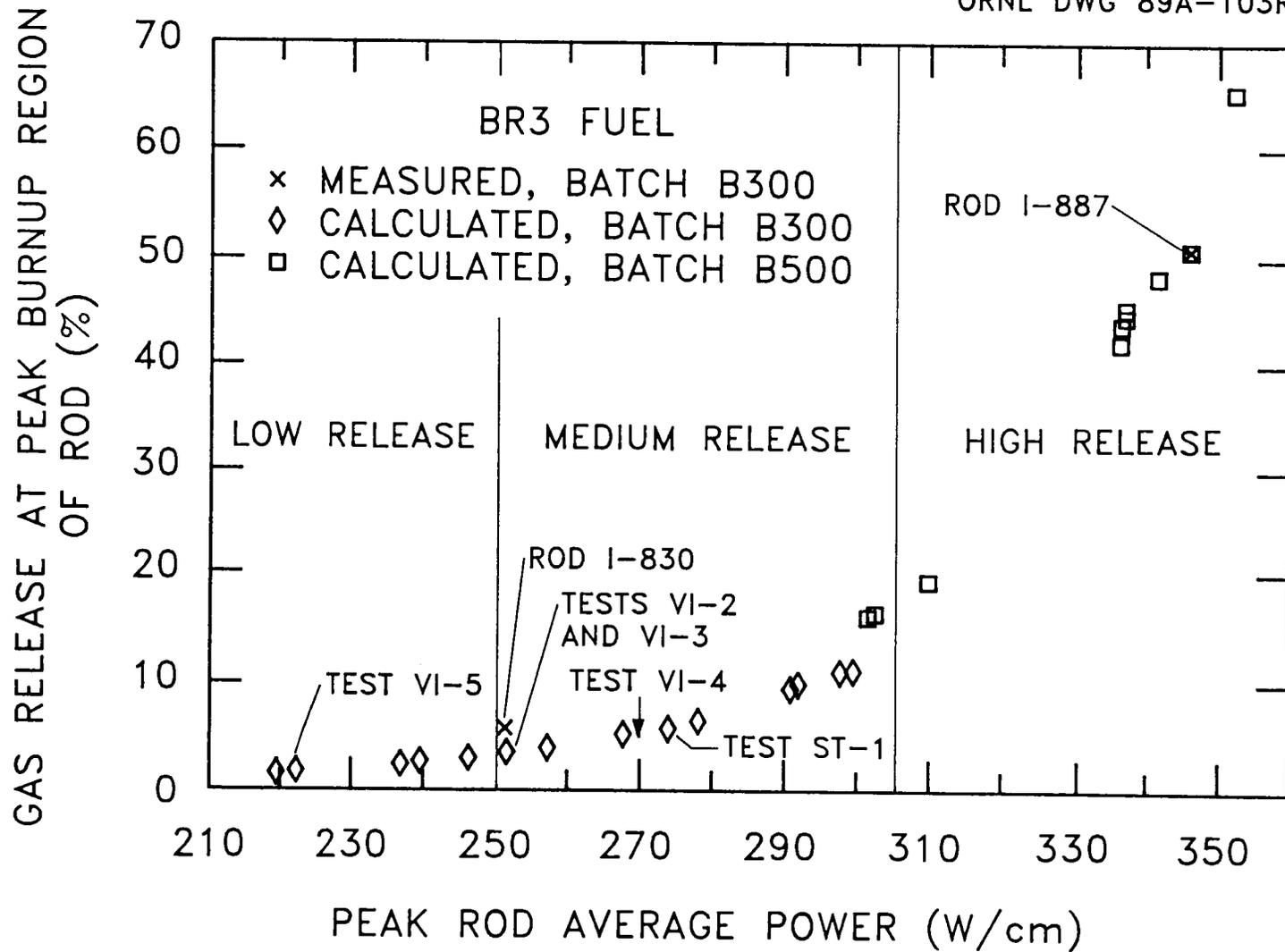


Fig. 5. In-reactor release of gaseous fission products as a function of power. The gas inside rods I-830 and I-887 was measured before sectioning.

Fig. 6, and the operating history, we estimate that ~2% of the total krypton generated in the fuel had been released during irradiation. The conversion from average gas release to release from the peak power (and peak burnup) location was made using the D' (empirical) method.¹¹

In addition to the test VI-5 fuel specimen, three other 15.2-cm-long specimens were cut from rod I-114 and prepared for future testing. Three short samples (1 to 2 cm long) were cut for metallographic examination, for dissolution and chemical analysis, and as an archive sample for possible future use.

Tapered end caps of Zircaloy-2 were pressed onto the ends of the test specimen, not as gas seals, but to prevent loss of the fractured UO₂ fuel during subsequent handling. The bottom end cap included a pin to facilitate vertical mounting. A small hole, 1.6 mm in diameter, was drilled through the cladding at midlength to serve as a standard leak for gas release during the heatup phase of the test. These details are shown in Fig. 4.

3.2 TEST CONDITIONS AND OPERATION

As in each of the previous experiments, the test apparatus was assembled by direct handling, which is possible because the hot cell and test apparatus are decontaminated after each test.

Also, new ceramic furnace internals, TGT liners of either platinum or stainless steel, and filter package components are used in each test. (The liner in TGT A was made of type 304 stainless steel foil, as opposed to platinum liners in TGTs B and C. In most previous tests, platinum liners were used to provide a relatively unreactive surface for the deposition of fission products, so that effects on the chemical forms of the deposits would be minimized. The stainless steel liner was used in test VI-5 in an effort to provide information about the effect of a realistic reactor material on the forms of the deposits.) Only the transfer and loading of the highly radioactive fuel specimen and the final closure of the furnace and containment box were done remotely. No in-cell operations were required during the test. Before heating was begun, the test apparatus was evacuated and purged with helium.

This test was intended to investigate fission product release at two temperatures (2000 and 2700 K) under strongly reducing conditions (hydrogen-helium atmosphere) to provide a direct comparison with test VI-3 in steam. The operating conditions are summarized in Table 5, and the temperature history (similar to test VI-3) is shown in Fig. 7. In tests VI-1 and VI-2, which used all ZrO₂ furnace ceramics, heatup rates of ~1 K/s were used. However, for the higher-temperature (2700 K) tests, VI-3 and VI-5, a ThO₂ furnace tube was required. Because of the greater susceptibility of ThO₂ to thermal shock at the lower temperatures, a slower heatup rate (~0.25 K/s) was necessary up to 2000 K. In test VI-5, the heatup rate from 2000 to 2700 K was increased to ~1.2 K/s, compared to ~0.3 K/s in test VI-3.

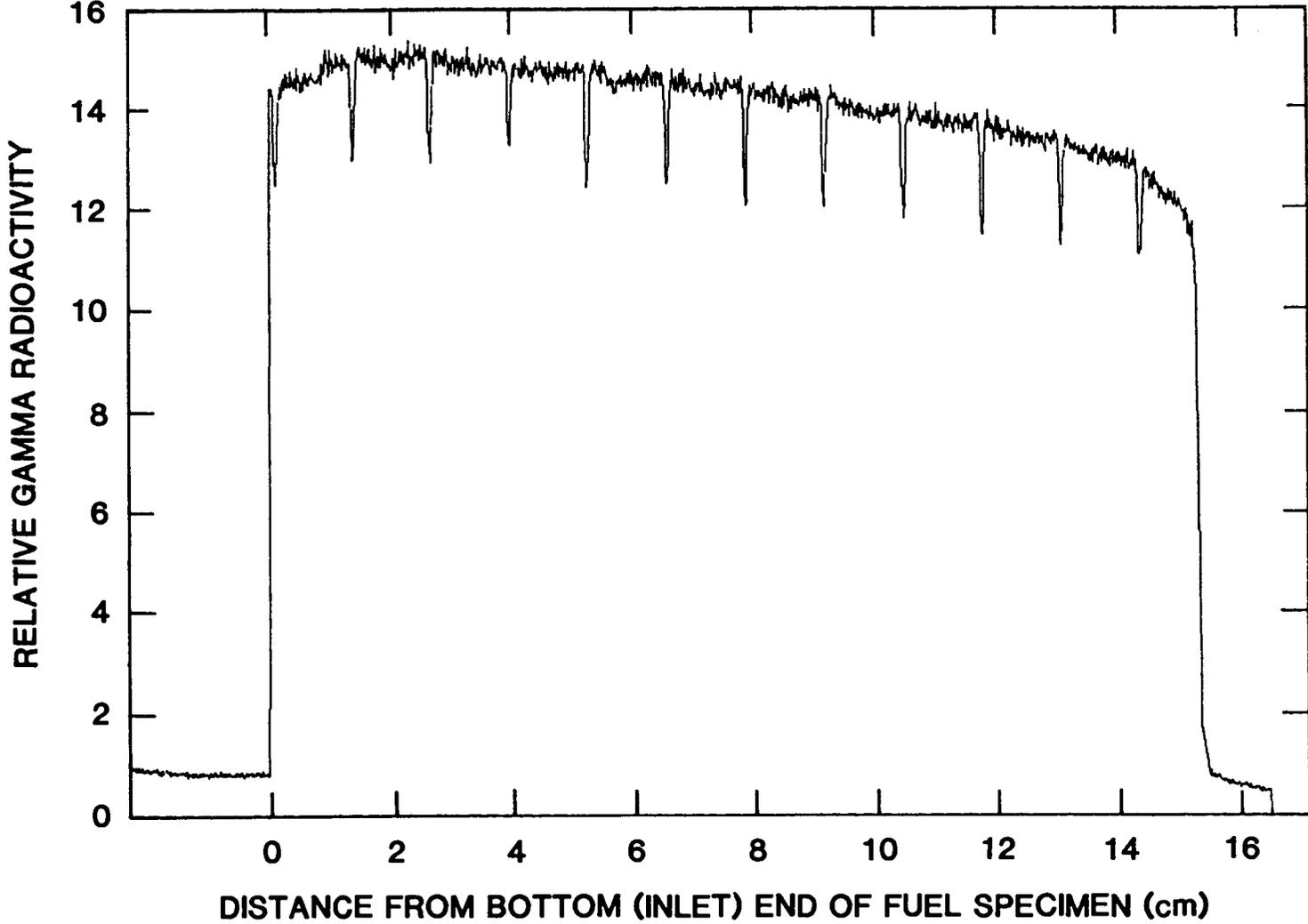


Fig. 6. Pretest gamma scan of specimen from rod I-114, heated in test VI-5. Note pellet-to-pellet gaps in radioactivity.

Table 5. Operating data for test VI-5

Specimen temperature (K):	
At start of heatup ramp	~550
During 10-min period to check pyrometers	1620
During first 20-min period (Phase A)	2015
During second 20-min period (Phase C)	2740
Heatup rate to Phase A plateau	0.25/s
Heatup rate to Phase C plateau	1.22/s
Cooldown rate to 2000 K	1.13/s
Time above 2000 K	61 min
Nominal gas flow rate data (L/min at 20°C, 1 bar): ^{a,b}	
50% H ₂ + He recirculation system	0.10
50% H ₂ + He to fuel specimen	0.80
Recirculation/purification system	1.5

^aMeasured by mass flowmeters.

^bAbsolute pressure in furnace during test was 99.1 kPa (743 mm Hg).

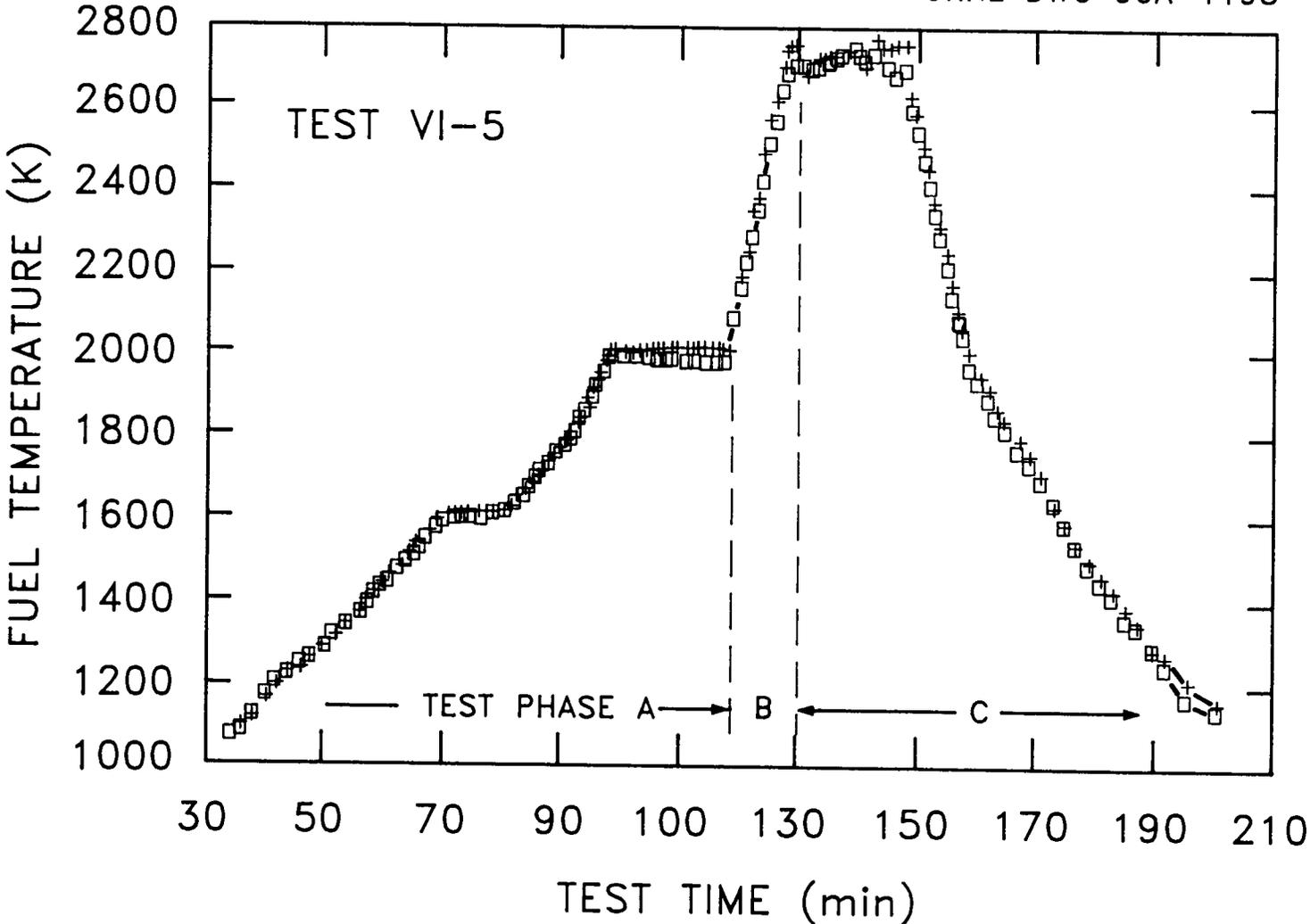


Fig. 7. Temperature history of test VI-5. Note collection periods for phases A, B, and C.

The more important events during the test are listed in the test chronology, Table 6. The time periods for operation of the three collection trains (Fig. 7) were for Train A, 0 to 118 min; for Train B, 118 to 128 min; and for Train C, 128 min to end of test, including cooldown. A preheat period was included to slowly heat the specimen to ~550 K prior to beginning hydrogen flow to the furnace. Time zero was defined as that time when the controlled heating ramp was begun, with stable gas (hydrogen + helium) flow through the warm furnace established. Temperature measurement and control were generally good. The 10-min period at ~1600 K was included to ensure heatup of ceramics in the outlet end of the furnace and to compare the data from the optical pyrometers before any significant release of fission products had occurred.

3.3 POSTTEST DISASSEMBLY AND EXAMINATION

After the test was completed, the apparatus was monitored for the distribution of radioactivity and then disassembled. Initially, the filter assemblies and the top furnace flange-TGT assembly, containing the TGT liners, were removed and transferred to another hot cell to avoid potential contamination from fuel handling. The ThO₂ top end plug could not be removed from the furnace tube to allow observation of the interior, so the cavity was filled with epoxy resin to preserve the geometry of the degraded fuel specimen for sectioning and subsequent microstructural examination. After removal of the fuel-furnace tube assembly from the furnace, it was analyzed in detail by gamma spectrometry. Efforts to obtain an X-ray view of the fuel geometry were unsuccessful because of the high density of the surrounding ThO₂ tube.

4. TEST RESULTS

4.1. GAMMA SPECTROMETRY DATA

All experimental components and collectors were analyzed after the test under well-defined geometry to determine the concentration of the gamma-emitting fission products. The empirical method of determining the effective shielding to obtain a mass balance for cesium among several of the ¹³⁴Cs gamma-ray energies was used.¹²

Pretest gamma spectrometric analysis of the 15.2-cm-long fuel specimen was used to determine the fission product inventories in the fuel. Long-lived gamma emitters - ¹⁰⁶Ru, ¹²⁵Sb, ¹³⁴Cs, ¹³⁷Cs, ¹⁴⁴Ce, and ¹⁵⁴Eu - were determined directly. A calculation by the computer program ORIGEN2, with corrections based on ¹³⁷Cs data, supplied the inventory values for other fission products, activation products, and fuel nuclides, as shown in Tables 3 and 4. The axial distributions of the gamma-emitting fission products were assumed to be similar to the gross gamma profile shown in Fig. 6. Other pretest data, based on measurements at 2-cm intervals, showed that the distributions of the major fission products were relatively uniform along the rod. Consequently,

Table 6. Chronology of test VI-5

Event/observation	Time		Furnace temperature corrected (K)	
	Clock (h)	Test (min)	Center	Bottom
Load fuel into furnace	945		RT ^a	RT
Complete leak checks	1100		RT	RT
Begin system preheat, TGTs and filters	1100		RT	RT
Begin furnace preheat, with gas flow to furnace	1230		RT	RT
Stable flow and temperature	1300		~550 ^b	~550 ^b
Test Phase A:				
Start ramp to ~1600 K	1321	0	~550 ^b	~550 ^b
First Kr detected	1351	30	~990	~995
First micro No. 1 T measurement	1353	32	~1015	~1020
Reached ~1600 K plateau	1432	71	1603	1620
Resume ramp to 2000 K	1442	81	1615	1621
First Cs detected, on TGT	1451	90	1773	1787
Rapid Kr release	1456	95	1902	1919
Reached 2000 K plateau, hold 20 min	1459	98	1998	2004
First Cs on filter	1503	102	2006	2021
End Phase A	1519	118	2003	2016
Test Phase B:				
Begin Phase B, rapid heatup	1519	118	2003	2016
Maximum release rates for Kr and Cs	1525	124	2428	2494
Reached 2700 K plateau, end Phase B	1529	128	2691	2746
Test Phase C:				
Begin Phase C, hold 20 min	1529	128	2691	2746
Kr and Cs on TGT leveling, Cs on filter still rising	1535	134	2710	2733
Cs on filter leveling	1548	147	2693	2755
End 2700 K plateau, reduce power	1549	148	2701	2756
Reduce cooling rate	1600	159	1971	2012
Last pyrometer measurements	1642	201	1142	1157
test essentially complete, power off	1653	212	~950 ^b	~950 ^b
End Phase C, stop gas flow	1930		RT	RT

we concluded that (1) the burnup was similarly uniform, and (2) the operating temperature was not high enough to cause significant migration of volatile species such as cesium.

The detailed posttest gamma scan of the fuel-furnace tube assembly, obtained with a 0.05-cm collimator, is shown in Fig. 8. Relocation of most of the gamma emitters is apparent, with the largest concentration 1 to 3 cm below the original bottom end of the fuel, indicating extensive meltdown/collapse during the test. Furthermore, most of this measured radioactivity was ^{154}Eu , almost all of the radio-cesium having been released from the furnace. Note the deposits in the ThO_2 top end plug, which were later shown to be primarily europium.

The posttest distributions of ^{106}Ru , ^{125}Sb , ^{144}Ce , and ^{154}Eu , as determined by gamma analysis of 1-cm-long sections of the fuel-furnace tube assembly, are shown in Fig. 9. In the region where the fuel was originally located (8 to 23 cm in Figs. 8 and 9), the dominant species was ^{106}Ru . High concentrations of ^{125}Sb and ^{144}Ce were found in the region just below the original fuel location. The ^{144}Ce is believed to be the best indicator for the location of UO_2 fuel, but the ^{125}Sb is believed to be associated with the Zircaloy cladding, which melted and ran down before the stack of fuel pellets collapsed.

The time and temperature at which the fuel rod collapsed in test VI-5 could not be determined precisely. The three radiation detectors that monitored activity in the top, middle, and bottom thirds of the fuel rod all appeared to show only the decrease in cesium activity with no sudden decrease (top) or increase (bottom) that would indicate fuel collapse. Therefore, the collapse must have occurred at a temperature above 2400 K (at which 50% of the cesium was still in the fuel rod), and possibly as high as 2700 K. The radiation detector viewing the bottom of the fuel rod began reading higher than the other two between 127 and 128 min run time. The fuel temperature was 2670 K at 127.5 min. The actual time of collapse is uncertain because of the poor collimation and high background for the "lower" radiation detector. Collapse must have occurred while the cladding/fuel combination was semimolten because of the degree of downward penetration of fission products below the original fuel location. Sectioning of the fuel remnants will be required to provide more specific information.

Gamma scanning of the fuel rubble and the original fuel location with a 2 mm \times 2 mm collimator was started. The objective was to look for the degree of uniformity of isotopes in the collapsed fuel region and to examine the horizontal distribution of gamma emitters in the original fuel region. Significant variations in the distribution of different isotopes were observed, but a malfunction of the gamma analysis system developed before sufficient data were obtained to determine any patterns. We plan to resume these measurements when resources permit.

The apparent difference in temperature at which fuel collapse occurred in tests VI-4 and VI-5 (~2150 K in VI-4 and >2300 K in VI-5)

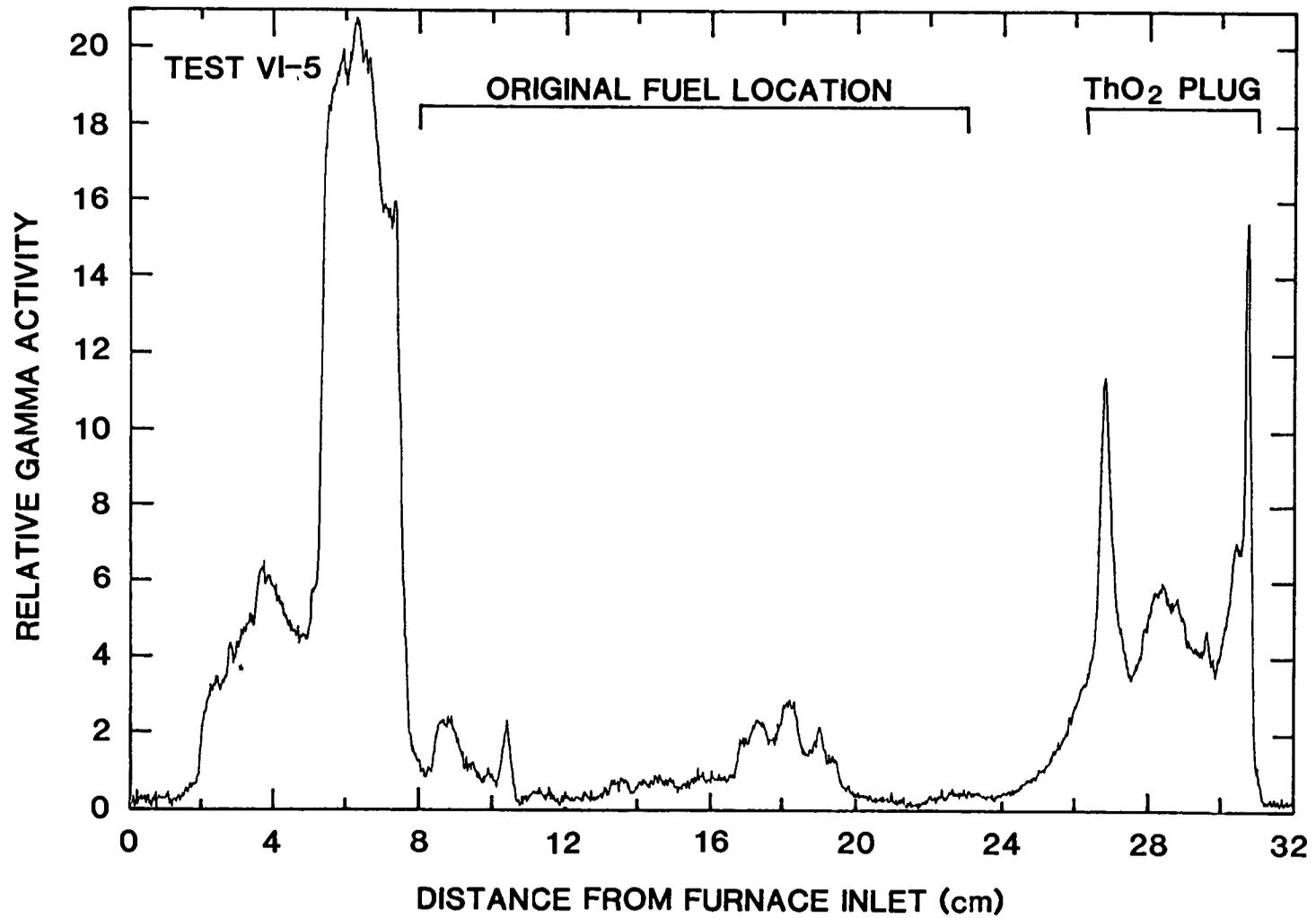


Fig. 8. Detailed gamma scan of test VI-5 fuel-furnace tube assembly after test.

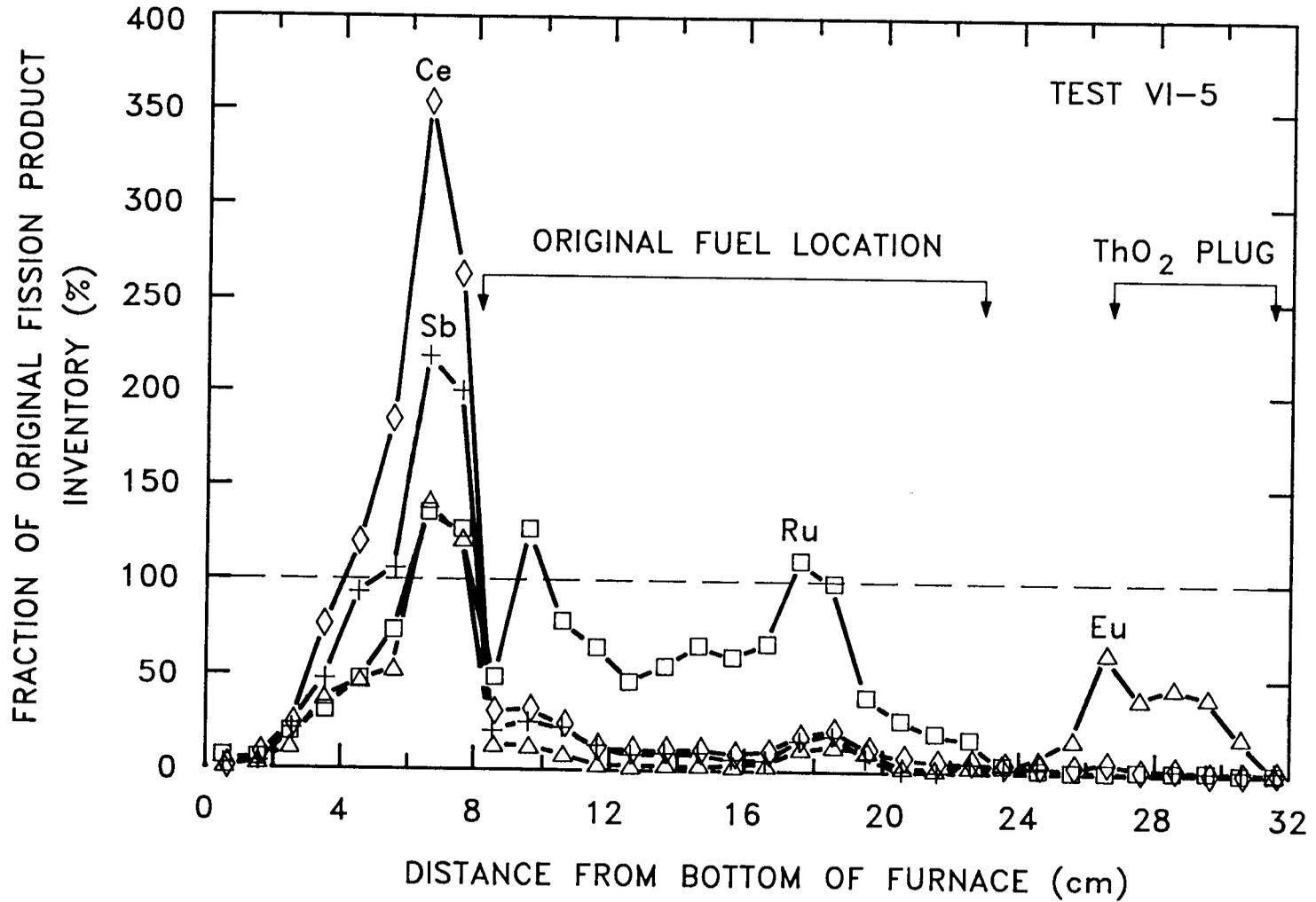


Fig. 9. Distribution of principal fission products in the fuel-furnace tube assembly after test VI-5.

could be a result of burnup difference (~47 MWd/kg in VI-4 and ~42 MWd/kg in VI-5). There is evidence to show that low burnup fuel resists fuel swelling and collapse even in contact with unoxidized Zircaloy. Two tests with trace-irradiated fuel heated by centered tungsten resistors in helium atmosphere were wet by the melted Zircaloy cladding but did not collapse.¹³ The fuel closest to the tungsten rod heater melted. Some of the unirradiated Zircaloy-clad fuel in the ACRR DF-4 test was exposed to a reducing atmosphere but did not collapse.¹⁴ Also, the unirradiated preheater fuel in ACRR ST-1 test did not collapse in spite of the hydrogen atmosphere and a maximum temperature of ~2400 K.¹⁵ The amount of wetting of the UO₂ by the Zircaloy in the last two examples is not known. The effect of fuel parameters, such as O/U ratio (low in the BR3 fuel case), is also unknown.

The results of tests VI-4 and VI-5 show that the VI apparatus is a useful facility for the study of fuel melt progression for any fuel type regardless of burnup or uranium enrichment.

As has been typical of these tests of high-burnup, long-decayed fuel, ¹³⁷Cs and ¹³⁴Cs were the dominant activities in almost all samples of released material and interfered with the analysis of less abundant fission products. The release behavior of krypton and cesium as functions of time and temperature is illustrated in Fig. 10; these curves show that most of the release occurred early in the high-temperature period, leaving very little volatile material available for release during the last half of the high-temperature period. More detail for krypton and cesium release behavior early in the test is presented in Fig. 11. This figure shows that, with rapid increase in temperature, krypton release precedes cesium release for a short period, and the release of cesium in vapor form increases more rapidly than the release in aerosol form. On-line analysis indicated that the krypton release value had reached >90% by the end of Phase B, when the fuel temperature had reached 2700 K (see Fig. 10). An estimated 2% of the krypton inventory had been released from the UO₂ to the pellet-cladding gap during irradiation and had been released when the rod was initially sectioned. The krypton remaining in the fuel when it reached 2700 K (~10%) was released within 10 min. A more complete history of cesium release and deposition by form for the entire test is shown in Fig. 12.

A summary of the fractional release results for Kr, Sb, Cs, and Eu, as determined by gamma spectrometry, is presented in Table 7. Although no data for ¹³⁴Cs are shown, the agreement with ¹³⁷Cs was consistently (within ±5%) good at all locations. The distribution of cesium within the test apparatus is shown in detail in Table 8. As has been noted previously, the largest fraction of the cesium (~60%) was released during Phase B, the 10-min heatup period from 2000 to 2700 K. Only ~9.4% of the total cesium remained to be released and collected during Phase C (20 min at 2700 K). About 7.5% of the released cesium was retained in the outlet end of the furnace, compared to ~24% retention in test VI-3 in steam. Since the temperatures and times were approximately the same in these two tests, the higher retention of cesium in steam suggests that most of the cesium was in a more reactive

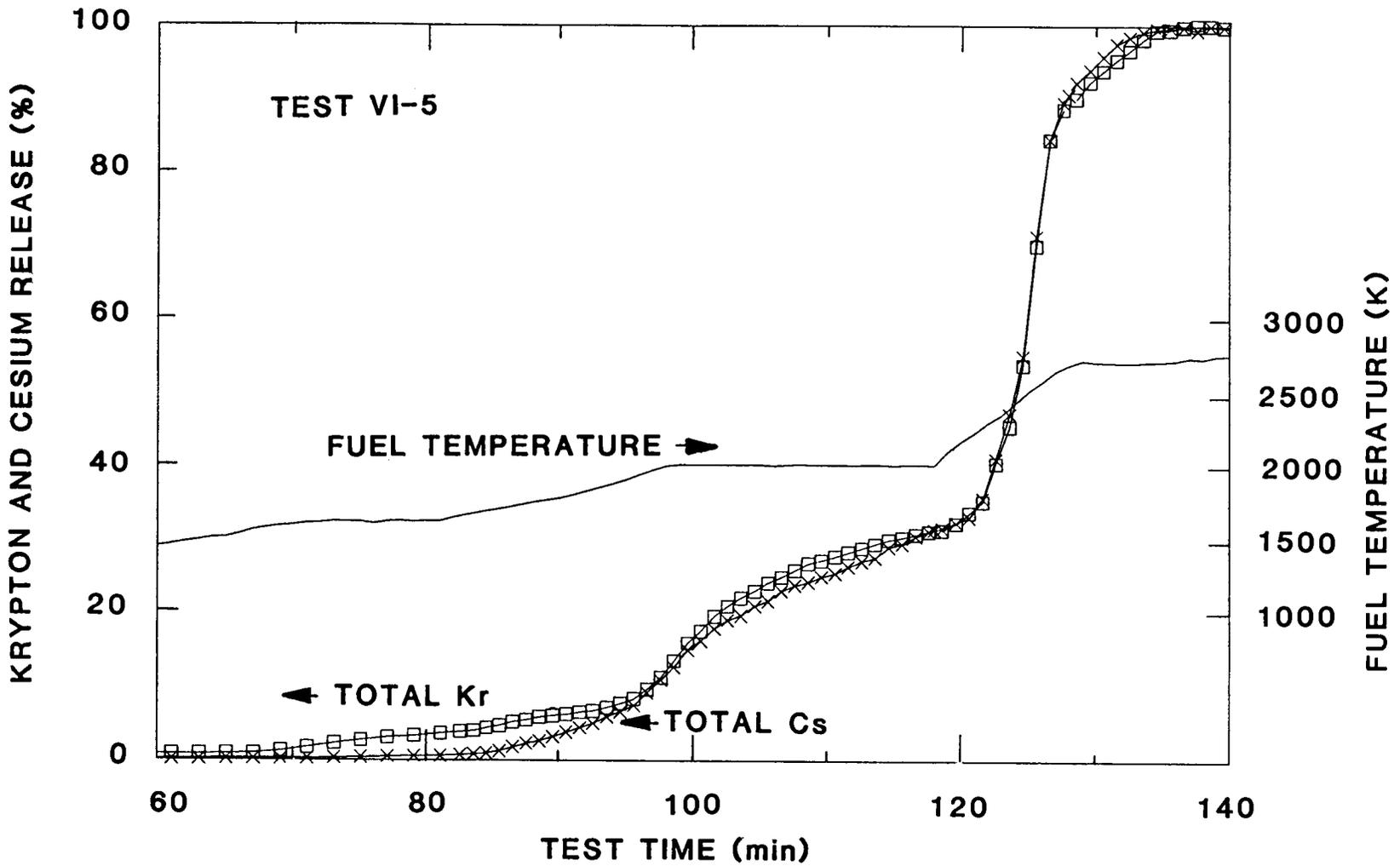


Fig. 10. Release behavior of krypton and cesium in test VI-5.

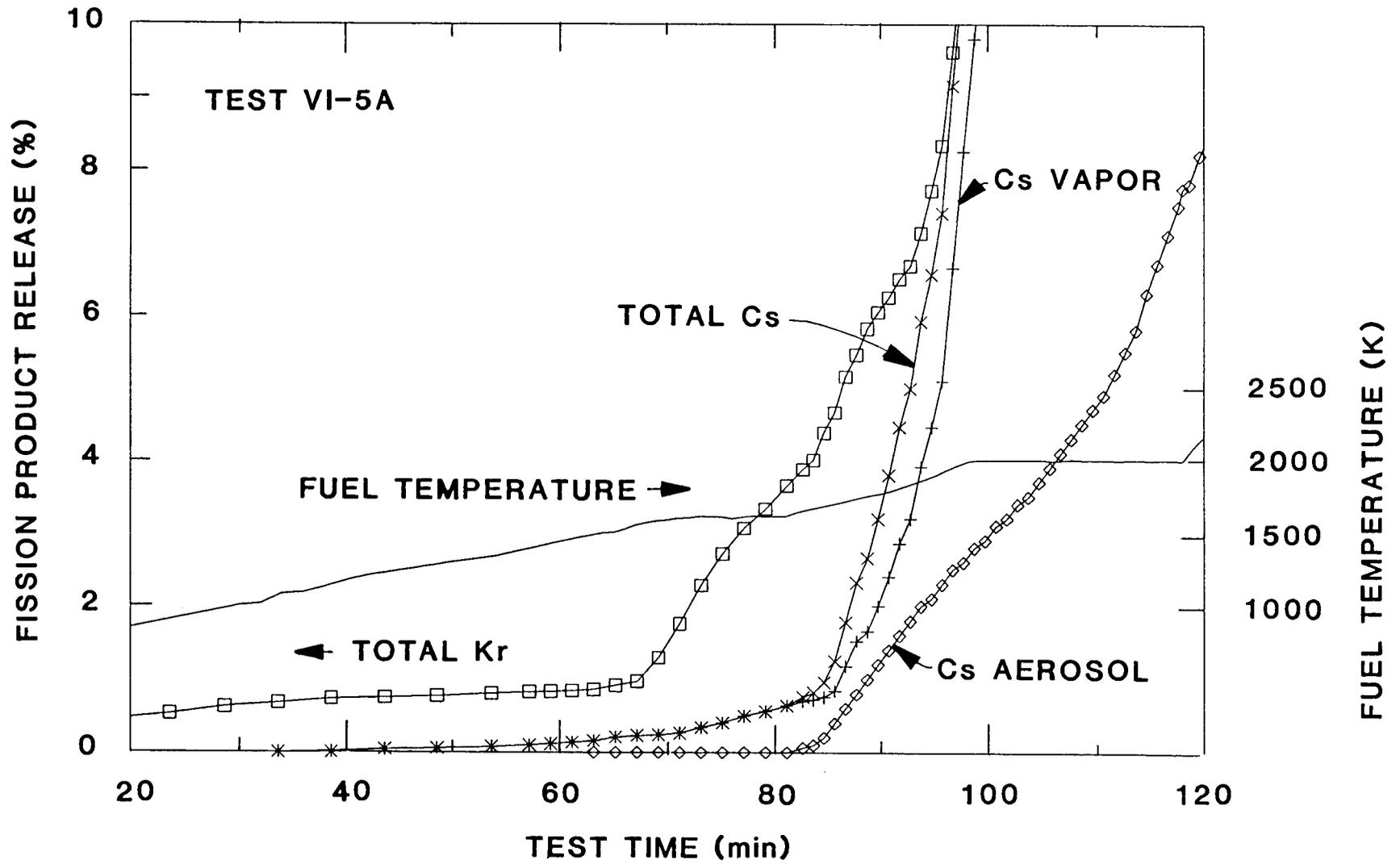


Fig. 11. Early release (test Phase A) of krypton and cesium in test VI-5.

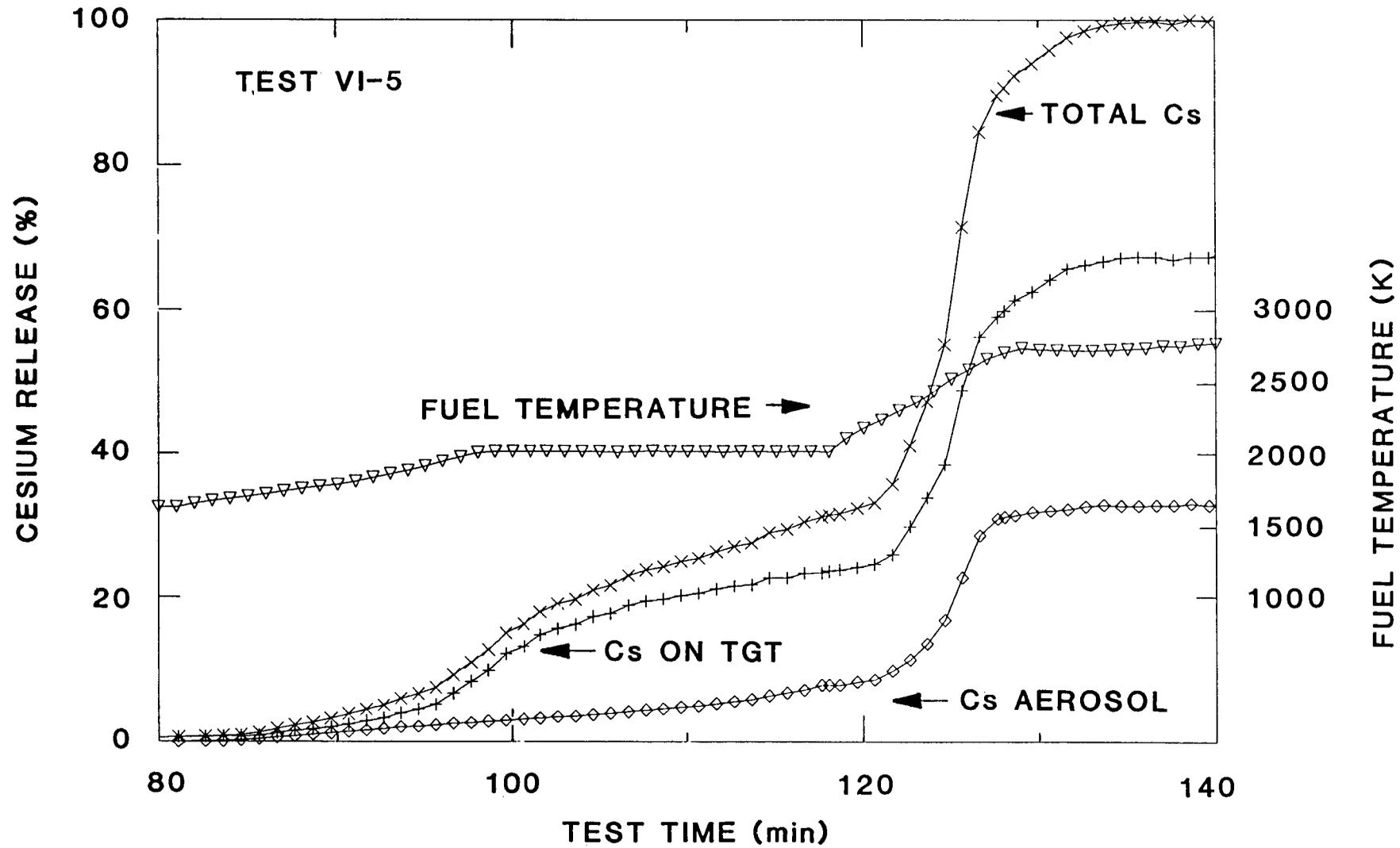


Fig. 12. Release behavior of cesium forms (vapor on TGT and aerosol on filters) in test VI-5.

Table 7. Summary of release data for test VI-5

Component/ collector	Operating time T >2000 K (min)	Percentage of fission product inventory released during each test phase ^a			
		⁸⁵ Kr ^b	¹²⁵ Sb ^c	¹³⁷ Cs	¹⁵⁴ Eu
Train A	20				
TGT			0	22.7	0.2
Filters			0	7.9	0.8
Total		31.0	0	30.6	1.0
Train B	10				
TGT			0	35.3	2
Filters			10	24.7	33
Total		58.1	10	60.0	35
Train C	31				
TGT			3	7.5	4
Filters			5	1.9	17
Total		10.9	8	9.4	21
Total for test	61	100	18	100	57

^aInventories based on pretest fuel analysis and ORIGEN2 calculations.

^bAn estimated 2% of the ⁸⁵Kr was released during irradiation; apparently, all of the remaining ⁸⁵Kr was released during test.

^cDistributions of Sb and Eu are approximate because of large Eu deposits in furnace and low Sb count rates.

Table 8. Cesium release and distribution data for test VI-5^a

Location	Approximate temperature (K)	Cesium found at each location		
		¹³⁷ Cs (mCi)	Total Cs (mg)	Total inventory (%)
Furnace components:				
Inlet region	500-1800	10	0.29	0.122
ThO ₂ plug	~2500	4	0.12	0.049
ZrO ₂ plug	~2000	551	15.88	6.723
Exit region	500-1800	55	1.59	0.671
Total		620	17.87	7.565
Train A:				
TGT A	470-1000	1706	49.17	20.815
TGT - filter line	430	35	1.01	0.427
First prefilter	405	500	14.41	6.101
Second prefilter	405	58	1.67	0.708
HEPA filters	405	4	0.12	0.049
Total		2303	66.37	28.10
Train B:				
TGT B	470-1070	2659	76.63	32.443
TGT - filter line	430	110	3.17	1.342
First prefilter	410	1729	49.83	21.096
Second prefilter	410	23	0.66	0.281
HEPA filters	410	2	0.06	0.024
Total		4523	130.35	55.19
Train C:				
TGT C	470-1010	568	16.37	6.930
TGT - filter line	430	8	0.23	0.098
First prefilter	410	124	3.57	1.513
Second prefilter	410	6	0.17	0.073
HEPA filters	410	1	0.03	0.012
Total		707	20.38	8.63
Total in dryer and cold charcoal		40	1.15	0.488
Total released from fuel		8193	236.12	99.96
Total in fuel (after test)		3.00	0.086	0.037

^aInventory based on measured data: 8196 mCi ¹³⁷Cs on July 1, 1986; 0.02882 mg Cs/mCi ¹³⁷Cs based on ORIGEN2.

form, probably CsOH in steam vs some other cesium vapor form in hydrogen. Also, most (65%) of the cesium was collected on the TGTs in test VI-5, an indication that the majority was in a vapor form, rather than associated with the aerosol. Conversely, tests in steam have consistently shown 60 to 70% of the cesium deposited on the filters as aerosol.

The indicated release values for ^{125}Sb and ^{154}Eu are shown in Table 9. Although these nuclides are of relatively low yield and are not among the most hazardous, they have intermediate half-lives and strong gamma rays, which make them easily detected. The release of europium (57%) was much higher than in any previous test. Very little europium, which is relatively involatile under oxidizing conditions, has been released in tests in steam, but 13.4% was released in test VI-4 at 2400 K in hydrogen. As shown in Table 9, significant fractions (6.7 and 55%, respectively) of the released Sb and Eu were deposited on ceramic surfaces in the outlet region of the furnace, where temperatures were believed to be 1500 to 2500 K during the test.

The relative distributions of fission product ^{106}Ru , ^{125}Sb , ^{144}Ce , and ^{154}Eu remaining in the fuel-furnace tube assembly after the test are shown in Fig. 10. Because of its similar chemical and physical properties, ^{144}Ce should be the best indicator of UO_2 position within the degraded fuel specimen. The position of this peak indicates that most of the UO_2 collapsed and/or disintegrated into the region slightly below the original bottom end of the fuel specimen. Some interactive melting with the Zircaloy cladding (location indicated by the ^{125}Sb peak), which melted and ran down to this region earlier in the test, is likely.

4.2 ANALYSIS FOR IODINE

Since iodine has no long-lived, gamma-emitting nuclides, analytical methods other than gamma spectrometry must be used. Neutron activation of ^{129}I to ^{130}I , which can be counted easily, is a proven, sensitive technique. Because the normally occurring iodine forms dissolve readily in basic solutions to form stable iodides, the collector components from this test were leached to remove this iodine for analysis. Because the final analyses of these samples are incomplete at this time, the results will be reported later.

Some iodine forms (organic iodides, elemental iodine, or hydrogen iodide) may pass through the TGTs, penetrate the filters, and adsorb onto the heated charcoal in the filter packages. For this reason, the charcoal cartridges were analyzed for ^{129}I by direct activation also. The results of these analyses are shown in Table 10. The fraction of the released iodine that was collected on the charcoal (2.85%) was higher in test VI-5 than in tests conducted in steam, normally <0.5%. The calculation for test VI-5 assumes that 100% of the iodine inventory was released.

Table 9. Fractional release and distribution of antimony and europium in test VI-5^a

Location	¹²⁵ Sb			¹⁵⁴ Eu		
	Amount found (mCi)	Total inventory (%)	Released (%)	Amount found (mCi)	Total inventory (%)	Released (%)
Furnace components:						
ThO ₂ plug	1.000	1.04	5.95	43.920	22.03	38.70
ZrO ₂ plug	0.000	0.00	0.00	11.860	5.95	10.45
Furnace tube	0.120	0.12	0.71	4.100	2.06	3.61
Exit Region	0.000	0.00	0.00	2.285	1.15	2.01
Furnace total	1.120	1.163	6.659	62.165	31.176	54.771
Train A:						
TGT A	0.000	0.00	0.00	0.126	0.06	0.11
TGT - filter line	0.000	0.00	0.00	0.043	0.02	0.04
First prefilter	0.000	0.00	0.00	0.565	0.28	0.50
Second prefilter	0.000	0.00	0.00	0.018	0.01	0.02
HEPA filters	0.000	0.00	0.00	0.002	0.00	0.00
Train A total	0.000	0.000	0.00	0.754	0.38	0.66
Train B:						
TGT B	0.000	0.00	0.00	1.753	0.88	1.54
TGT - filter line	0.357	0.37	2.12	2.048	1.03	1.80
First prefilter	8.350	8.67	49.64	27.590	13.84	24.31
Second prefilter	0.000	0.00	0.00	0.112	0.06	0.10
HEPA filters	0.000	0.00	0.00	0.010	0.01	0.01
Train B total	8.707	9.040	51.766	31.513	15.804	27.765
Train C:						
TGT C	2.599	2.70	15.45	3.738	1.87	3.29
TGT - filter line	0.179	0.19	1.06	1.056	0.53	0.93
First prefilter	4.000	4.15	23.78	13.610	6.83	11.99
Second prefilter	0.172	0.18	1.02	0.592	0.30	0.52
HEPA filters	0.025	0.03	0.15	0.090	0.05	0.08
Train C total	6.975	7.241	41.47	19.086	9.57	16.82
Total released from fuel	16.802	17.44	99.89	113.518	56.93	100.02
Total in fuel (after test)	79.50	82.54		85.91	43.08	

^aInventories based on measured data: 96.32 mCi ¹²⁵Sb and 199.4 mCi ¹⁵⁴Eu.

Table 10. Iodine found on heated charcoal cartridges in test VI-5

Train	¹²⁹ I on charcoal (% of total inventory)			
	First	Second	Third	Total
A ^a	1.39	3.1E-4	5.6E-4	1.39
B	1.42	5.2E-3	<3E-4	1.43
C	2.9E-2	<2.1E-3	<3E-4	3.1E-2
Total				2.85

^aTGT A contained an unoxidized stainless steel liner.

One plausible explanation for the large amount of volatile iodine is that it was caused by the combination of the hydrogen atmosphere and the unoxidized stainless steel liner in TGT A. The volatile iodine in the B train must have resulted from a slow reaction between the stainless steel liner and a deposited iodine species, accompanied by diffusion back into the B train.

A second possible source of the volatile iodine is radiation decomposition of an iodine species collected as particulate material on the glass wool and HEPA filters. The activated charcoal cartridges are part of the filter pack assembly and in the case of VI-5, the filter/charcoal assembly remained sealed for 5 months. The usual time before disassembly is 5 to 9 weeks.

4.3 THERMAL GRADIENT TUBE DEPOSITS

The stainless steel liner in TGT A was removed without difficulty. Attempts to remove the platinum liners from TGTs B and C, however, revealed that these liners were firmly stuck to the surrounding Inconel tubes. Inspection of the inlet (bottom) ends of these tubes showed droplets of very hard, apparently previously molten material connecting the platinum liners to the Inconel tubes. The platinum liner was removed from TGT B with the loss of only a small piece of the liner, but removal of the platinum liner from TGT C proved to be much more difficult. Because the attachments between the liner and TGT C were more extensive and appeared to extend several centimeters from the inlet, in situ leaching and gamma scanning were carried out before further efforts toward removal were attempted.

Samples of the hard metallic material connecting the platinum liners to the Inconel TGTs were collected for analysis; but because of the attendant high gamma radioactivity, no analyses have been obtained

to date. Tin from the Zircaloy cladding appears to be a very likely constituent of the alloy, with platinum and perhaps nickel from the Inconel being other constituents. (In the hydrogen atmosphere of this high-temperature test, relatively large amounts of tin would be expected to volatilize from the molten Zircaloy and to be transported in the elemental form to the TGTs. The Pt-Sn phase diagram shows a number of compositions with a broad range of melting points.¹⁶ Since the TGTs experienced temperatures $>750^{\circ}\text{C}$ at the inlet end during the time vapors and aerosols were being transported from the fuel, the compound PtSn_4 , with a melting point of 745°C , appears to be a likely candidate. The cladding contained ~ 320 mg of tin initially.

The stainless steel liner from TGT A and the platinum liner from TGT B were gamma scanned separately, and the platinum liner, which could not be readily removed from TGT C, was scanned in place. As shown in Fig. 13, the liner from TGT A contained a large cesium deposition peak where the deposition temperature was $\sim 600^{\circ}\text{C}$ (875 K). A shoulder on this peak at $\sim 550^{\circ}\text{C}$ (825 K) suggested the presence of a second cesium species. Because the platinum liner in TGT A of test VI-4 (also in a helium-hydrogen atmosphere) exhibited very similar peaks, these data indicate that the peaks were formed by condensation of cesium species rather than by chemical reaction with the TGT liner deposition surfaces. Based on the results of previous tests, CsI is believed to be a major component of this peak. In addition to cesium, TGT A from test VI-5 contained small ($\sim 0.2\%$ of the fuel inventory) but significant europium deposits also. These results are consistent with the high releases observed for cesium ($\sim 100\%$) and europium (57%) in this test.

The platinum liner from TGT B exhibited three large cesium deposition peaks (Fig. 13). The high, narrow peak at the entrance is likely to be at least partially a result of flow transition effects. The second peak, at $\sim 775^{\circ}\text{C}$ (1050 K), appears to be characteristic of deposition of a specific chemical form, as yet unidentified. The broad third peak, extending from ~ 700 to $\sim 400^{\circ}\text{C}$ (975 to 675 K), is probably a combination of peaks resulting from deposition of a mixture of cesium forms, including CsI.

Compared to TGTs A and B, only $\sim 25\%$ as much radioactivity was found in the deposits in TGT C. About 90% of the cesium, the dominant radioactive species, was released during the first two phases of the test. In addition to cesium, small amounts of ^{60}Co , ^{125}Sb , and ^{154}Eu were found in TGT C. (The ^{60}Co probably evolved from the Zircaloy cladding - cobalt is a minor constituent of Zircaloy - at the very high temperature.) These elements showed no consistent patterns in their profiles, suggesting that the primary mode of transport and deposition was in association with aerosols.

4.4 DEPOSITION OF CESIUM ON STAINLESS STEEL

The liner in TGT A was made of 0.003-in.-thick Type 304 stainless steel foil; it was in the as-received condition except for annealing in He-4% H_2 to aid forming.

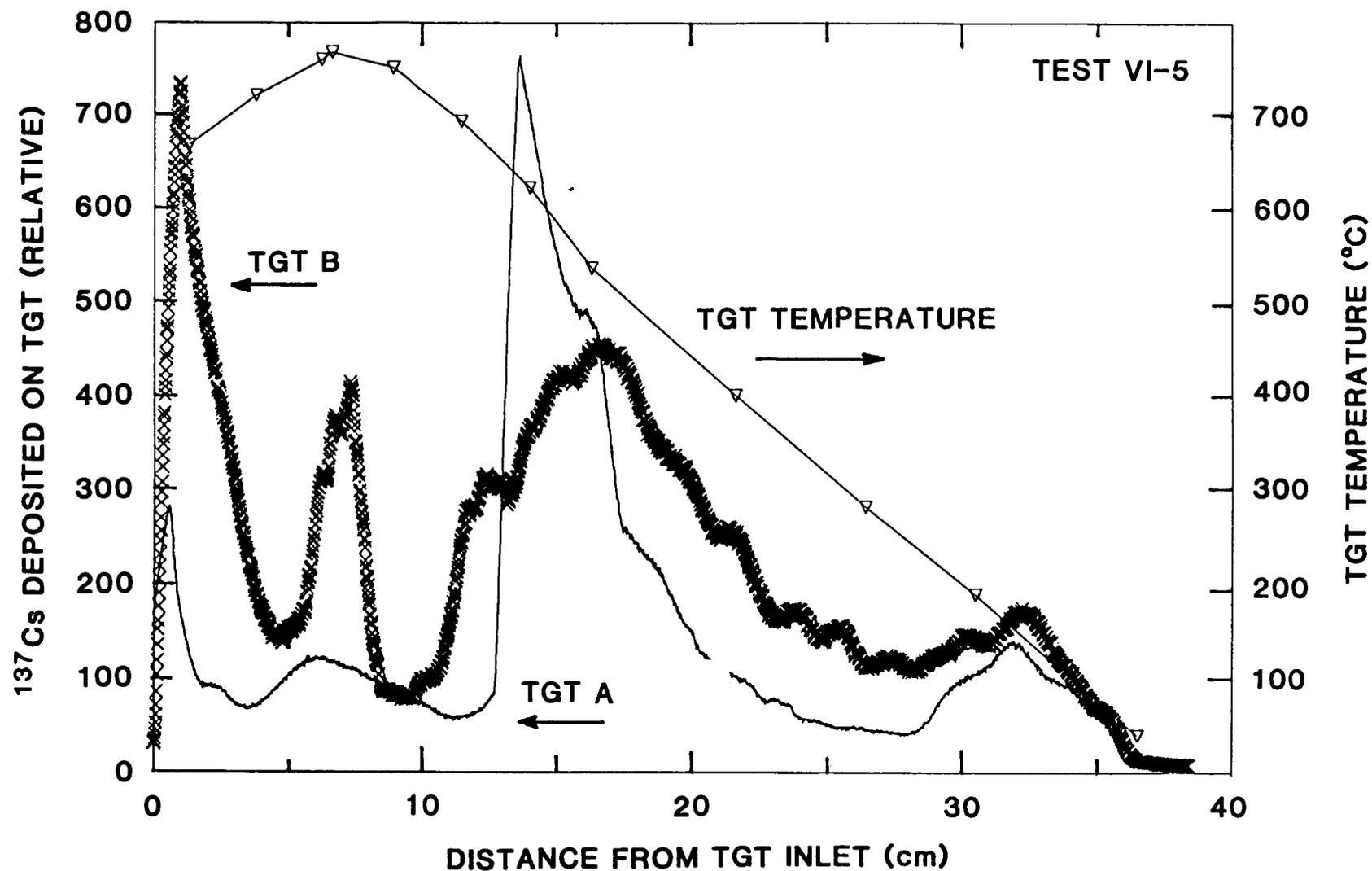


Fig. 13. Distribution of ^{137}Cs along thermal gradient tubes A (SST) and B (Pt) in test VI-5. Note temperature profile also. Peak concentrations are $\sim 3 \text{ mg Cs/cm}^2$.

As mentioned above, the cesium peak at 14 cm on the stainless steel liner of TGT A appears to be the result of condensation of a vapor species. The deposit between 2 and 13 cm could have resulted from a slow reaction between a cesium vapor species and the stainless steel liner surface. It is common (but not yet proved to be correct) to assume a constant rate of reaction and to express this rate as a deposition velocity. The maximum deposit was at 6.5 cm where the temperature was 1050 K (775°C). The surface concentration reached 0.36 mg Cs/cm², and the gas-phase concentration averaged 2.7×10^{-4} mg Cs/cm³ at 1050 K. Particulate cesium was not included in the gas-phase concentration. The major cesium accumulation in TGT A occurred over a 20-min period. These numbers result in a deposition velocity of ~0.5 cm/s. Condensation of a cesium species could have contributed to this rate as suggested by the deposition peak on the platinum liner of TGT B at the same temperature.

4.5 SSMS ANALYSES

Small smear samples of the material deposited on the filters were collected on graphite electrodes for analysis by spark-source mass spectrometry (SSMS). This technique has been used extensively in past experiments; it has advantages in that it is sensitive, not only for the radionuclides but also for the stable nuclides, including structural materials and impurities in the system. Its disadvantage is relatively low precision, a factor of 2 to 3. Unfortunately, the facility for conducting these measurements is currently not in operation, and re-start is uncertain. If the SSMS facility is returned to service, the samples will be analyzed and the results will be reported.

4.6 ICP-ES ANALYSES

Samples of the acidic leach solutions from the filters were submitted for inductively coupled plasma-emission spectrometry (ICP-ES) analyses for non-gamma-emitting elements. This technique is well suited for measuring several of the fission product elements, primarily cations, and uranium. Unfortunately, it is not useful for iodine analysis. Because of the high levels of radiocesium in most of the samples, large dilutions have been required to avoid excess radiation dose to the analyst. Unfortunately, these large dilutions have resulted in poor sensitivities for measuring the elements of interest in previous tests. The sensitivity for Te has been disappointingly low, but useful data for Mo, Ba, and U have been obtained in most cases.

As a result of delays in collecting the samples from test VI-5 and in obtaining the desired analyses, no ICP-ES results are currently available. The results, along with those from activation analysis for iodine, will be reported in the near future.

4.7 MASSES OF DEPOSITS IN TGTS AND ON FILTERS

The filters and TGT liners are weighed before and after the test to determine the mass of material collected during the test. Immediately after disassembly of the filter packages, the filters were inspected and photographed, then packaged for weighing. Although the prefilters exhibited light deposits, no deposits were visible on the HEPA filters, indicating efficient collection of most of the aerosols by the previous filters. The masses of material collected at the various locations are listed in Table 11 and illustrated in Fig. 14. As would be expected, the greatest masses were collected at the highest temperatures, during Phases B and C. The total mass collected (0.681 g) was greater than in the lower-temperature test VI-4 (0.401 g), but much less than in the steam atmosphere test VI-3 at the same temperature (3.168 g).

Table 11. Vapor and aerosol deposits in test VI-5

	Weight of deposits (g) ^a			
	Train A	Train B	Train C	Total
Thermal gradient tube (TGT)	0.104	0.050 ^b	0.030 ^b	0.184
Filters:				
Prefilter 1 ^c	0.045	0.267	0.172	0.484
Prefilter 2	0.002	0.003	0.005	0.010
HEPAs	0.0	0.003	0.0	0.003
Total filters	0.047	0.273	0.177	0.497
Total TGT and filters	0.151	0.323	0.207	0.681
Aerosol concentration ^d at 423 K (g/m ³)	7.3	36	7.2	12

^aPrecision = ± 0.003 g.

^bEstimated value; fusion of platinum TGT liner to TGT prevented accurate measurement.

^cIncludes estimated mass of deposits in connecting tubes, based on ¹³⁷Cs data.

^dAssumes all aerosol was formed at temperatures ≥ 1900 K and that the average temperature in the TGTs and filters was 423 K.

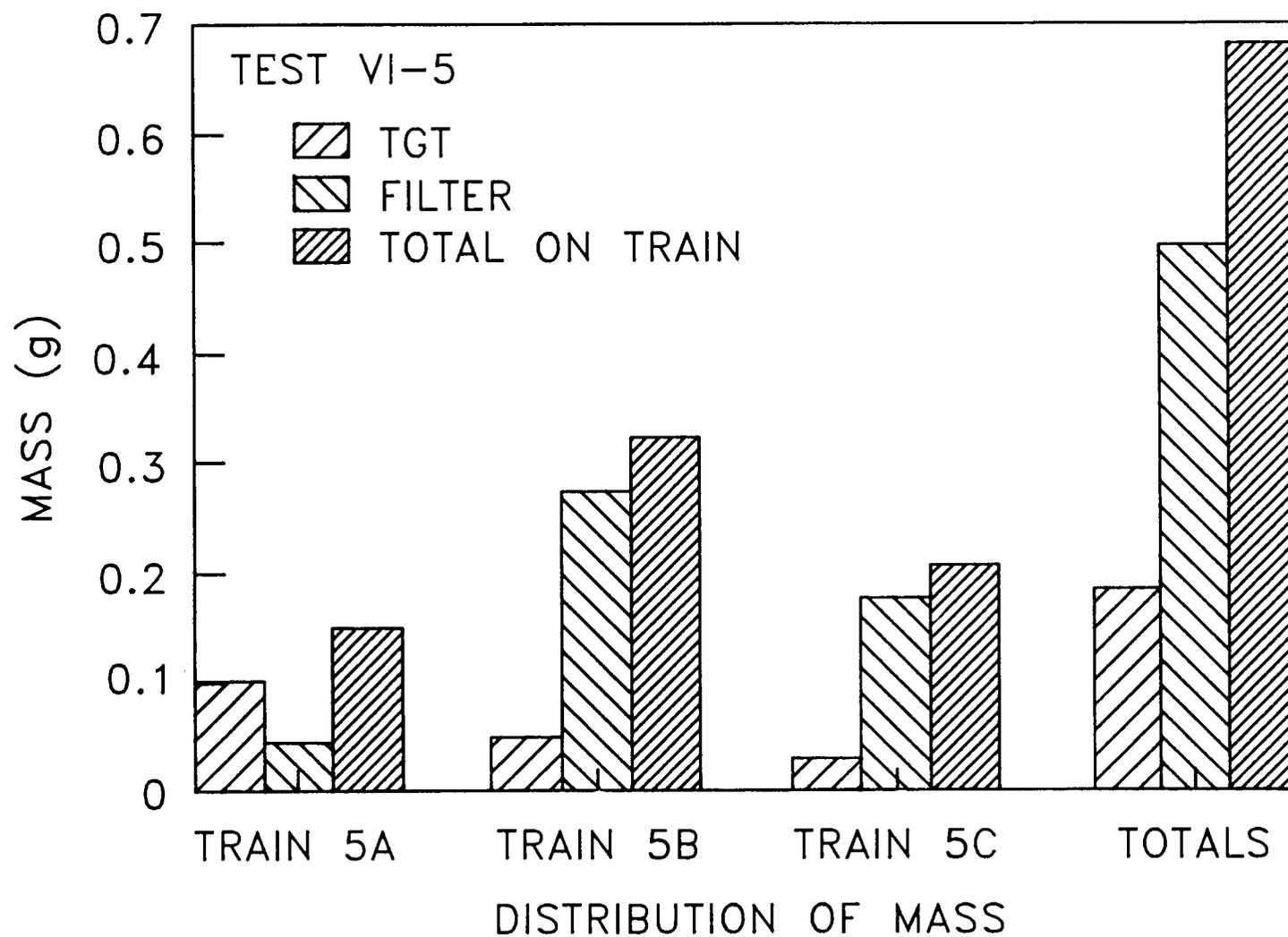


Fig. 14. Mass distribution of material released during test VI-5. The masses for TGT B and TGT C were estimated and may be revised later.

Most of the material deposited in the TGT liners is believed to be a result of vapor condensation, and the material deposited on the filters probably was transported primarily as aerosol. The average mass concentration of vapor and aerosol during each test phase was calculated from the masses collected and the total gas flow during that period (see Table 11). To reduce the distortion of the heatup and cooldown periods, it was assumed that no aerosol was formed at temperatures below 1900 K. These data show that the average mass concentration of the aerosol was highest during test Phase B and that this average mass concentration declined during the test as the supply of the more volatile material was depleted.

5. COMPARISON OF RELEASE DATA WITH PREVIOUS RESULTS

The fission product release data from this test (VI-5) have been compared with the results from earlier experiments and also with results from a comprehensive NRC review of all relevant fission product release data.¹⁷ Because tests VI-3 and VI-5 were essentially identical except for atmosphere - oxidizing in VI-3 and reducing in VI-5 - we were particularly interested in comparing the results from these two tests. In addition, test VI-4 was conducted in hydrogen, but at a lower temperature (2440 K) than VI-5, thereby providing a temperature comparison in the same atmosphere; and test VI-2, in steam, was at only a slightly lower temperature (2300 K). The release data from test VI-5 are compared with these earlier tests in Table 12.

As these data show, the fractional releases of krypton and cesium were always similar, except for the lower krypton value for test VI-2, where some of the gas was known to have been lost. Further, the increase in the releases of krypton and cesium with increasing temperature was consistent and reached ~100% at 2700 K. The results indicate no significant difference in the effects of the reactive atmospheres, steam and hydrogen, on these elements.

The release behaviors of antimony and europium, on the other hand, were quite different and reflected the effect of the reactive atmospheres on the elements (Table 12). In the tests at 2000 K for ~30 min, very little (<1%) release of antimony or europium was observed in either atmosphere. At 2300 to 2400 K, however, differences in release were apparent and became more exaggerated at 2700 K. Much higher release of antimony was observed in steam, whereas europium release was much higher in hydrogen. This behavior is consistent with the chemical characteristics of these elements. Previous work has shown that, although antimony release from the UO₂ may be similar to that of cesium, the antimony tends to combine with metallic Zircaloy and be retained until the zirconium is fully oxidized.¹⁸ Consequently, in tests in steam, antimony release is delayed until the cladding becomes oxidized, then occurs rapidly. In tests in hydrogen, the zirconium oxidation, which must depend on reduction of the UO₂ or furnace ceramics, is slow and/or incomplete. In addition, antimony release is limited to relatively small fractions, because the antimony is retained in the metallic zirconium. Based on the results of previous tests, the

Table 12. Comparison of release data from BR3 fuel in tests VI-3, VI-4, and VI-5

Test No.	Maximum temperature (K)	Effective time ^a (min)	Reactive atmosphere	Fission product released (% of inventory)			
				Kr	Sb	Cs	Eu
VI-3A	2000	34	Steam	26	0	22	0
VI-5A	2000	33	Hydrogen	31	0	31	1
VI-2(A+B)	2300	25	Steam	>31 ^b	49	54	0
VI-4(A+B+C)	2400	31	Hydrogen	85	3.9	95	13
VI-3(A+B+C)	2700	46	Steam	100	99	99	<1
VI-5(A+B+C)	2700	31	Hydrogen	100	18	100	57

^aIncludes estimates for heatup and cooldown effects.

^bSome ⁸⁵Kr lost during test VI-2.

release fractions for tellurium are expected to be similar to those of antimony in both types of atmosphere.¹⁹

Only in tests in hydrogen has significant release of europium been observed. (Trace release of europium in test HT-4, which was believed to have resulted from localized melting of the Zircaloy cladding and reduction of the adjacent UO_2 , was an exception.) As is typical for the rare earths, the stable oxide, Eu_2O_3 , has a much lower vapor pressure than the element. Therefore, it appears that in steam atmosphere tests, the fission product europium becomes fully oxidized before it can escape the fuel, but in a hydrogen atmosphere, europium is not fully oxidized and escapes much more easily as a lower oxide. Achard and Albert reported the reduction of Eu_2O_3 to Eu_3O_4 in hydrogen at 1300 to 1600°C (1573 to 1873 K), with the product being volatilized at >1600°C.²⁰ In test VI-5, the major deposits of europium were found in the outlet end of the furnace, at locations of very high temperature (probably 1500 to 2000°C), in apparent agreement with the observations of Achard and Albert. Since the environment of the molten Zircaloy is even more reducing than the hydrogen atmosphere, and since the highest release rate period for europium is during cladding meltdown, the influence of the molten Zircaloy may be stronger than that of the hydrogen. Under these conditions, an even lower oxide might form. The behavior of barium and lanthanum, and also of other rare earths, should parallel that of europium.

As in previous tests, the released behaviors of both ^{85}Kr and of ^{134}Cs and ^{137}Cs were quite similar in test VI-5 (see Fig. 10). The integral release curves for Cs vs temperature in two tests (VI-3 in steam and VI-5 in hydrogen) are shown in Fig. 15. The two periods of constant temperature (2000 and 2700 K) are apparent. The release curves for ^{85}Kr for these two tests are essentially identical to the Cs curves.

Because the ^{85}Kr and ^{137}Cs were collected and measured on-line at 1-min intervals during the test, both the continuous integral releases and the minute-by-minute release rates can be determined. The release rates for these two nuclides are compared, as functions of temperature, in Figs. 16(a) and 16(b). The corresponding data from both test VI-3 and VI-5 are compared with the release rates calculated using the CORSOR-M model.²¹ These comparisons show that both elements behaved similarly in both tests, whether in steam (VI-3) or in hydrogen (VI-5). During the temperature interval 2250 to 2700 K, both krypton and cesium were released at somewhat higher rates in test VI-5. The number of data points (one per minute) in Figs. 16(a) and 16(b) indicates the more rapid heatup rate in test VI-5. Both the higher heatup rate and the fuel collapse in test VI-5 may have promoted the higher release rates. The vertical clusters of points at 2000 and 2700 K identify the 20-min periods at constant temperature and, for the period at 2000 K, also show the decline in release rate with fraction released from the fuel.

The same on-line release data for krypton and cesium were used to illustrate the increase in integral release with temperature, as shown in Figs. 17 and 18, respectively. The most applicable data from both

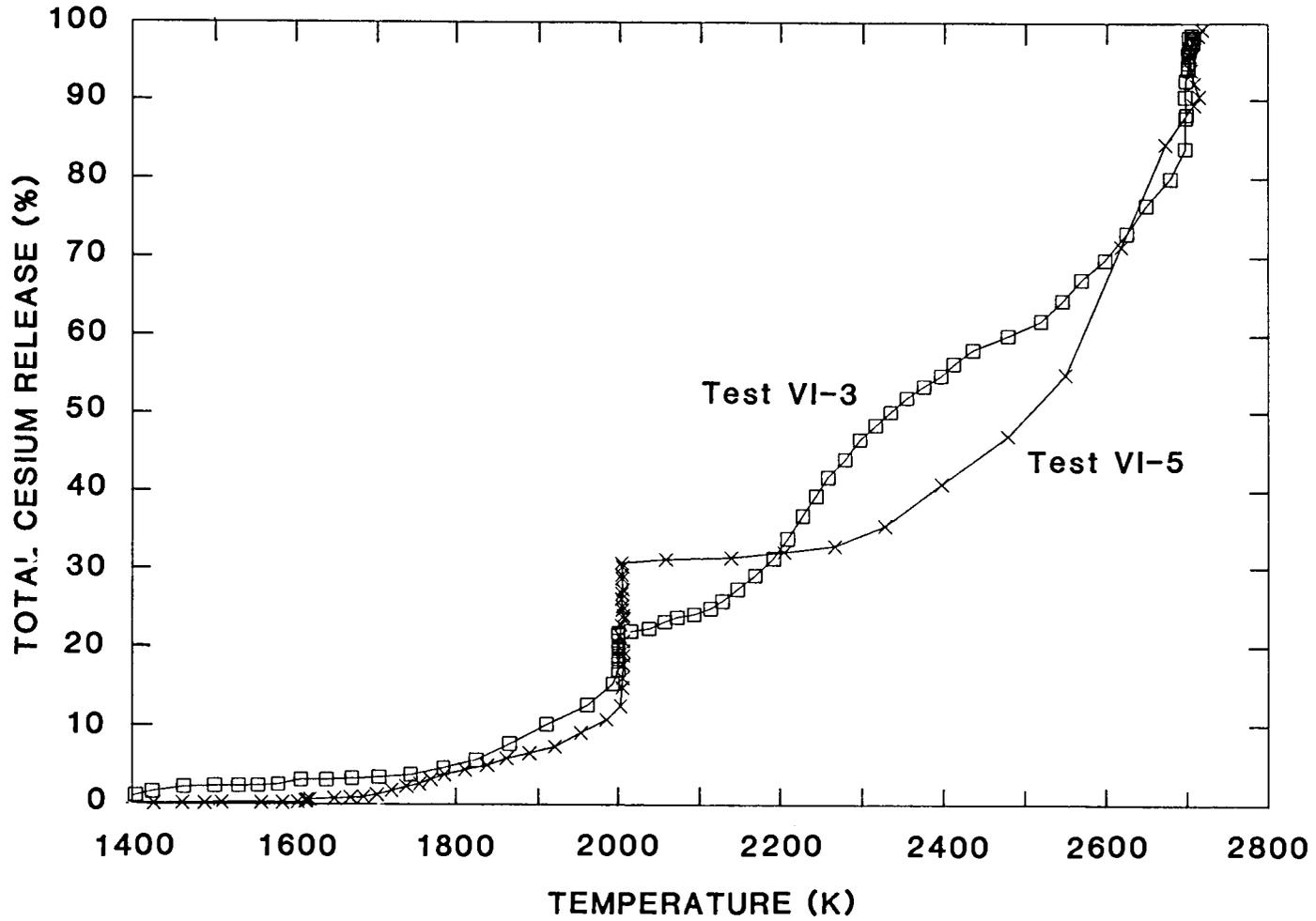


Fig. 15. Comparison of integral cesium releases in test VI-3 (steam atmosphere) and VI-5 (hydrogen atmosphere) as functions of temperature.

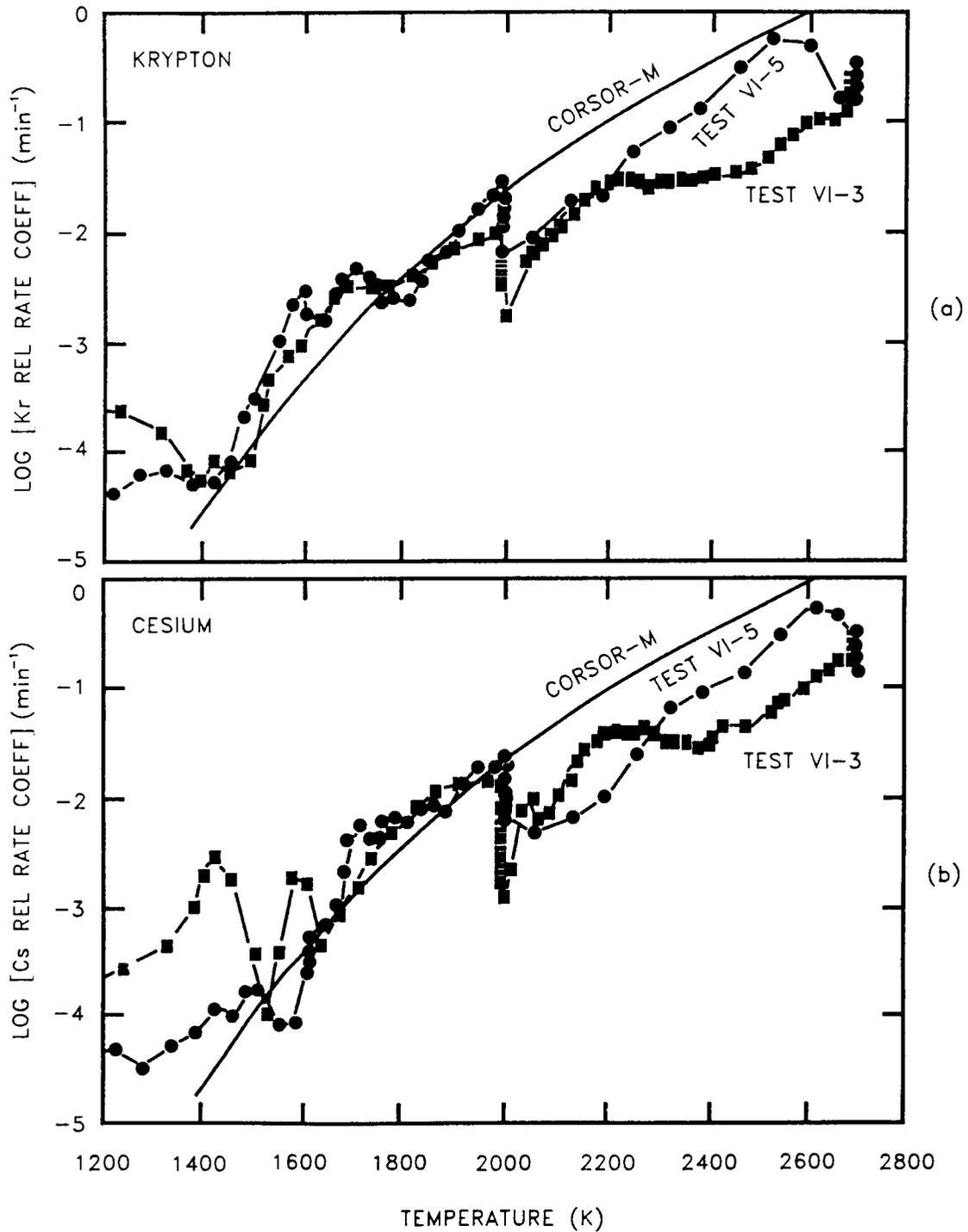


Fig. 16. Release rate coefficients for krypton (a) and cesium (b) vs temperature in tests VI-3 (steam atm) and VI-5 (hydrogen atm).

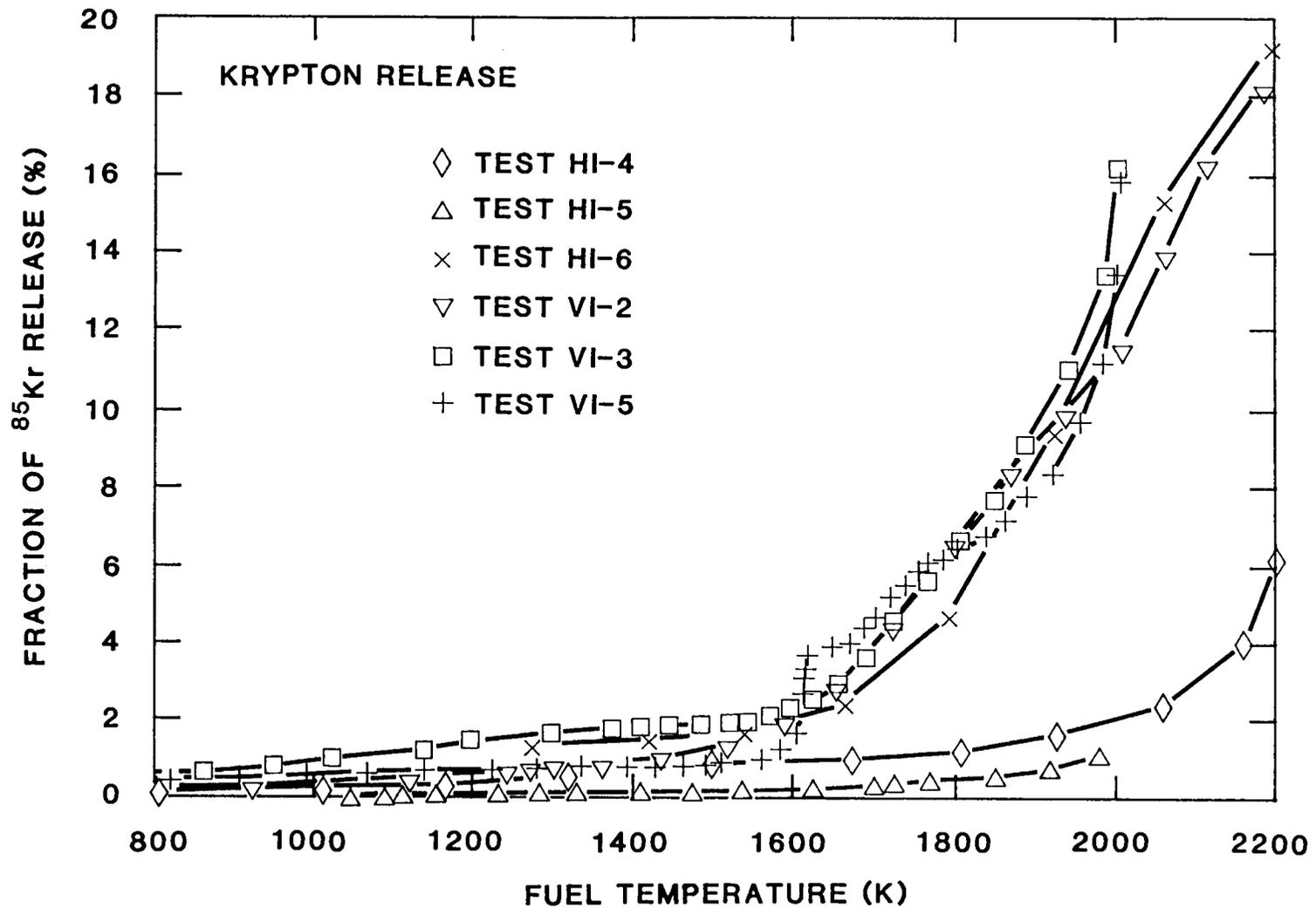


Fig. 17. Integral release of krypton as a function of temperature in six fission product release tests. Note lower release at high temperature for tests HI-4 and HI-5.

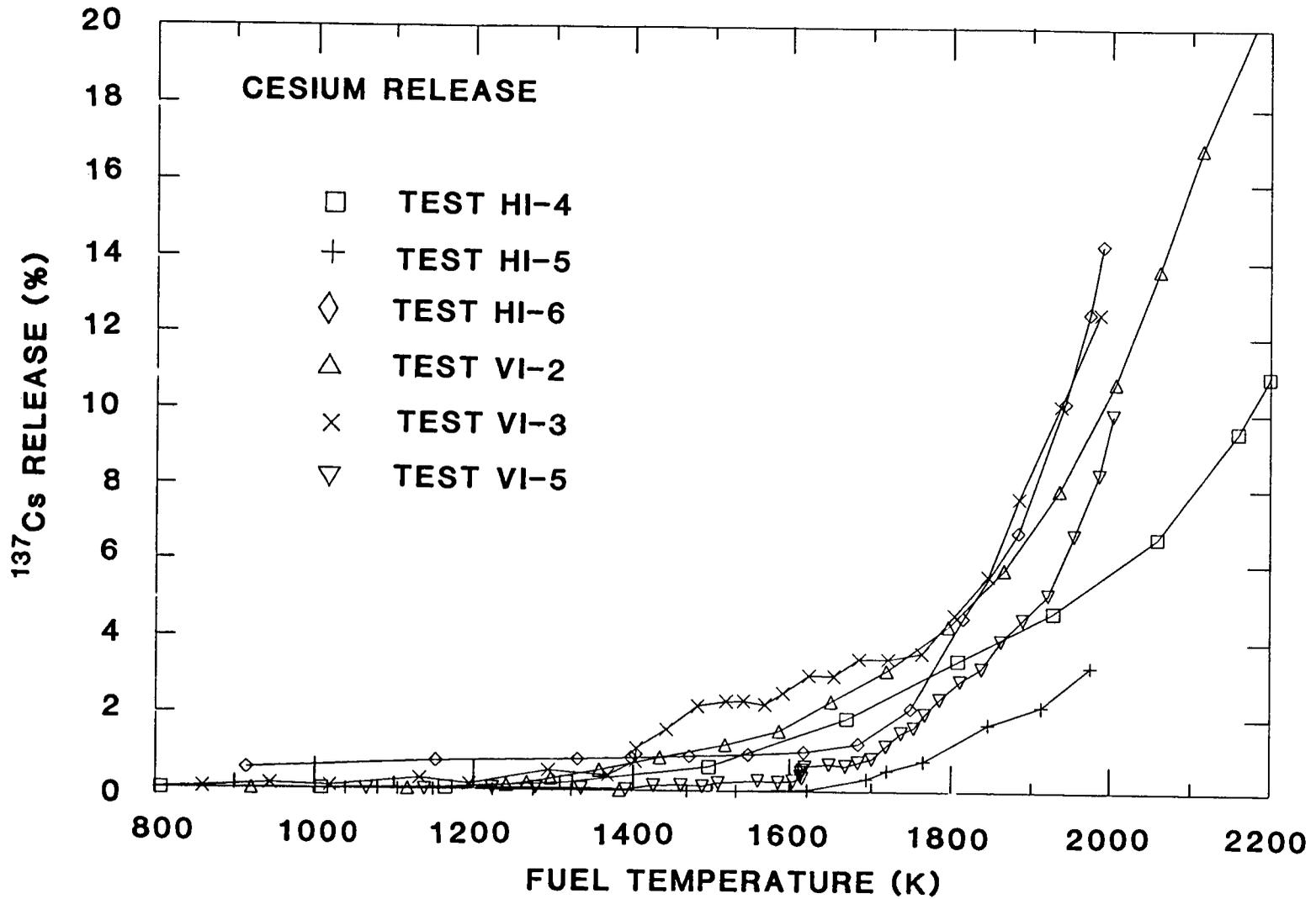


Fig. 18. Integral release of cesium as a function of temperature in six fission product release tests. Only test VI-5 was conducted in hydrogen, all others in steam.

the HI series and the VI series of fission product release tests at ORNL are presented. For krypton (Fig. 17), two tests (HI-4 and HI-5) showed significantly lower release at the higher temperatures, above 1600 K. The reason for these lower values is not apparent but may be related to the fact that the fuel specimens were different - Peach Bottom fuel in test HI-4 and Oconee fuel in test HI-5 - from the other fuels tested. In the comparison of cesium results (Fig. 18), somewhat more scatter in the curves for individual tests is apparent. Again, the HI-4 and HI-5 release values at 1900 to 2100 K are lower than in other tests but are barely outside the scatter band for the six tests. These low temperature releases are believed to be very sensitive to burnup and irradiation temperature, which have strong influences on the linkage of grain boundary gas bubbles to form escape tunnels.

The same on-line data for krypton and cesium were used to calculate the minute-by-minute diffusion coefficients during test VI-5. The results are plotted in Fig. 19 where they are compared with a curve representing the ORNL diffusion model. This model uses data from previous tests, most of which were in steam, and includes the effects of grain size and burnup. As may be seen in Fig. 19, the values from test VI-5 fall slightly above the curve, but follow the general shape of the curve very well. The divergence of the measured values from the model at low temperatures (<1600 K) is based on poor counting statistics and is not considered to be significant.

6. CONCLUSIONS

In view of the preliminary nature of this report and the fact that some important analyses/results have not yet been obtained, a thorough interpretation of the results of test VI-5 is impossible at this time. Upon completion of the currently delayed work, a comprehensive interpretation will be published. However, several significant observations are appropriate at this time.

1. This was the second test at 2700 K for 20 min and the first test at this high temperature in hydrogen. Because all of the test equipment operated well and the planned test conditions were accomplished, the test was considered to be highly successful and the test apparatus and technique were shown to be suitable for future tests of this type.
2. The total release values for the more volatile fission products - especially Kr, Sb, Cs, and Eu - were determined for the specific conditions of this test. The release values were 100% for Kr and Cs, 57% for Eu, and 18% for Sb. Comparison of the release data from test VI-5 with the results from similar previous tests showed generally consistent behavior of the measured fission products. The fractional release of iodine will be determined and reported later. The distribution of the ruthenium remaining in the fuel specimen suggests that some relocation occurred during the test, and additional investigation of this possibility is planned.

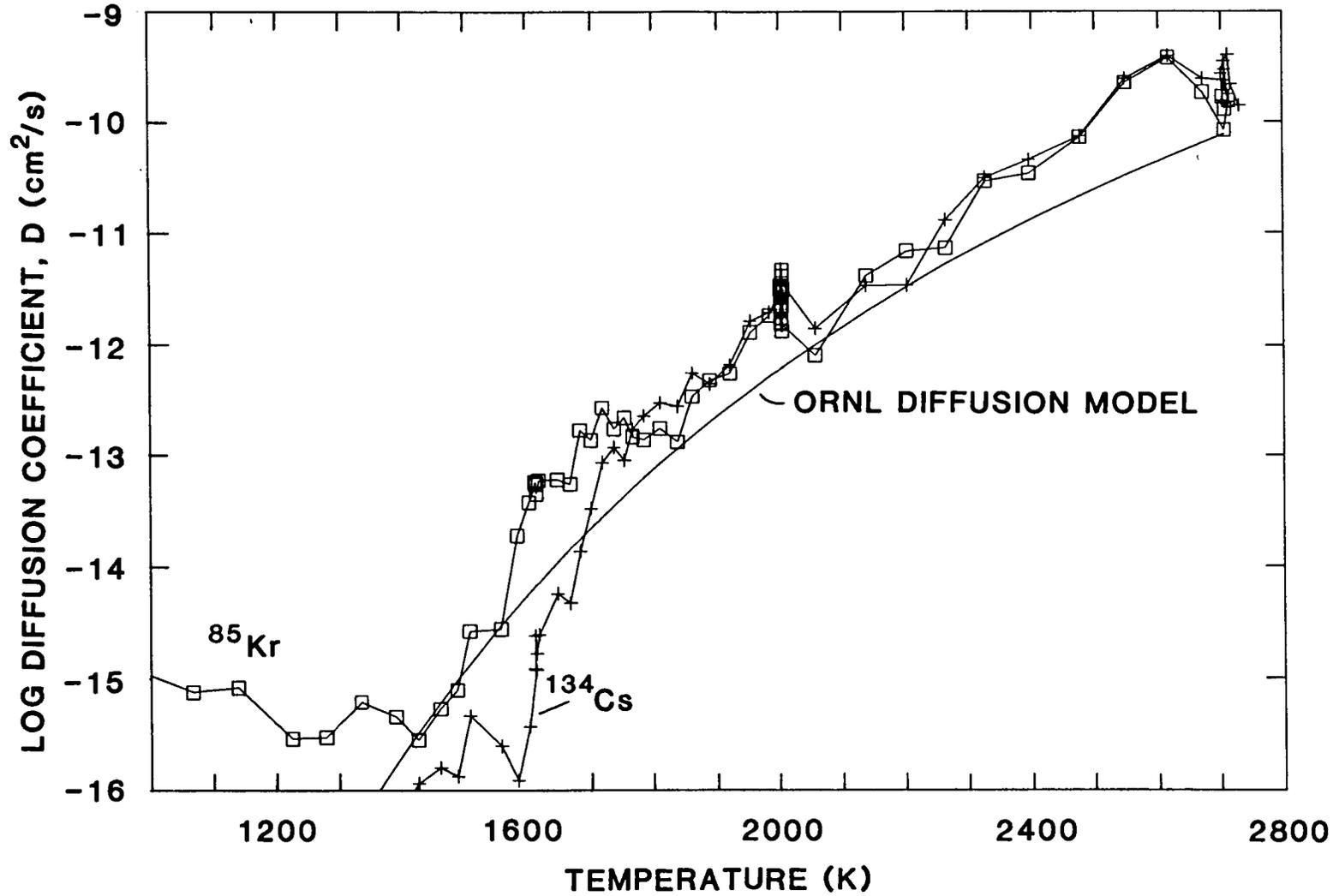


Fig. 19. Comparison of diffusion coefficients for krypton and cesium in test VI-5 with the ORNL diffusion model.

3. Analyses still in progress will supply more information about the release of the non-gamma-emitting fission products (Te, Ba, Mo, etc.) and uranium.
4. Measurements of the masses of deposits collected on the TGTs and the filters verified, as would be expected, that the mass release rates were highest at the maximum test temperature and that the ratio of aerosol to vapor increased with both temperature and time during the test. The total mass released, however, was only ~20% as much as in a similar test in steam, showing the strong effect of the different reactive atmospheres on mass release and transport. This finding is significant in the design and operation of in-containment filtration systems for LWRs and the calculation of aerosol settling rates.
5. Comparison of the results from test VI-5 with those from previous tests showed that the release behavior of krypton and cesium was very similar in almost all tests and in both test atmospheres, steam and hydrogen. The different reactive atmospheres, however, caused drastically different behavior in some fission products: antimony (and probably tellurium) release was much higher in steam than in hydrogen. For europium, and probably for barium and strontium, the release was much higher in hydrogen (test VI-5) than in steam (test VI-3). These behavior variations result from the different chemical properties of these elements.

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71. C. A. Alexander, Battelle Memorial Institute, 505 King Ave., Columbus, OH 43201
72. F. Panisko, Pacific Northwest Laboratory, P.O. Box 999, Richland, WA 99352
73. K. Y. Suh, Fauske & Associates, Inc., 16W070 West 83rd St., Burr Ridge, IL 60521
74. C. S. Lee, INER, Division of Chemistry, P.O. Box 3-6, Lung-Tan, 32500, Taiwan, Republic of China
75. F. C. Iglesias, Ontario Hydro, Nuclear Safety Department, 700 University Avenue, Toronto, Ontario, M5G 1X6, Canada
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M. F. Osborne, R. A. Lorenz, J. R. Travis, C. S. Webster,
J. L. Collins

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10. SUPPLEMENTARY NOTES

11. ABSTRACT (200 words or less)

Test VI-5, the fifth in a series of high-temperature fission product release tests, was conducted in a flowing mixture of hydrogen and helium. The test specimen was a 15.2-cm-long section of a fuel rod which had been irradiated to a burnup of ~42 MWd/kg. Using a hot cell-mounted test apparatus, the fuel rod was heated in an induction furnace under simulated LWR accident conditions to two test temperatures, 2000 K for 20 min and then 2700 K for an additional 20 min.

Based on fission product inventories measured in the fuel or calculated by ORIGEN2, analyses of test components showed total releases from the fuel of ~100% for ⁸⁵Kr, ¹³⁴Cs, and ¹³⁷Cs; 18% for ¹²⁵Sb; and 57% for ¹⁵⁴Eu. Almost 3% of the iodine was collected in a volatile form. In the hydrogen atmosphere, the Zircaloy cladding melted, ran down, and reacted with the UO₂ and some of the fission products. The total mass released from the furnace to the collection system was ~0.681 g. The results from this test were compared with the CORSOR-M and ORNL diffusion release models.

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