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Data Summary Report for Fission Product Release Test HI-6

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Prepared for the
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OPERATED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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Chemical Technology Division

DATA SUMMARY REPORT FOR FISSION PRODUCT RELEASE TEST HI-6

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Prepared for the
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Prepared by the
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ABSTRACT

The sixth in a series of high-temperature fission product release tests was conducted for 1 min at 1950°C in a steam—helium atmosphere. The 15.2-cm-long test specimen was a section of fuel rod which was irradiated to 40.3 MWd/kg in the Monticello BWR.

Based on fission product inventories, analyses of test components by gamma spectrometry and neutron activation showed total releases of 29.6% for ^{85}Kr, 33.1% for ^{137}Cs, 24.7% for ^{129}I, 6.0% for ^{110m}Ag, and 0.06% for ^{125}Sb. Rubidium and bromine were also detected in the collection system by spark-source mass spectrometry. The fractions released of these chemical analogs of cesium and iodine should have been similar to those obtained for cesium and iodine.

Cesium reacted with the oxidized surface in the stainless steel thermal gradient tube; steam oxidation occurred at temperatures >600°C. Cesium that reacted at ~800°C was considerably more difficult to remove by leaching with simulated "LWR coolant" at 53°C than cesium that reacted or otherwise deposited at ~600°C.

The thermal gradient tube deposition profile indicated that iodine probably deposited as cesium iodide.

A comparison was made of Cs, I, and Kr release rate coefficients obtained in the HI and HT test series with NUREG-0772 values. The coefficients obtained in the HI tests were factors of 19 and 3.2 times lower than the HT tests and NUREG-0772 values, respectively.

CONTENTS

	Page
ABSTRACT	iii
FOREWORD	vii
ACKNOWLEDGMENTS	ix
LIST OF FIGURES	xi
LIST OF TABLES	xiii
1. EXECUTIVE SUMMARY	1
2. INTRODUCTION	2
3. TEST DESCRIPTION OF TEST HI-6	3
3.1 FUEL SPECIMEN DATA	3
3.2 EXPERIMENTAL APPARATUS	9
3.3 TEST CONDITIONS AND OPERATING PROCEDURE	14
3.4 POSTTEST DISASSEMBLY AND SAMPLE COLLECTION	14
4. TEST RESULTS	20
4.1 TEST DATA	20
4.2 POSTTEST DATA	23
4.2.1 Results from Gamma Spectrometry	23
4.2.2 Results of Neutron Activation Analysis for Iodine	27
4.2.3 Results of SSMS Analysis	27
4.2.4 Mass of Material Collected in the Thermal Gradient Tube and in the Filter Package	31
4.2.5 Thermal Gradient Tube Results	32
4.2.6 Leachability of ^{134}Cs from Stainless Steel in Simulant LWR Coolant Water at 53°C	32
4.2.7 Results from Analysis of Aerosol Sampler	35
4.2.8 Gamma Spectrometric Analysis of the Fuel Specimen	39
4.2.9 Fuel Examination	41
5. COMPARISON OF RELEASE DATA FROM THE HI AND HT TEST SERIES	41
6. CONCLUSIONS	49
7. REFERENCES	50

FOREWORD

This document is the sixth in a series of reports describing the conduct and results of fission product release testing of commercial LWR fuel. The other reports describing work under this fission product release program are:

1. M. F. Osborne, R. A. Lorenz, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-1, NUREG/CR-2928 (ORNL/TM-8500), December 1982.

2. M. F. Osborne, R. A. Lorenz, J. R. Travis, C. S. Webster, and K. S. Norwood, Data Summary Report for Fission Product Release Test HI-2, NUREG/CR-3171 (ORNL/TM-8667), February 1984.

3. M. F. Osborne, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-3, NUREG/CR-3335 (ORNL/TM-8793), April 1984.

4. 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-4, NUREG/CR-3600 (ORNL/TM-9001), June 1984.

5. M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, and C. S. Webster, Design, Construction, and Testing of a 2000°C Furnace and Fission Product Collection System, NUREG/CR-3715 (ORNL/TM-9135), September 1984.

6. J. L. Collins, M. F. Osborne, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Observed Behavior of Cesium, Iodine, and Tellurium in the ORNL Fission Product Release Program, NUREG/CR-3930 (ORNL/TM-9316), February 1985.

7. K. S. Norwood, An Assessment of Thermal Gradient Tube Results from the HI Series of Fission Product Release Tests, NUREG/CR-4105 (ORNL/TM-9506), March 1985.

8. 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-5, NUREG/CR-4037 (ORNL/TM-9437), May 1985.

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LIST OF FIGURES

Number		Page
1	Dimensions of fuel specimen used in test HI-6	4
2	Gross gamma scan of fuel rod BND-0304 from which the test specimen was cut	8
3	Fission product release furnace	10
4	Fission product release and collection system	11
5	Photograph of (a) fission product release furnace, (b) thermal gradient tube, and (c) filter package in steel containment box before test HI-2	12
6	Components of fission product collection system	13
7	Data acquisition and processing system for fission product release test	15
8	Posttest view of stainless steel thermal gradient tube	19
9	Temperature and flow history for test HI-6	21
10	Collection of ^{85}Kr and ^{134}Cs as functions of temperature and time in test HI-6	22
11	Release rate (% of the initial inventory/min) as a function of test temperature and time for cesium and krypton	24
12	Deposition profiles for cesium and iodine in the stainless steel TGT in test HI-6	33
13	Deposition behavior of iodine in the TGT liners used in tests HI-1 through HI-6	34
14	Rate of removal of ^{134}Cs from stainless steel TGT sections by leaching with a simulant LWR coolant at 53°C	36
15	Miniature aerosol deposition sampler	37
16	Distribution of cesium along the graphite sampler	38
17	Comparison of pretest and posttest ratios of ^{134}Cs (1365 keV) to ^{154}Eu (1244 keV) along the fuel rod segment in test HI-6	42
18	Radial sections through the fuel specimen after test HI-6. The cuts were made at ~2-cm intervals, with section 1 near the inlet end and section 7 near the outlet end	43
19	Comparison of release rate coefficient data from HI and HT tests with curves from NUREG-0772	48

LIST OF TABLES

Number		Page
1	Data for fuel specimen used in test HI-6	5
2	Principal radionuclides and selected stable nuclides in H. B. Robinson fuel specimen HI-6 after 516-d decay	6
3	Amounts of principal fission and activation product elements in Monticello fuel and cladding after 516 d of decay	7
4	Analysis of gas removed from plenum and void spaces of Monticello fuel rod BND-0304 (G-5)	9
5	Summary of changes in apparatus and test conditions for the six HI tests	16
6	Operating data for test HI-6	17
7	Analytical techniques for fission product release analysis	18
8	Distribution and fractional release of fission products in test HI-6	25
9	Posttest distribution of cesium in the experimental apparatus	26
10	Fractional release and distribution of silver and antimony found by gamma spectrometry in test HI-6	28
11	Posttest distribution of iodine in experimental apparatus (results of activation analysis for ^{129}I)	29
12	Results of spark-source mass spectrometric analysis of samples from test HI-6	30
13	Mass of material collected on thermal gradient tube and filter package in test HI-6	31
14	Aerosol sampler data for test HI-6	39
15	Comparison of percentage cesium released as determined by two methods	41
16	Operating conditions for ORNL fission product release tests	45
17	Data summary for fission product release tests	46
18	Release rate coefficients for fission product release tests	47

DATA SUMMARY REPORT FOR FISSION PRODUCT RELEASE FROM TEST HI-6

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K. S. Norwood
J. R. Travis
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1. EXECUTIVE SUMMARY

The objective of this report is to present a thorough documentation of the observations and results collected for fission product release test HI-6. Complete interpretation and correlation of these results with related experiments and with theoretical behavior will be included in a subsequent topical report, which will consider the results of several tests over a range of test conditions. Similar data reports for five previous tests in this series and two other related reports are listed in the Foreword.

The fuel specimen used in this test was cut from a standard fuel rod irradiated in the Monticello BWR from May 1974 to February 1976 to a burnup of 40.3 MWd/kg. The fabrication and irradiation history of this specimen is detailed, and the fission product inventories, as calculated by ORIGEN, are listed in Sect. 3. In addition, the test conditions and post-test analysis procedures are described.

The primary test goals were to heat the specimen at 2000°C for 20 min in a flowing steam–helium atmosphere and to examine the deposition behavior of the released fission products in a stainless steel thermal gradient tube (TGT) liner. Platinum liners have been used in all the other HI tests. Because of a flow restriction in the downstream filter package, the test was terminated after heating the specimen only 1 min at 1950°C. For fission product release calculations which include release during heatup and cooldown, it is recommended that the total effective time for release at 1950°C be 2.5 min.

In Sect. 4 the test results and some preliminary interpretations are presented. The most important results are:

1. Total release values for ^{85}Kr, ^{137}Cs, and ^{129}I were 29.6, 33.1, and 24.7%, respectively. In addition, 6.0% of the ^{110m}Ag and 0.06% of the ^{125}Sb were released.

2. Of the above released nuclides, all of the ^{85}Kr, 99.3% of the ^{129}I, 76% of the ^{137}Cs, 33% of the ^{125}Sb, and 0.3% of the ^{110m}Ag escaped from the furnace to the collection system. The Cs, Sb, and Ag retained in the furnace were found on the ZrO_2 ceramics at the outlet end. Previous tests indicated that CsOH readily reacted with ZrO_2 and was deposited in the temperature range 800 to 1200°C under similar conditions; the most probable reaction product would be Cs_2ZrO_3 (cesium metazirconate).

3. Gamma spectrometric analysis of the fuel specimen before and after the test indicated that 39% of the ^{134}Cs was released. This independent measurement compares reasonably well with the corresponding values based on ORIGEN inventories for ^{137}Cs (33.1%).

4. The deposition profile for cesium in the TGT suggested that a large portion of the cesium reacted with the stainless steel surfaces at the inlet end where the temperature was >600°C during the test and where surface oxidation occurred. Cesium ferrite ($CsFeO_2$) was probably formed.

5. The TGT deposition profile for iodine indicated that it deposited as cesium iodide.

6. Only the fission products mentioned above and rubidium and bromine were detected by spark-source mass spectrometric (SSMS) analysis of samples taken at key locations in the collection system.

7. A brief study of the leachability of cesium from stainless steel TGT sections by simulated LWR coolant at 53°C revealed that it was significantly more difficult to remove cesium that had deposited at higher temperatures (800 compared with 600°C). The leachant contained ~3750 ppm boron and ~1000 ppm sodium at a pH of 7.8.

8. As in previous HI tests, a large fraction of the released iodine (66.3%) and cesium (25.4%) transported to the filter package as aerosol. For the six HI tests, the average fractions of iodine and cesium transported to the filter have been 64 and 31%, respectively.

In Sect. 5, a comparison was made of Cs, I, and Kr release rate coefficients obtained in the HI and HT test series with NUREG-0772 values. The coefficient values obtained in the HI tests were factors of 19 and 3.2 times lower than the HT tests and NUREG-0772 values, respectively.

2. INTRODUCTION

This report summarizes data from the sixth test in a series designed to investigate fission product release from light water reactor (LWR) fuel in steam throughout the temperature range 1400 to ~2400°C.[1] Earlier tests, conducted under similar conditions at temperatures of 500 to 1600°C, have been reported by Lorenz et al.[2-6] 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 heatup accidents in LWRs. The primary objectives of this project are:

1. to determine fission product release from discharged LWR fuel at temperatures up to and including fuel melting (~2400°C);

2. to identify, to the extent possible, the chemical and physical forms of the released fission products;

3. to compare the observed fission product behavior with the physical and chemical changes in the fuel specimens;

4. to correlate the results with data from related programs and develop a consistent source term model applicable to any LWR fuel subjected to a spectrum of accident conditions.

Tests of high-burnup LWR fuel are emphasized in this program; however, the applicability of simulated fuel (unirradiated UO_2 containing a range of fission product elements) will also be investigated, but only at test temperatures >2000°C. As in the first five tests, this test was conducted in a flowing mixture of steam and helium at atmospheric pressure.

Test temperatures in the induction furnace were limited by material properties to a maximum of ~2000°C. Higher-temperature tests are planned but will require the replacement of the ZrO_2 ceramics with ThO_2.

This report provides a fairly complete description of test HI-6 and a tabulation of all the results obtained. As noted in the previous data summary reports,[7-11] no attempt was made to thoroughly evaluate and correlate the data; this will be done and included in a subsequent topical report covering the first series of fission product release tests at temperatures up to 2000°C. However, a comparison was made of Cs, I, and Kr release rate coefficients obtained in the HI and HT tests series with NUREG-0772 values.

3. DESCRIPTION OF TEST HI-6

The primary objectives of this test were to obtain release data at ~2000°C for a period of 20 min in a steam atmosphere and to examine the deposition behavior of the released fission products in a stainless steel thermal gradient tube (TGT)* liner. Because of a flow restriction that developed during the test, the fuel specimen could be maintained at 1950°C for only 1 min. As described in Sect. 5, the effective time for fission product release at 1950°C is estimated to be 2.5 min.

3.1 FUEL SPECIMEN DATA

The test specimen was a 15.2-cm-long section (Fig. 1) of rod BND-0304 (G-5) which operated in the Monticello BWR from May 1974 to February 1980 to a burnup of 40.3 MWd/kg. Details of the irradiation and of the characteristics of this particular specimen are listed in Table 1;[13] calculated fission product inventories for the specimen are shown in Tables 2 and 3.

An axial scan of gamma radioactivity along the fuel rod is shown in Fig. 2. Reductions in burnup resulting from the flux depression caused by the grid spacers are apparent. This gamma scan is dominated by ^{134}Cs and ^{137}Cs but also includes the emissions from such long-lived gamma-emitting nuclides as ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{144}Ce, and ^{154}Eu.

The total gas release (Kr and Xe) from the entire fuel rod was reported to be 1.6%;[13] the gas analysis data are given in Table 4. Since very little gas could have been released from the lower-burnup, lower-temperature regions near the ends of the rod, we assume that all gas released came from the high-burnup region between 51 and 305 cm. We

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*The term "thermal gradient tube" will be abbreviated frequently to TGT.

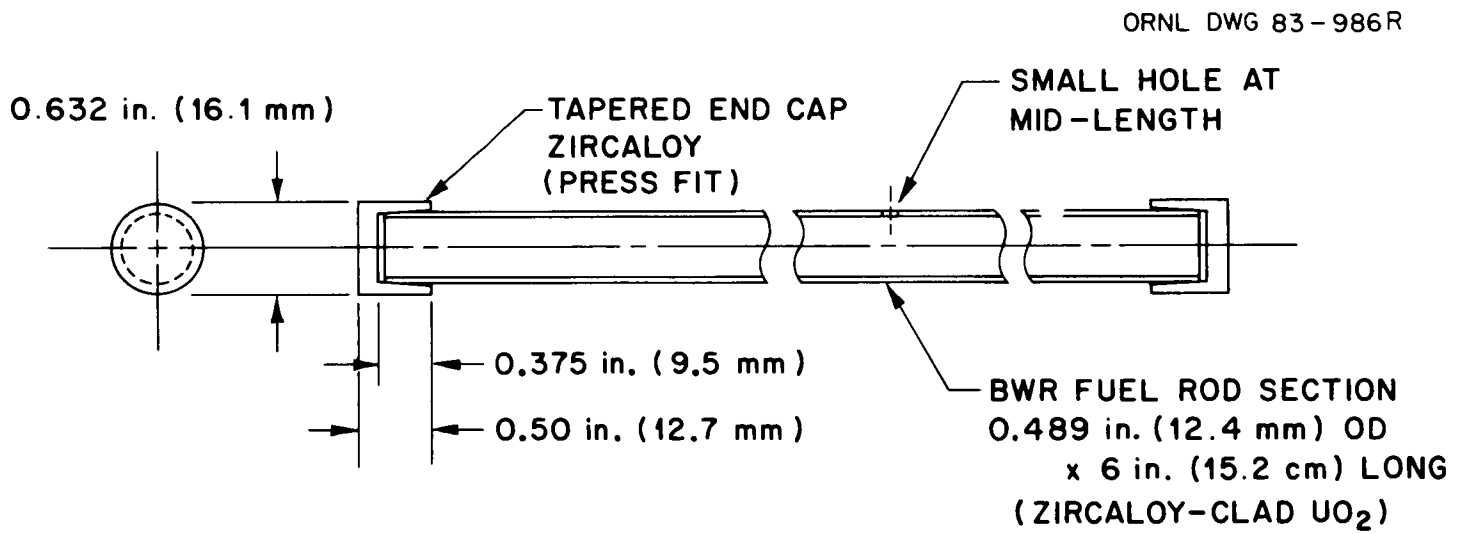

Fig. 1. Dimensions of fuel specimen used in test HI-6.

Table 1. Data for fuel specimen used in test HI-6

Fuel rod identification	BND-0304 (G-5) Section 2
Irradiation data	
Period	May 1974 to February 1980
Average burnup of rod	~31.4 MWd/kg
Burnup of test specimen	~40.3 MWd/kg
Fuel rod characteristics (unirradiated)	
Zircaloy-4 cladding	1.243 cm (0.489 in.) OD 1.072 cm (0.422 in.) ID
Initial enrichment	2.87% ^{235}U
Test specimen characteristics	
Length	15.2 cm (6.0 in.)
Specimen fuel loading	126.0 g UO_2 (112.3 g U)
Weight of Zircaloy cladding and end caps	44.2 g
Total weight of specimen	170.2 g
Gas release during irradiation	1.6% (from whole rod) 2% (from specimen)

Table 2. Principal radionuclides and selected stable nuclides in H. B. Robinson fuel specimen HI-6 after 516-d decay[a]

Nuclide	Amount in fuel		Amount in specimen[b]	
	(g/MT)	(Ci/MT)	(mg)	(mCi)
^{83}Kr	42.51	0.0	4.774	0.0
^{84}Kr	133.3	0.0	14.97	0.0
^{85}Kr	222.8	8,744	25.02	982
^{86}Kr	213.1	0.0	23.93	0.0
^{90}Sr	553.5	75,530	62.16	8,482
^{93}Sr	826.9	2.079	92.86	0.234
^{99}Tc	910.4	15.44	102.2	1.734
^{106}Ru	54.64	182,900	6.136	20,540
^{110m}Ag	0.259	1,231	0.029	138.2
^{113m}Cd	0.324	70.17	0.036	7.880
^{125}Sb[c]	9.180	9,482	1.031	1064.8
^{129}I	221.9	0.039	24.92	0.004
^{128}Xe	5.294	0.0	0.595	0.0
^{130}Xe	20.85	0.0	2.341	0.0
^{131}Xe	464.5	0.0	52.16	0.0
^{132}Xe	1,395	0.0	156.7	0.0
^{134}Xe	1,780	0.0	199.9	0.0
^{136}Xe	2,689	0.0	302.0	0.0
^{134}Cs	89.50	115,800	10.05	13,004
^{137}Cs	1,366	118,900	153.4	13,352
^{144}Ce	58.75	187,500	6.598	21,056
^{147}Pm	76.86	71,280	8.631	8,005
^{154}Eu	50.96	13,760	5.723	1,545
^{155}Eu	16.58	7,714	1.862	866.3
Total[d]	41,470	1,367,000	4,657	153,514

[a]Calculated by C. W. Alexander on Aug. 9, 1984, using the ORIGEN computer program and assuming a burnup of 40.3 MWd/kg and a 516-d decay to July 15, 1981.

[b]Original uranium content of 15.2-cm fuel specimen was 112.3 g; initial enrichment was 2.87% ^{235}U. Thus, the fuel specimen was 112.32 g/10^6 g = 0.01123% of a metric ton.

[c]There was an additional 1.276 g/metric ton of ^{125}Sb formed as cladding activation product (0.143 mg in specimen). Therefore, the total ^{125}Sb in the specimen was 1.174 mg.

[d]Total includes all other isotopes.

Table 3. Amounts of principal fission and activation product elements in Monticello fuel and cladding after 516 d of decay[a]

Element	Mass		Element	Mass	
	g/MT[b]	HI-6 specimen[c] (mg)		g/MT	HI-6 specimen (mg)
Se	65.39	7.34	Sn	108.5	12.18
Br	24.85	2.79	Sb	30.27	3.40
Kr	412.6	46.34	Te	598.5	67.20
Rb	387.4	43.50	I	291.4	32.72
Sr	945.0	106.1	Xe	6354	713.5
Y	508.5	57.10	Cs	3302	370.8
Zr	4108	461.3	Ba	1868	209.7
Mo	4039	453.5	La	1456	163.5
Tc	910.4	102.2	Ce	2865	321.7
Ru	2857	320.8	Pr	1345	151.0
Rh	518.9	58.27	Nd	4765	535.0
Pd	1996	224.1	Pm	76.86	8.63
Ag	106.2	11.92	Sm	954.9	107.2
Cd	164.8	18.52	Eu	205.5	23.07
In	2.16	0.24	Gd	173.9	19.53
Total of all fission products	41,470	4657			
U	9.476×10^5	1.064×10^5			
^{235}U	(3924)	(441)			
Pu	1.00×10^4	1.123×10^3			

[a]Calculated by C. W. Alexander on Aug. 14, 1984, using the ORIGEN computer program and assuming a burnup of 40.3 MWd/kg and a 516-d decay to July 15, 1981.

[b]MT = metric ton.

[c]Original uranium content of 15.2-cm fuel specimen was 112.3 g; initial enrichment was 2.87% ^{235}U. Thus, the fuel specimen was 0.01123% (112.3 g/10^6 g) × 100 of a metric ton.

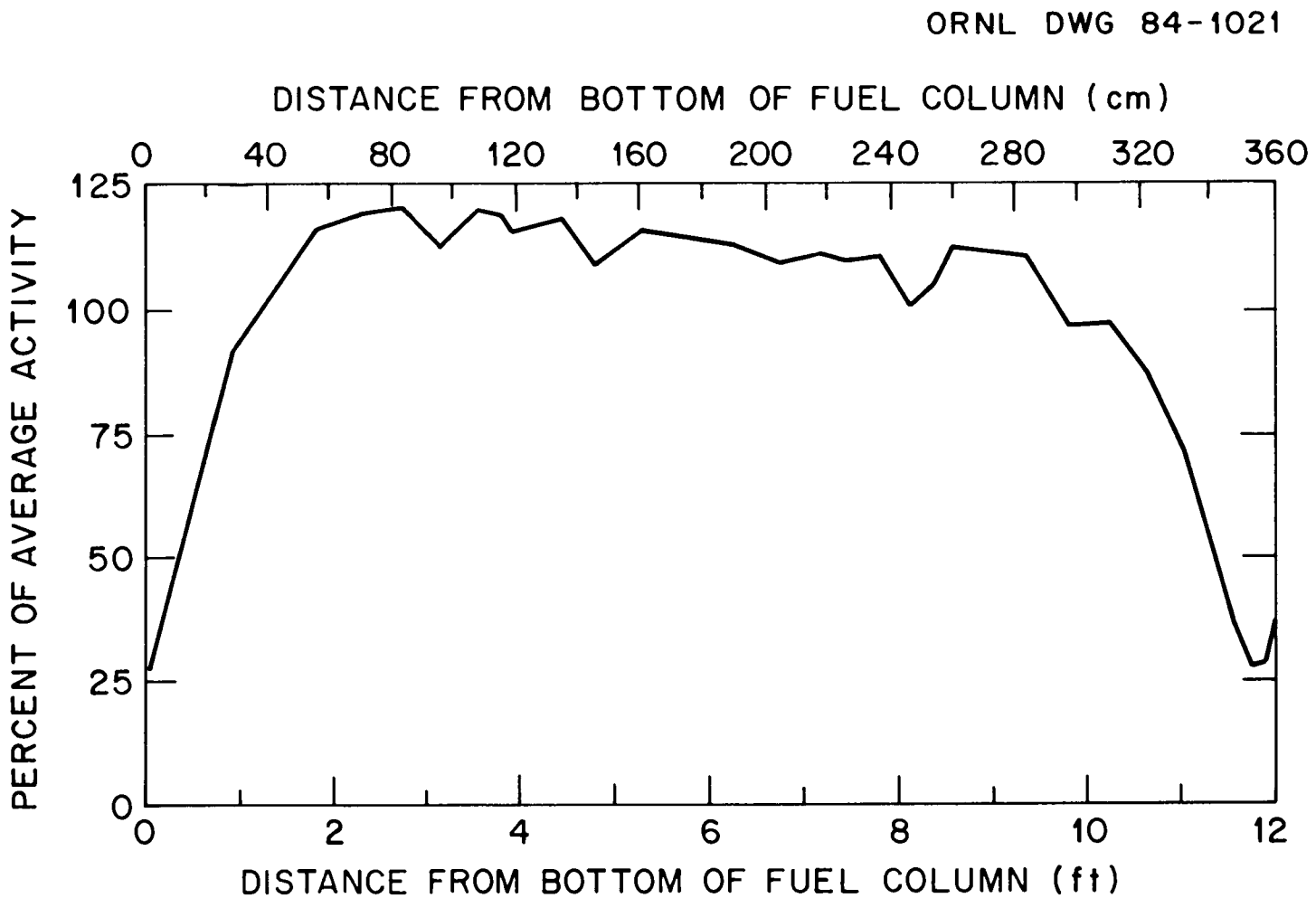

Fig. 2. Gross gamma scan of fuel rod BND-0304 from which the test specimen was cut.

Table 4. Analysis of gas removed from plenum and void spaces of Monticello fuel rod BND-0304 (G-5)

Component	Fraction of total (mol %)	Volume[a] (cm^3, STP)
H_2	<0.1	<0.1
He	56.8	67.4
N_2	<0.2	<0.2
O_2	<0.1	<0.01
Ar	<0.1	0.04
H_2O	<0.1	<0.1
Kr	4.8	5.7
Xe	38.4	45.6
Total		118.7

[a]Analyzed by Battelle Columbus Laboratory in 1981 for DOE.

calculate that ~2% of the krypton and xenon was released from the test specimen during irradiation.

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 UO_2 fuel during subsequent handling. A small hole, 1.6 mm in diam, was drilled through the cladding at midlength to serve as a standard leak for gas release during the heatup phase of the test (Fig. 1).

3.2 EXPERIMENTAL APPARATUS

The fuel specimen was heated in an induction furnace, as illustrated in Fig. 3. This furnace was developed from designs used in previous experimental efforts: fission product release tests,[3-5] fuel rod burst experiments,[14] and molten fuel tests.[15] The graphite susceptor was protected from steam oxidation by a blanket of helium gas that leaked through joints in the ZrO_2 ceramics into the flowing helium stream (see Fig. 3). The furnace was mounted inside a stainless steel containment box in a hot cell, as shown in Figs. 4 and 5. The fission product collection system included a stainless steel TGT, a graphite rod aerosol sampler, fiberglass filters, heated charcoal (for iodine adsorption), and cooled charcoal (for inert fission gas adsorption). The steam was collected in a condenser and a dryer, as indicated in Fig. 6, prior to reaching the cooled charcoal. Instrumentation included two tungsten vs rhenium thermocouples and an optical pyrometer for temperature measurement of the test specimen, three NaI(Tℓ) radiation detectors connected to a multichannel analyzer,

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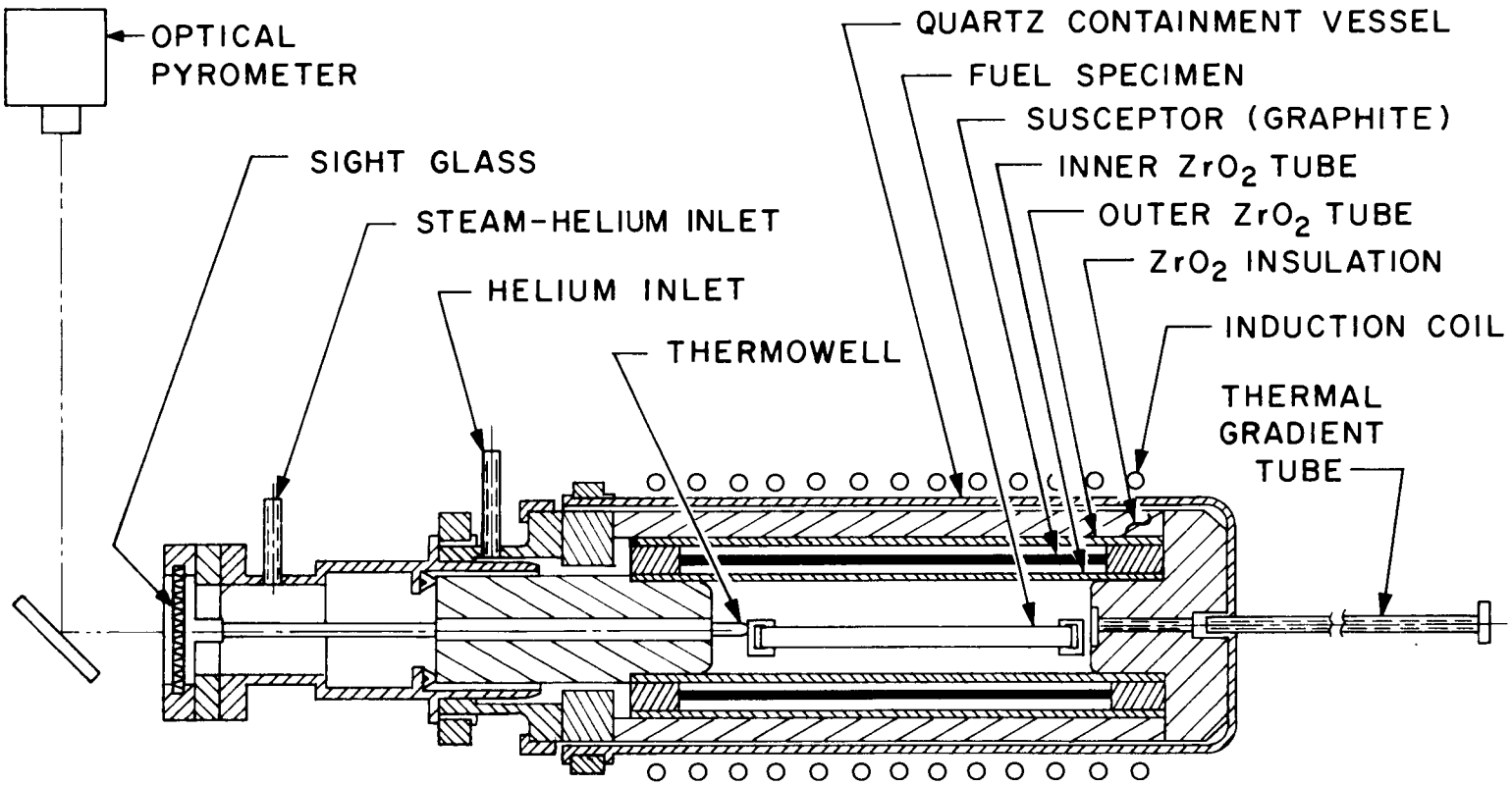

Fig. 3. Fission product release furnace.

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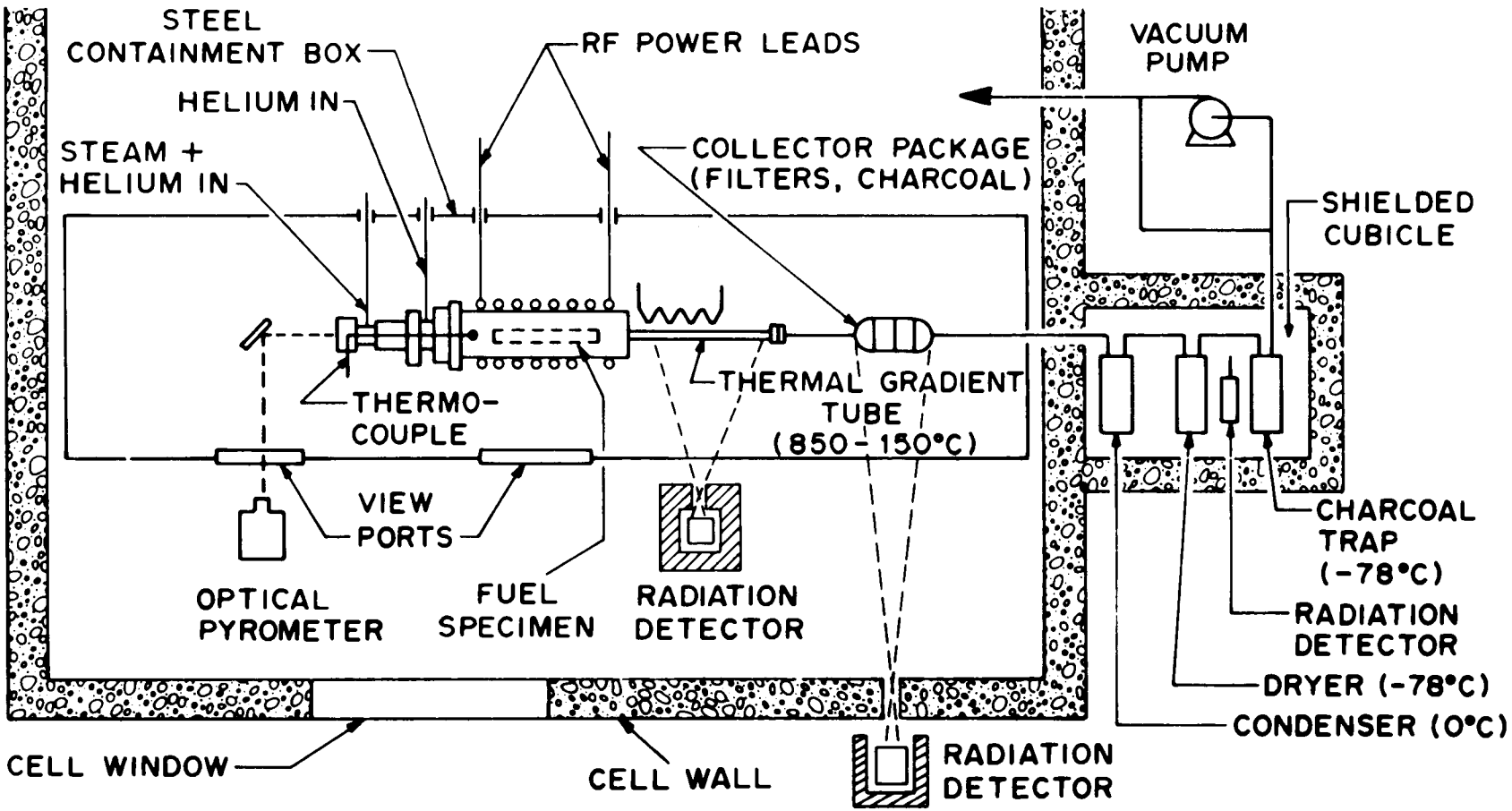

Fig. 4. Fission product release and collection system.

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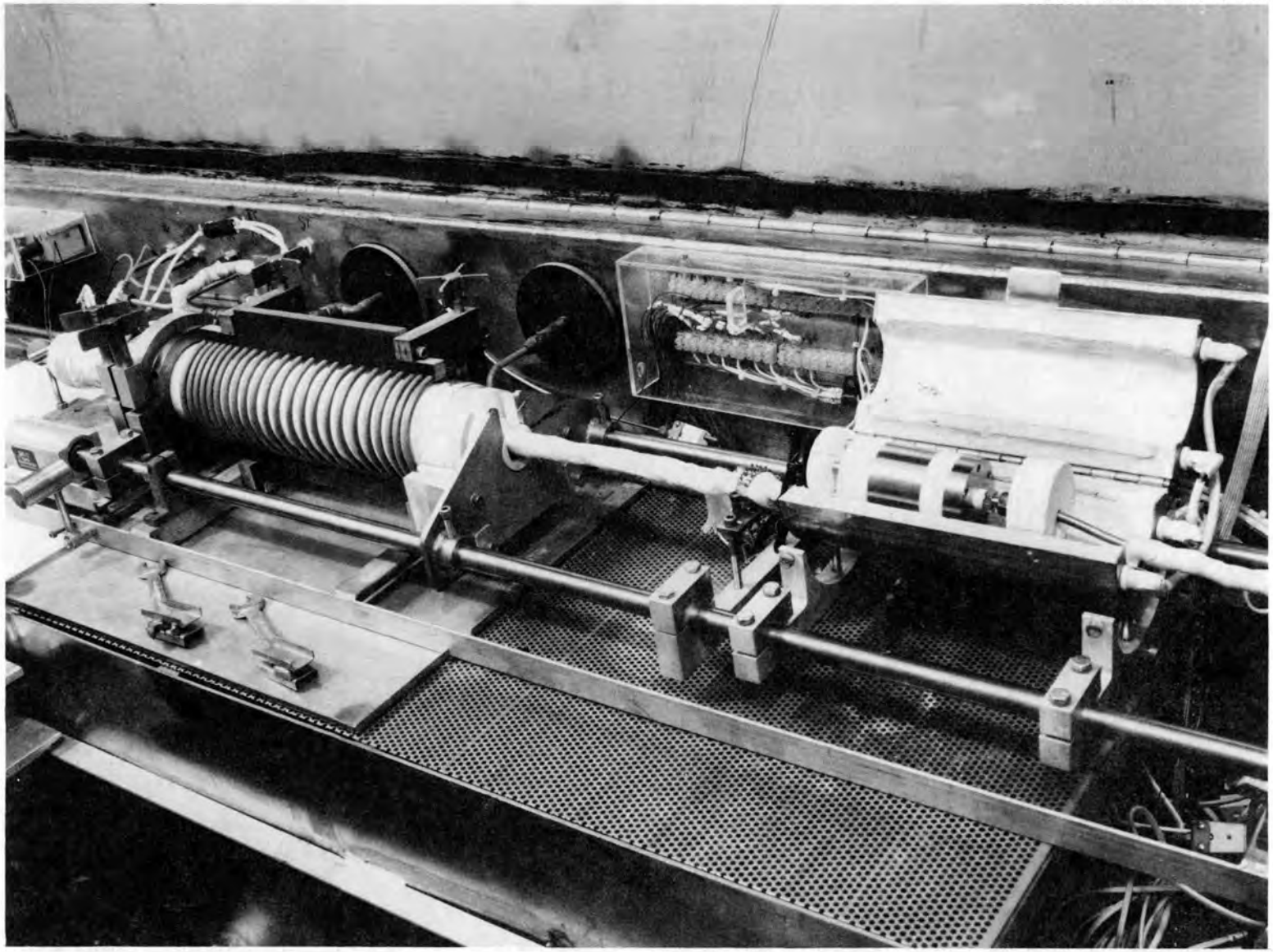

Fig. 5. Photograph of (a) fission product release furnace, (b) thermal gradient tube, and (c) filter package in steel containment box before test HI-2.

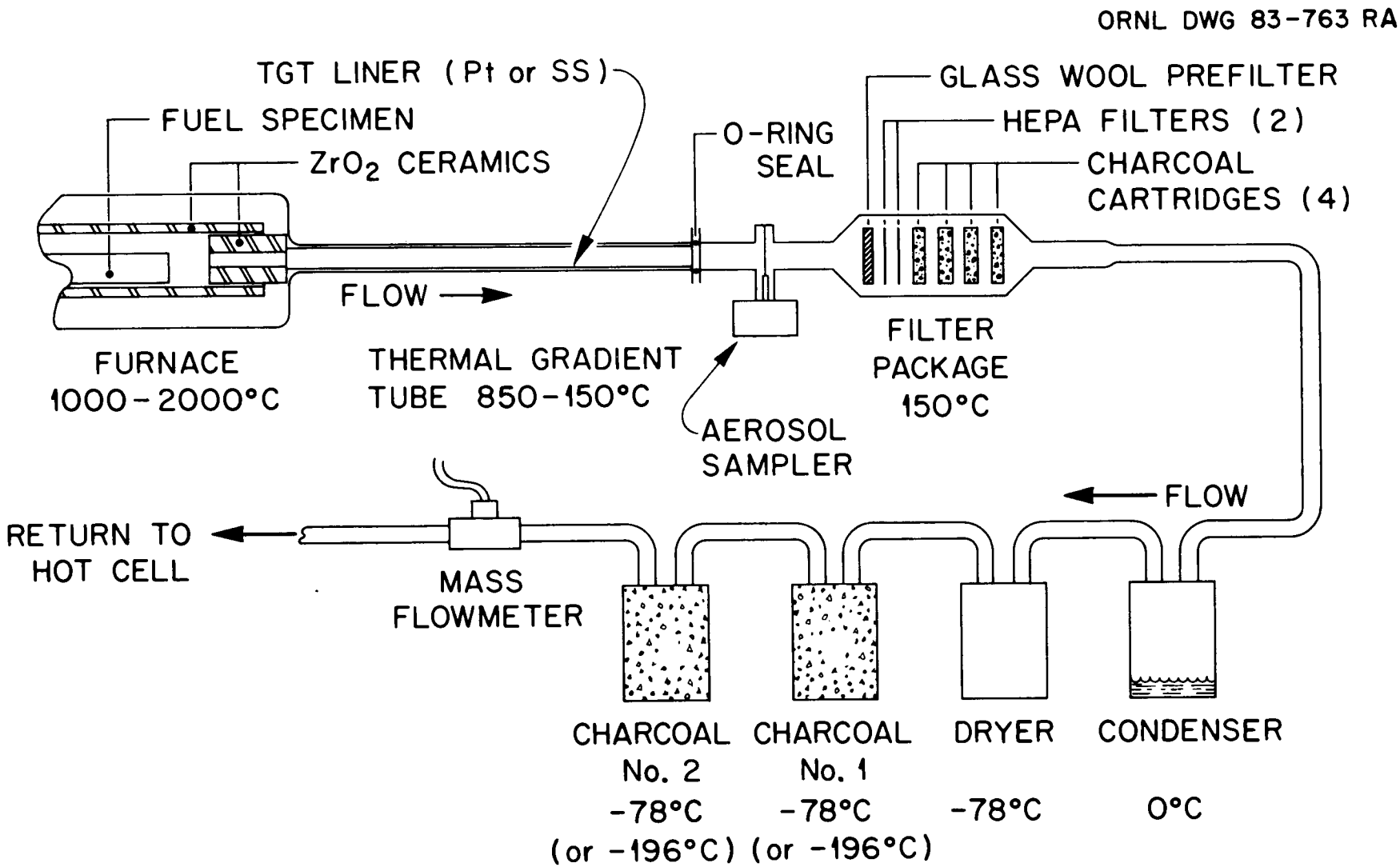

Fig. 6. Components of fission product collection system.

and conventional electrical and gas flow instruments. A data acquisition system (Fig. 7) was used to record test data at 1-min intervals, and several individual chart recorders maintained continuous records of temperatures and flow rates. Differences in apparatus materials and conditions for this test, compared with the previous tests, are summarized in Table 5.

Prior to conducting the test, the furnace ceramics were cleaned and heated to 1000°C in air for several hours to remove organics and impurities. This treatment was helpful, but was not completely effective in removing impurities such as sulfur and chlorine.

3.3 TEST CONDITIONS AND OPERATING PROCEDURE

The HI-6 test was conducted under the operating conditions listed in Table 6. As in each of the previous experiments, the experimental apparatus was assembled by direct handling, which is possible because the hot-cell and test apparatus are decontaminated after each test. Also, new furnace, TGT, and filter package internals are used in each test. Transfer and loading of the highly radioactive fuel specimen and final closure of the furnace and containment box required the use of master slave manipulators. Once the hot cell was closed and sealed, no in-cell operations were required during the test. Prior to testing and before the steam flow was begun, the test apparatus was evacuated, and the furnace was preheated to ~300°C and purged with helium. Also, all connecting lines to the furnace—thermal gradient tube—filter pack assemblies were preheated to at least 125°C to prevent steam condensation during the test.

3.4 POSTTEST DISASSEMBLY AND SAMPLE COLLECTION

After the test was completed, the apparatus was disassembled. The filter package and the TGT liner were removed first and then transferred to another hot cell to avoid potential contamination from fuel handling. Attempts to remove the fuel specimen from the furnace showed that it was attached to the ZrO_2 furnace tube. The fuel specimen end caps could be viewed. Neither was melted; the cap at the inlet end appeared to be oxidized. No cracks in the ZrO_2 furnace tube were noted. To maintain the posttest orientation of the specimen, it was cast in epoxy as was done in test HI-3.[9]

After the highly radioactive test components were removed from the hot cell, the cell was decontaminated. Gamma-ray analyses were carried out on all test components to determine the quantity of each released nuclide. The analytical techniques used are summarized in Table 7. The TGT and fuel specimen were longitudinally gamma scanned at 1-cm intervals to determine fission product distribution. The data for the fuel specimen were compared with pretest data (Sect. 4.2.8). The fuel specimen was subsequently transferred to the High Radiation Level Examination Laboratory, where it was cut into radial sections for detailed inspection (Sect. 4.2.9).

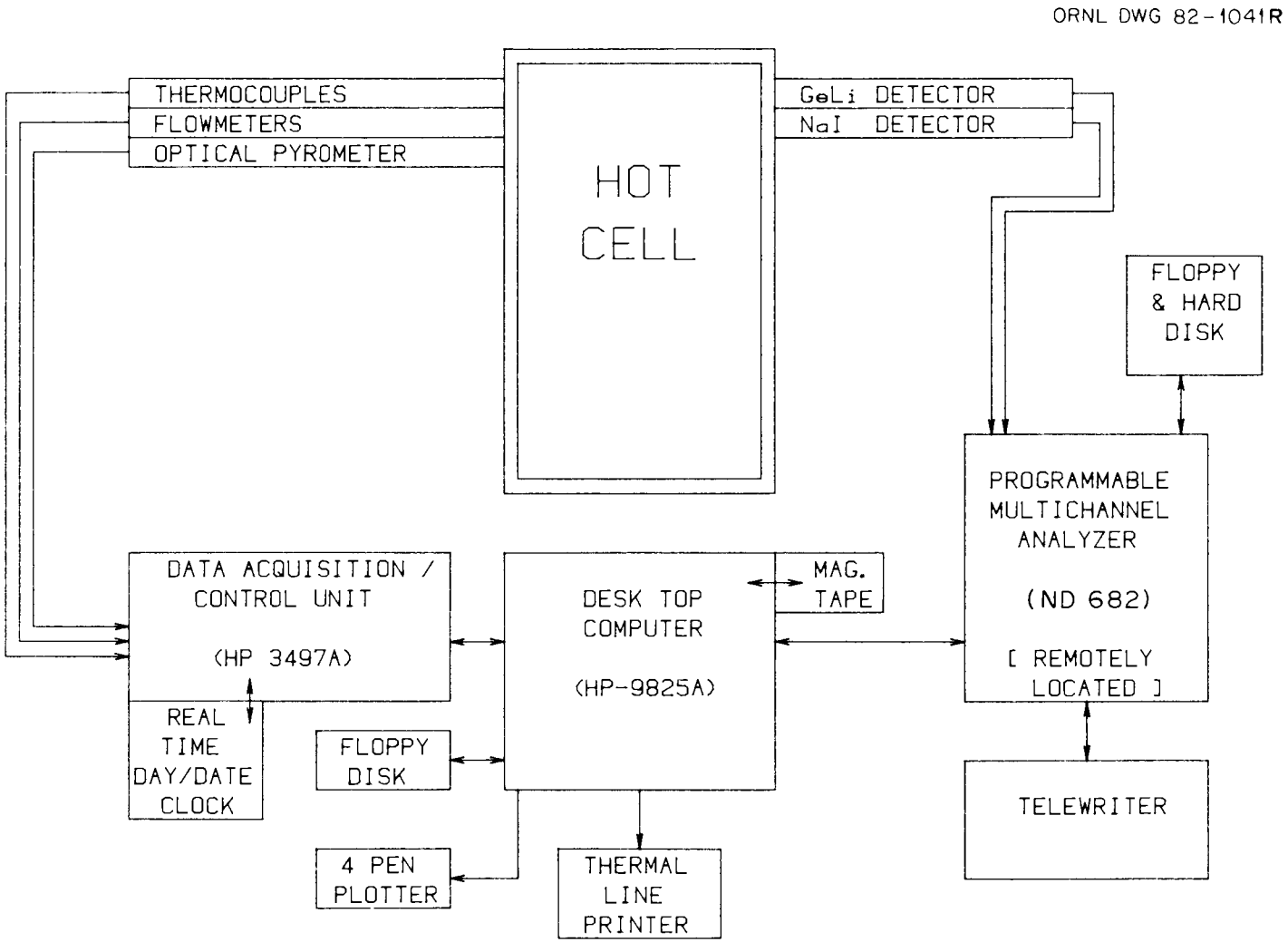

Fig. 7. Data acquisition and processing system for fission product release test.

Table 5. Summary of changes in apparatus and test conditions for the six HI tests

Component/condition	Test HI-1	Test HI-2	Test HI-3	Test HI-4	Test HI-5	Test HI-6
Furnace thermocouple	Pt-10% Rh vs Pt (bare)	Pt-10% Rh vs Pt (bare)	W-5% Re vs W-26% Re (ZrO_2 thermowell)	W-5% Re vs W-26% Re (ZrO_2 thermowell)	Pt-10% Rh vs Pt (bare)	W-5% Re vs W-26% Re (ZrO_2 thermowell)
Susceptor	Tungsten	Tungsten	Graphite	Graphite	Graphite	Graphite
Pretreatment of fibrous ZrO_2 insulator	None	Zirconyl nitrate (1 coat)	Zirconyl nitrate (1 coat)	Zirconyl nitrate (2 coats)	Zirconyl nitrate (1 coat)	Zirconyl nitrate (1 coat)
Thermal gradient tube liner	Platinum	Platinum/gold	Platinum	Platinum	Platinum	Stainless steel
Thermal gradient tube length (cm) diameter (cm)	30.5 0.38	35.6 0.38	35.6 0.38	35.6 0.38	35.6 0.38	37.0 0.425
Connector, thermal gradient tube to filter pack	3.2 mm ID × 76 mm long Teflon	3.2 mm ID × 76 mm long Teflon	4.6 mm ID × 190 mm long stainless steel	4.6 mm ID × 190 mm long stainless seel	4.6 mm ID × 190 mm long stainless steel	4.6 mm ID × 190 mm long stainless steel
Entrance cone to filter pack	Teflon	Teflon	Stainless steel	Stainless steel	Stainless steel	Stainless steel
Glasswool prefilter	30 mm diam, Teflon screen	30 mm diam, Teflon screen	51 mm diam, stainless steel screen	51 mm diam, stainless steel screen	51 mm diam, stainless steel screen	51 mm diam stainless steel screen
Dryer and cold charcoal temperature (°C)	-78	-78	-195	-195	-78	-78
Inert gas	Argon	Argon	Helium	Helium + 0.05% H_2	Helium	Helium

Table 6. Operating data for test HI-6

Specimen temperature at start of heating ramp	250°C
Heatup rate	2.3°/s (135°/min)
Maximum test temperature	2000°C ± 50
Time at maximum temperature	~1 min
Nominal flow rate data:[a]	
He purge to graphite susceptor	0.14 L/min
He to steam generator	0.28 L/min
Steam into system[b]	3.0 L/min
Volume flow data:[c]	
He purge to susceptor	5.25 L
He to steam generator[d]	10.68 L
H_2 generated[e]	11.0 L
CO generated[f]	2.39 L

[a]As measured by mass flowmeters.

[b]Began test at 3 L/min, but decreased to ~0.3 L/min at 1950°C, then recovered to ~1 L/min during cooldown.

[c]As measured by totalizers on mass flowmeters during the 38 min of steam flow into apparatus, operating at room temperature = 23°C.

[d]Absolute pressure in furnace during test was 0.1027 MPa (770 mm Hg).

[e]At atm = 0.0974 MPa (730.2 mg Hg).

[f]Based on 1.28-g weight loss from graphite susceptor during test, assuming all carbon converted to CO.

Table 7. Analytical techniques for fission product release analysis

Technique	Time	Location	Elements
Gamma spectrometry	Pretest, posttest	Fuel specimen	Long-lived, high-energy fission products — Ru, Ag, Sb, Cs, Ce, Eu
	On-line	Thermal gradient tube	Cs, Kr
	Posttest	Furnace components, thermal gradient tube, filters	Ru, Ag, Sb, Cs, Ce, Eu
Activation analysis	Posttest	Charcoal, solution from furnace, thermal gradient tube, filters	I, Br
Spark-source mass spectrometry	Posttest	Samples from furnace, thermal gradient tube, filters	All (detectable) except C, N, O, Be, He, and H
Scanning electron microscope, with energy dispersive x-ray system	Posttest	Graphite rod for sampling aerosol	Elements with atomic numbers ≥11

Upon opening the filter package, it was discovered that it had been inadvertently installed backward. Rather than the aerosol collecting on the glasswool prefilter, it deposited primarily on the charcoal. Much of the charcoal at the entrance was coated with a whitish deposit. This reverse arrangement of the filter package apparently had caused the flow restriction which required the test to be ended prematurely. As the desired test temperature of 2000°C was approached, the system pressure started to build to an unacceptable level forcing the early termination.

Figure 8 shows the posttest view of the stainless steel TGT liner. The pretest cut grooves were spaced at 1-cm-long intervals. This simplified posttest cutting of the TGT for sampling purposes with a tube cutter. Graphite electrode smear samples were taken at five locations for spark-source mass spectrometric (SSMS) analysis. (Samples were also taken of the whitish deposit on the charcoal.)

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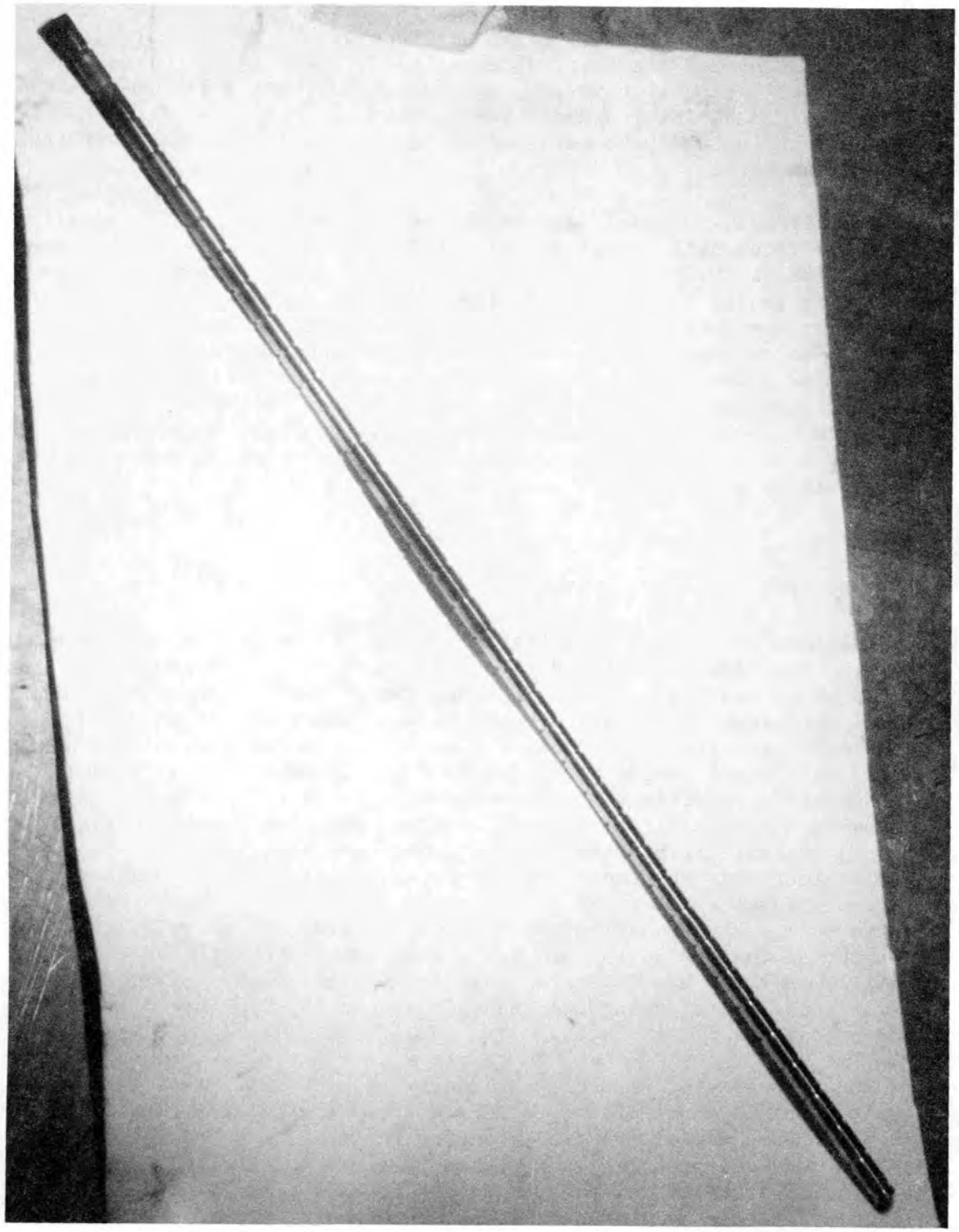

Fig. 8. Posttest view of stainless steel thermal gradient tube.

Each TGT section and each filter was analyzed by gamma spectrometry, before and after leaching successively with basic ($NH_4OH + H_2O_2$) and acidic ($HNO_3 + HF$) solutions. (Two sections of the TGT were leached first in simulated PWR cooling water at 53°C. This is discussed in Sect. 4.2.6.) The quartz vessel, the quartz TGT, and a few of the furnace outlet end ceramic components were also leached in a similar fashion. Iodine release values were obtained by neutron activation analysis of the leach solutions.

The filters, charcoal, and TGT liner were weighed before and after the test. These data showed that the TGT and filter package components gained 0.139 and 0.084 g, respectively. A portion of the weight gained by the TGT was caused by steam oxidation. Most of the visible oxidation occurred at the hotter inlet end where the temperature during the test was >600°C. The two sections of the TGT were disconnected and weighed separately. The inlet section was only 12 cm long, yet ~71% of the weight gain occurred there. Also, ~51% of the increased weight of the filter package components was on the charcoal. The remainder was on the HEPA filter located behind the charcoal. None of the other components in the filter package gained weight.

4. TEST RESULTS

4.1 TEST DATA

The temperature and flow history of the entire test is presented in Fig. 9. These are corrected data. The tip of the thermocouple was located ~2 cm upstream of the inlet end cap of the fuel specimen. The optical pyrometer recorded the surface temperature of the end cap. Based upon special temperature calibration runs, the temperature at the outlet end of the 15.2-cm-long fuel rod segment should have been ~75° cooler. Test operating conditions are summarized in Table 6. The helium flow rates were determined from mass flow rates, and the steam flow rates were based on pretest calibrations of the steam generator and on the amount of water collected in the condenser and dryer (51.9 g). All weight loss from the graphite susceptor (1.28 g) was assumed to result from the steam reaction to form carbon monoxide and hydrogen. Using the data in Table 6 and the reported conversion factors for the measurement of CO and H_2 in the mass flowmeter, it was estimated that ~40% of the 44.2 g of zirconium in the fuel specimen cladding and end caps should have converted to ZrO_2 by steam oxidation.

The release histories of ^{85}Kr to the cold charcoal traps and of the ^{137}Cs to the TGT and filter package are shown in Fig. 10. These values were determined as relative rates during the test, and posttest quantitative measurements were made with a multichannel analyzer to determine the actual fractional releases. The detector for the TGT was positioned at a point where only the lower-temperature end of the tube could be viewed. As indicated by the release curve, significant releases of krypton and cesium occurred as the fuel specimen cooled. (Estimated total cesium release with time is also shown.)

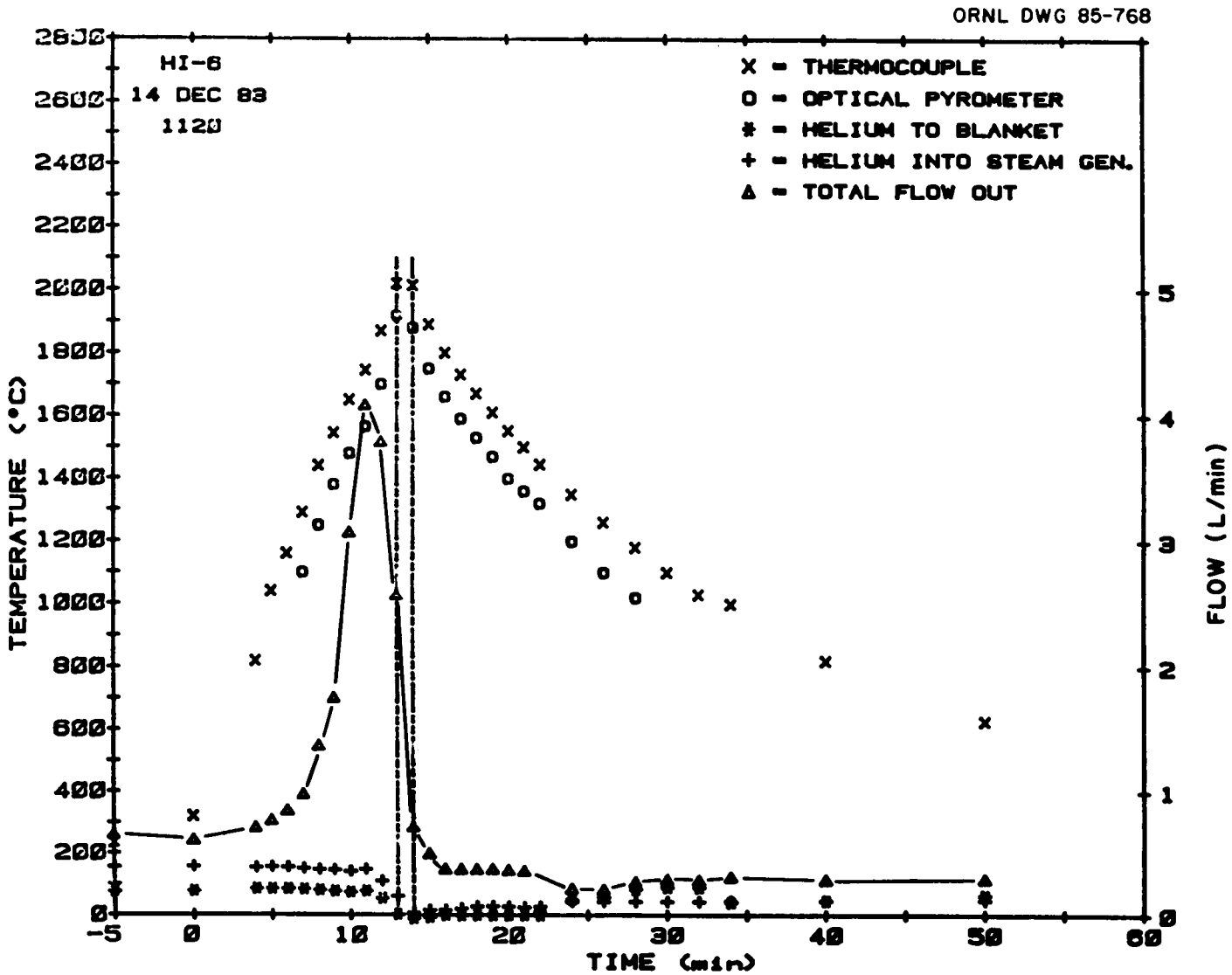

Fig. 9. Temperature and flow history for test HI-6.

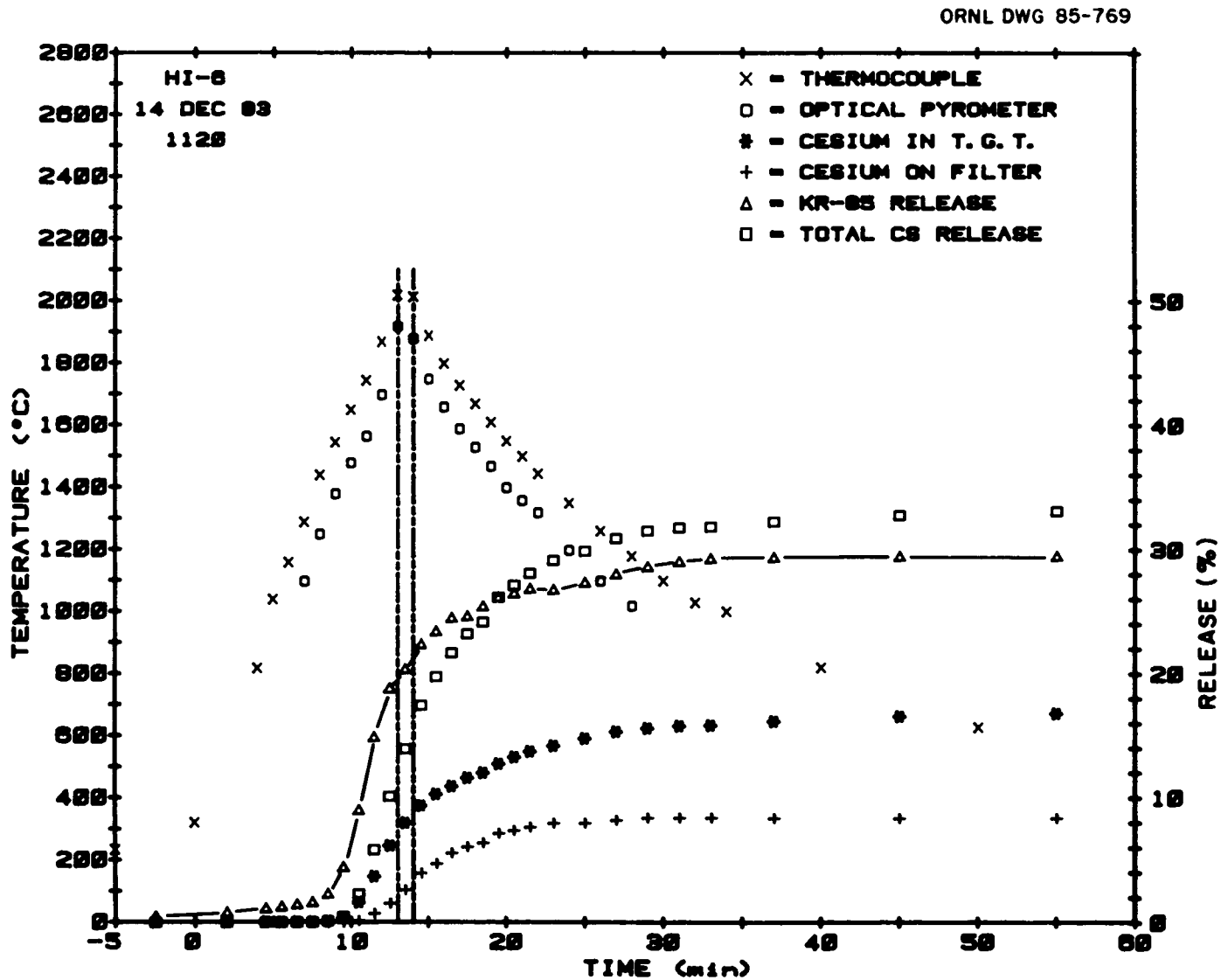

Fig. 10. Collection of ^{85}Kr and ^{134}Cs as functions of temperature and time in test HI-6.

Figure 11 shows how the release rate (% of initial inventory/min) changed for ^{85}Kr and ^{134}Cs with test temperature and time. The largest release rates for both occurred while heating the specimen to the test temperature. A maximum krypton value of 5.9%/min was obtained as the temperature neared 1750°C, but then it declined rapidly as the temperature continued to climb. For cesium, a high value of 4.3%/min occurred as the temperature approached 1850°C; thereafter, the rate declined rapidly, but not as fast as for krypton. Figure 9 shows that the point in time during the test where the helium flow rates to the steam generator and blanket were reduced was coincident with the rapid drop in release rate. Obviously, only the values up to that time are meaningful.

4.2 POSTTEST DATA

Gamma-ray spectrometric analysis of the disassembled apparatus components revealed that the principal radioactive species present were ^{137}Cs and ^{134}Cs. Unfortunately, the high level of cesium activity interfered with the analysis of less abundant fission products. A summary of the fractional release results for the various system components, as determined by gamma spectrometry and by neutron activation analysis for ^{129}I, is presented in Table 8. Note that the krypton and cesium releases are similar, 31.6 and 33.1% respectively, a usual observation in this series of tests. The krypton value includes an estimated 2.0% of the segment inventory which was released to the pellet-clad gap during irradiation. This gas was collected prior to the parent fuel rod being cut into segments.

The apparent release of antimony from the fuel specimen was low, 0.058%. Of the measured antimony that was released, ~67% of it was found on ceramic component surfaces near the inlet end of the test specimen; the remainder was detected at the inlet end of the platinum TGT (after leaching). No antimony was detected (before leaching) in the SSMS samples taken along the TGT or on filter package component surfaces.

Release of antimony has been very limited in the HI tests where the Zircaloy cladding of the test specimens was not completely oxidized; this has been the case in all the tests except for HI-2. Volatile antimony is believed to react with the unoxidized cladding and be retained. Posttest examination of the HI-6 fuel specimen revealed that only the inlet end was heavily oxidized; as was pointed out above, this is near where the antimony was found in the furnace.

4.2.1 Results from Gamma Spectrometry

Table 9 provides the detailed results of gamma spectrometric analysis for ^{137}Cs. Approximately 23.8% of the released cesium remained inside the ZrO_2-lined furnace, primarily on components at the outlet end. Of the ^{137}Cs that escaped the furnace, ~67% deposited in the stainless steel TGT, and the remainder deposited in the filter package. No cesium was found beyond the filter package. About 15 times more cesium was released than iodine. The ORIGEN value for the Cs/I ratio (mass) was 11.3 (Table 3).

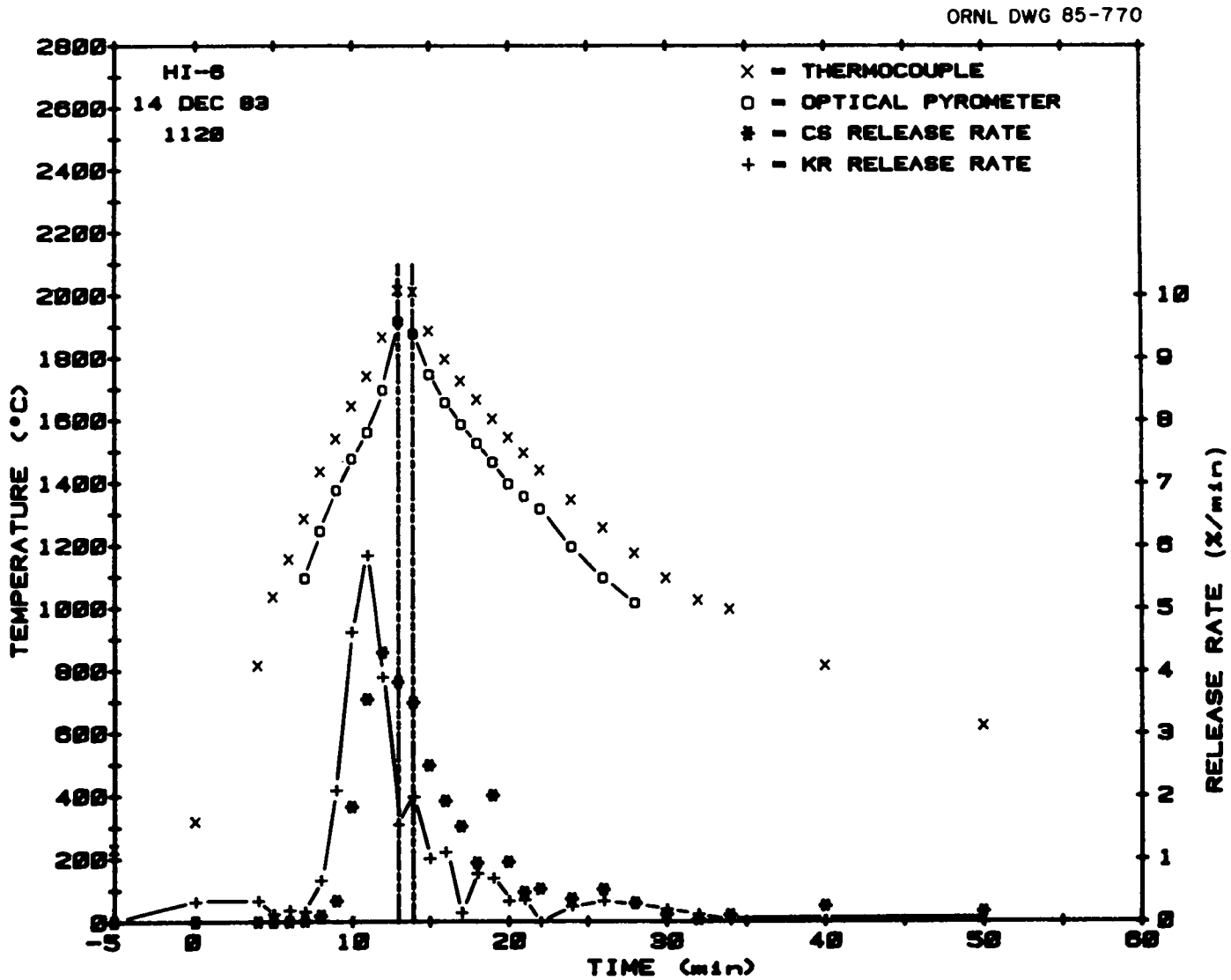

Fig. 11. Release rate (% of the initial inventory/min) as a function of test temperature and time for cesium and krypton.

Table 8. Distribution and fractional release of fission products in test HI-6

Test component or collector	Temperature or range (°C)	Fraction of fission product inventory[a] found (%)				
		^{85}Kr	^{137}Cs	^{129}I	^{125}Sb	^{110m}Ag[b]
Furnace	1000–2000	0	7.88	0.17	0.039	5.94
Thermal gradient tube	180–850	0	16.82	8.15	0.019	0.02
Aerosol sampler	150					
Filter package	140	0	8.40	16.38	0	0
Cold charcoal	-78	29.6	0	0	0	0
Totals		31.6[c]	33.10	24.70	0.058	5.96

[a]Inventory based on ORIGEN calculations by C. W. Alexander.

[b]Large amounts of ^{134}Cs and ^{137}Cs interfered with analyses of ^{125}Sb and ^{110m}Ag prior to leaching.

[c]About 2.0% of the ^{85}Kr was released during irradiation and measured during rod examination. If this amount had not been released previously, it would have escaped during the high-temperature test; therefore, it is included in the total release value for comparison with cesium and iodine data.

Table 9. Posttest distribution of cesium in the experimental apparatus

Location	Temperature (°C)	Cesium found in each location: Amount (μCi ^{137}Cs)	Amount (mg total Cs)	Percentage of specimen inventory[a]	Percentage of released
Furnace components					
Inlet end components	~1000	2.796+3[b]			
Outer ZrO_2 tube	~2000	2.011+2			
Fibrous ZrO_2 insulator	1000–1800	1.768+3			
Graphite susceptor	~2100	1.623+3			
Outlet end components	1000–1500	6.734+4			
ZrO_2 outlet end plugs	1000–2000	9.520+4			
Miscellaneous debris	800–2000	2.398+4			
Quartz vessel	~800	2.280+3			
Total		1.052+6	29.22	7.88	23.80
Thermal gradient tube[c]	850–180				
Quartz tube		6.793+2			
SS section 1		8.179+5			
SS section 2		8.416+5			
SS section 3		3.732+5			
SS section 4		1.805+5			
SS section 5		3.224+4			
Total		2.246+6	62.37	16.82	50.81
Filter package[d]	~140				
Total		1.122+6	31.16	8.40	25.39
Other components					
Condenser	0	0.0			
Dryer	-78	0.0			
Cooled charcoal	-78	0.0			
Total		0.0	0.0	0.0	0.0
Total all components		4.420+6	122.75	33.10	100.0

[a]Based on an average burnup of 40.3 MWd/kg, the test fuel specimen contained 370.81 mg Cs and 13.352 Ci of ^{137}Cs (= 2.78×10^{-5} mg Cs/μCi ^{137}Cs); decay corrected to July 15, 1981 (ORIGEN calculation).

[b]Exponential notation: 2.796+3 = 2.796×10^3, etc.

[c]A stainless steel thermal gradient tube was used rather than platinum.

[d]The filter package was inadvertently installed backward. About 98% of the cesium was found on the charcoal and adjacent HEPA filter paper (~52% on charcoal).

The large concentration of cesium found on the zirconia outlet end plug indicates reaction probably occurred (perhaps the formation of cesium zirconate). This has been observed in all the HT tests.

Table 10 gives the distribution of ^{110m}Ag and ^{125}Sb in the apparatus as determined by gamma spectrometry. The release values of 5.96 and 0.06% for Ag and Sb, respectively, are much lower than the values for Cs, I, and Kr (Table 7) as observed in all tests. However, it should be noted that the silver and antimony values are minima because of the interference from cesium. Most of the released silver (99.8%) was found on the zirconia ceramic plugs which were positioned at the outlet end of the zirconia furnace liner. Also, ~67% of the detected antimony was found on these plugs; the remainder was found at the entrance end of the stainless steel TGT (first cm).

4.2.2 Results of Neutron Activation 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. Iodine forms dissolve readily in basic solutions to form stable iodides; in our samples, large amounts of highly radioactive cesium were also dissolved. Small aliquots of the solutions were chemically treated to remove cesium, loaded onto ion-exchange media, and irradiated, and the 12.4-h ^{130}I was counted. The results of these analyses, along with data for fractional iodine release and cesium/iodine ratios at various apparatus locations, are summarized in Table 11. The total fraction of iodine released, 24.7%, was a minimum value. Since iodine cannot be detected directly, it must be leached from the surfaces of the components as discussed above. With this procedure, there is always some uncertainty about the iodine being completely removed by the leaching steps. Because the filter package was installed and used in reverse order during the test, the amount of elemental iodine could not be determined. In the previous tests of the HI series, the average percentage of the released iodine to reach the filter package was 63.6. In this test, 66.3% was found in the filter package, thus demonstrating similar partitioning between condensation in the TGT and association with particles, 62% was found on the charcoal, and the remainder on the HEPA filter located behind the charcoal. Only 0.03% was found beyond this filter.

4.2.3 Results of SSMS Analysis

Elemental analyses were obtained by SSMS for graphite electrode smear samples at five locations along the stainless steel TGT liner and one from the whitish deposit on the charcoal in the filter package. Following the sampling, the amount of ^{137}Cs in each sample was determined by gamma spectrometry. As shown in Table 12, the only fission products detected, other than cesium and iodine, were the chemical analogs, rubidium and bromine. The major impurities that were transported from the furnace components to the collection system were S, Cl, Al, Ca, Zn, P, and Na. Stainless steel elements (Fe, Cr, and Ni) that were made available for

Table 10. Fractional release and distribution of silver and antimony found by gamma spectrometry in test HI-6

Location	Temperature (°C)	Silver			Antimony		
		(μCi ^{110m}Ag)	(μg Ag)[a]	Percentage of inventory	(μCi ^{125}Sb)	(μg Sb)[b]	Percentage of inventory
Furnace components							
Inlet end components	~1000	17.4			22.3		
ZrO_2 insulator tubes	1000–1800				ND[c]		
Graphite susceptor	2100				ND		
Outlet end components	1000–2000				ND		
ZrO_2 outlet end plugs	1000–2000	8198			392.3		
Miscellaneous debris	800–2000						
Quartz vessel	~800				ND		
Total		8213.4			414.6		
Thermal gradient tube	180–850	12.5[d]			202.8[e]		
Filter package	140	3.8			0		
Total all components		8230	710	5.96	617.4	2.0	0.06

[a]Based on inventory data: 11.92 mg Ag and 138.2 mCi ^{110m}Ag in specimen.

[b]Based on inventory data: 3.40 mg Sb and 1064.8 mCi ^{125}Sb in specimen.

[c]ND = not determined.

[d]Detected only after leaching.

[e]Located on first cm of TGT at the entrance.

Table 11. Posttest distribution of iodine in experimental apparatus (results of activation analysis for ^{129}I)

Location	Temperature (°C)	Iodine found at each location: Amount (μg ^{129}I)	Amount (μg total I)	Percentage of specimen inventory[a]	Percentage of released	Cs/I ratio (μg Cs/μg I)
Furnace components						
Quartz vessel	~800	2.05	2.69			23.6
ZrO_2 outlet end plugs	1000–2000	41.54	54.54			485.6
Total[b]		43.59	57.23	0.17	0.71	463.9
Thermal gradient tube[c]	850–180					
Quartz tube		0.18	0.24			78.7
SS section 1		13.53	17.76			1281.2
SS section 2		5.13	6.74			3473.8
SS section 3		143.80	188.81			55.0
SS section 4		1699.50	2231.45			2.3
SS section 5		167.48	219.90			4.1
Total		2029.62	2664.90	8.15	32.97	23.4
Filter package[d]	~140					
Total		4082.09	5359.79	16.38	66.32	5.8
Other components						
Condenser	0	0.04	0.05			
Total		0.04	0.05	(1.6×10^{-4})	(0.001)	
Total all components		6155.34	8081.97	24.70	100.00	15.3

[a]Based on an average burnup of 40.3 MWd/kg, the test specimen contained 24.92 mg ^{129}I and 32.72 mg total iodine; decay corrected to July 15, 1981.

[b]Small amounts of iodine probably were present on other components of the furnace that were not leached.

[c]A stainless steel thermal gradient tube was used rather than platinum.

[d]The filter package was inadvertently installed backward. About 96% of the iodine was found on the charcoal and adjacent HEPA filter paper (~60% on the charcoal).

Table 12. Results of spark-source mass spectrometric analysis of samples[a] from test HI-6

Thermal gradient tube sample No.	Sample location (distance from inlet end of TGT) (cm)	Elemental fraction (at. %)														
		Cs[b]	Rb[b]	I[b]	Br[b]	Ca	K	Na	Al	Fe	Cr	Ni	Cl	S	Si	P
1	0.3	0.84	0.54			3.8		7.5	19.0	1.2		2.0	24	37		1.1
2	5.3	2.8	2.3			0.45	0.18	0.77	0.53	38	7.0	3.0	2.4	33	8.9	0.02
3	12.3	0.92	1.1			4.5		0.29	8.0	4.0		3.8	20	56		1.4
4	17.3	3.5	1.0	0.87		8.0	1.1	0.70	6.1	12			14	52		1.0
5	27.3	3.2				12	2.0	1.8	17	17			47			
Filter package		10	3.8	3.9	1.5	0.12	0.30	25	0.13	0.21	0.16	0.10	2.0	37		7.7

[a]Samples were taken of surface deposits at key locations along the thermal gradient tube and also from deposited material in the filter package.

[b]Fission products; all the other elements given came from apparatus materials.

sampling by oxidation of the TGT at temperatures >600°C were also detected.

Neutron activation analysis revealed that iodine deposited primarily in the filter package and in a narrow peak in the TGT (see Sect. 4.2.6). This was also indicated by the SSMS data.

4.2.4 Mass of Material Collected in the Thermal Gradient Tube and in the Filter Package

As observed in previous tests in this series, deposits of significant mass were found in the TGT and in the filter package. The masses of material collected in the TGT and filter package are listed in Table 13. In addition to the materials released from the fuel specimens and furnace vessel components, these data include an undetermined mass of oxygen in the form of oxides of the components of Inconel and stainless steel at the inlet (high temperature >600°C) end of the TGT. Approximately 45% of the increased weight in the TGT and in the filter package was due to the deposition of cesium and iodine. If it is assumed that the release fraction of rubidium, the chemical analog of cesium, was similar to that of cesium, the fractional contribution of the weight gain in the TGT and filter package due to fission products would be 50%. The fission products detected in the TGT and filter package by the various analytical methods were Cs, I, Rb, Br, and Sb. The weight contributions by antimony and bromine were negligible. The major nonfission product elements detected in the TGT (by SSMS) other than the constituents of stainless steel were, in decreasing order of dominance: S, Cl, Al, and Ca. In the filter package, the major ones were S, Na, and Cl.

Table 13. Mass of material collected on thermal gradient tube and filter package in test HI-6

Component	Mass[a] (mg)
Thermal gradient tube	
Section 1 (12 cm)	102[b]
Section 2 (25 cm)	42
Filter package[c]	
Charcoal	~40
Filters	41
Total	~225

[a]Weight increase by direct measurement.

[b]At the higher-temperature end, oxidation of the stainless steel probably contributed significantly to this value.

[c]In this test, the filter package was inadvertently reversed so that the charcoal cartridges preceded the filters. This reversal probably caused the observed flow restriction.

4.2.5 Thermal Gradient Tube Results

The type 304L stainless steel (~67% Fe, ~20% Cr, ~10% Ni, 2% Mn, 1% Si) TGT was 37.15 cm long and was composed of two interconnected sections with the inlet end section being 12 cm long (Fig. 8). The inner diameter was 0.425 cm.

Figure 12 shows the deposition profiles for cesium and iodine in the TGT. The surface concentration of cesium rose and fell several times between 2 and 5 mg/cm (of length) from the entrance to ~18 cm downstream where the temperature gradient during the test had been between 850 and 500°C. Beyond that point, the concentration decreased rapidly. Visual inspection of the cut sections revealed that the inner surfaces of the sections that were heated to temperatures >600°C were oxidized with the degree of oxidation appearing to be heavier with increased temperature. Inspections were made before and after leaching, and the surfaces of sections ≤600°C showed little or no evidence of oxidation. Similar oxidation behavior was observed in the simulant test C-8[16] where a stainless steel TGT was also used.

The cesium peak at 500°C appears to be associated with the iodine peak. The location of the iodine peak is in the temperature region where CsI has been observed to deposit in simulant control tests. The iodine peak location in test HI-6 is shown also in Fig. 13 compared with the iodine peaks that were obtained in the other HI tests.

The relatively flat cesium profile at the inlet end (Fig. 12) where the steam oxidation occurred indicates that cesium reaction with the surface may have occurred. The most probable reaction products are cesium ferrite,[17] cesium chromate,[18] and cesium silicate.[19]

4.2.6 Leachability of ^{134}Cs from Stainless Steel in Simulant LWR Coolant Water at 53°C

The collection behavior of cesium in the stainless steel TGT in this test was different from that observed in previous tests where platinum thermal gradient tubes were used. The TGT profile for cesium in Fig. 12 shows that the most significant fraction deposited at the hot inlet end starting at the entrance at ~880°C and extending to ~19 cm downstream, where the temperature was ~500°C. In most of the previous tests, on the other hand, major cesium peaks were observed in the temperature range 600 to 300°C.

A leach study was conducted to examine the leachability of cesium from two sections of the stainless steel TGT; section 1 from the hottest region (840 to 800°C), which was a 6-cm-long section at the entrance, and section 3 from a cooler region (675 to 550°C), which was located 12 to 16.9 cm from the entrance.

To make the study meaningful, a stock leach solution was prepared that simulated cooling water which might be found following pressurized

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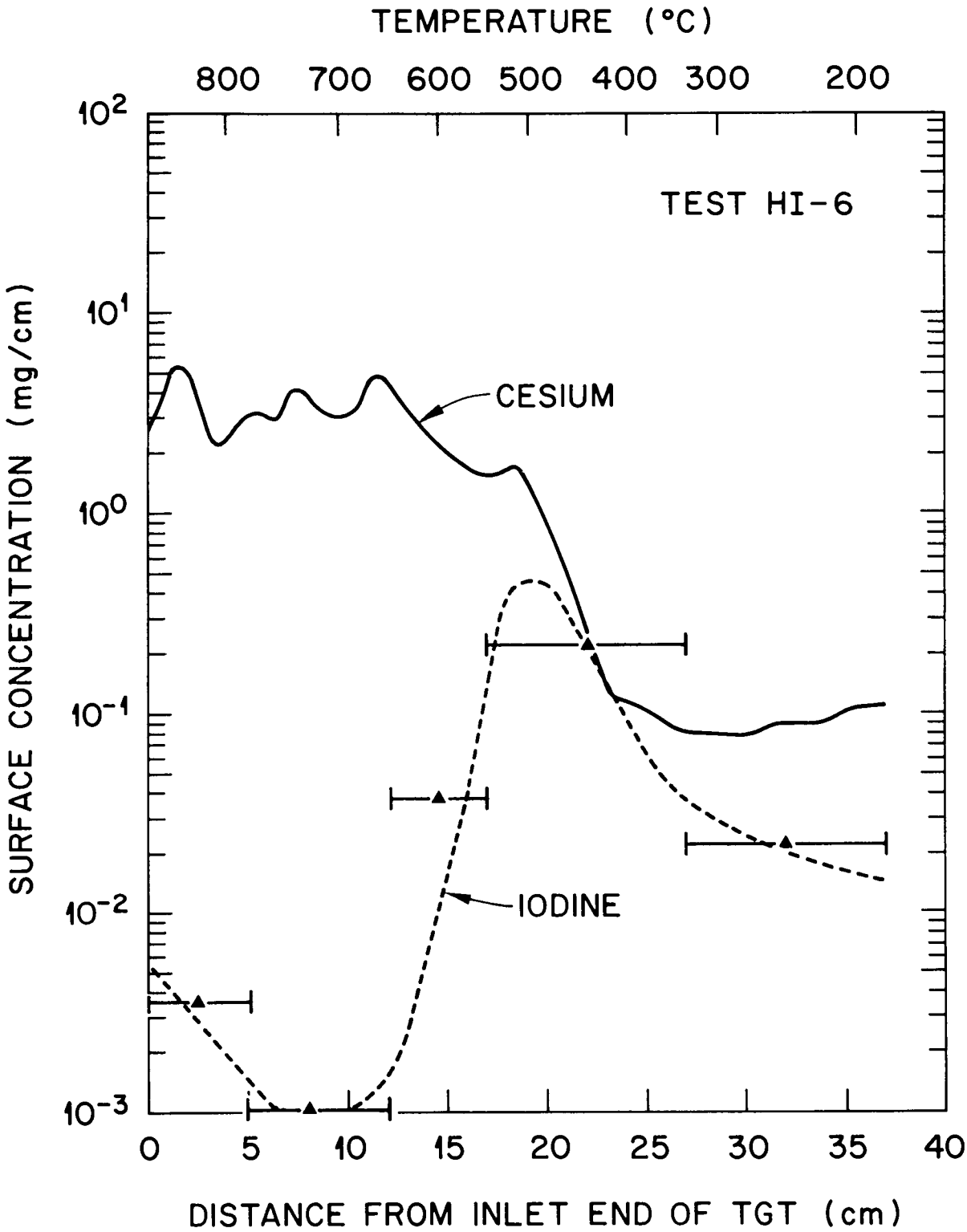

Fig. 12. Deposition profiles for cesium and iodine in the stainless steel TGT in test HI-6.

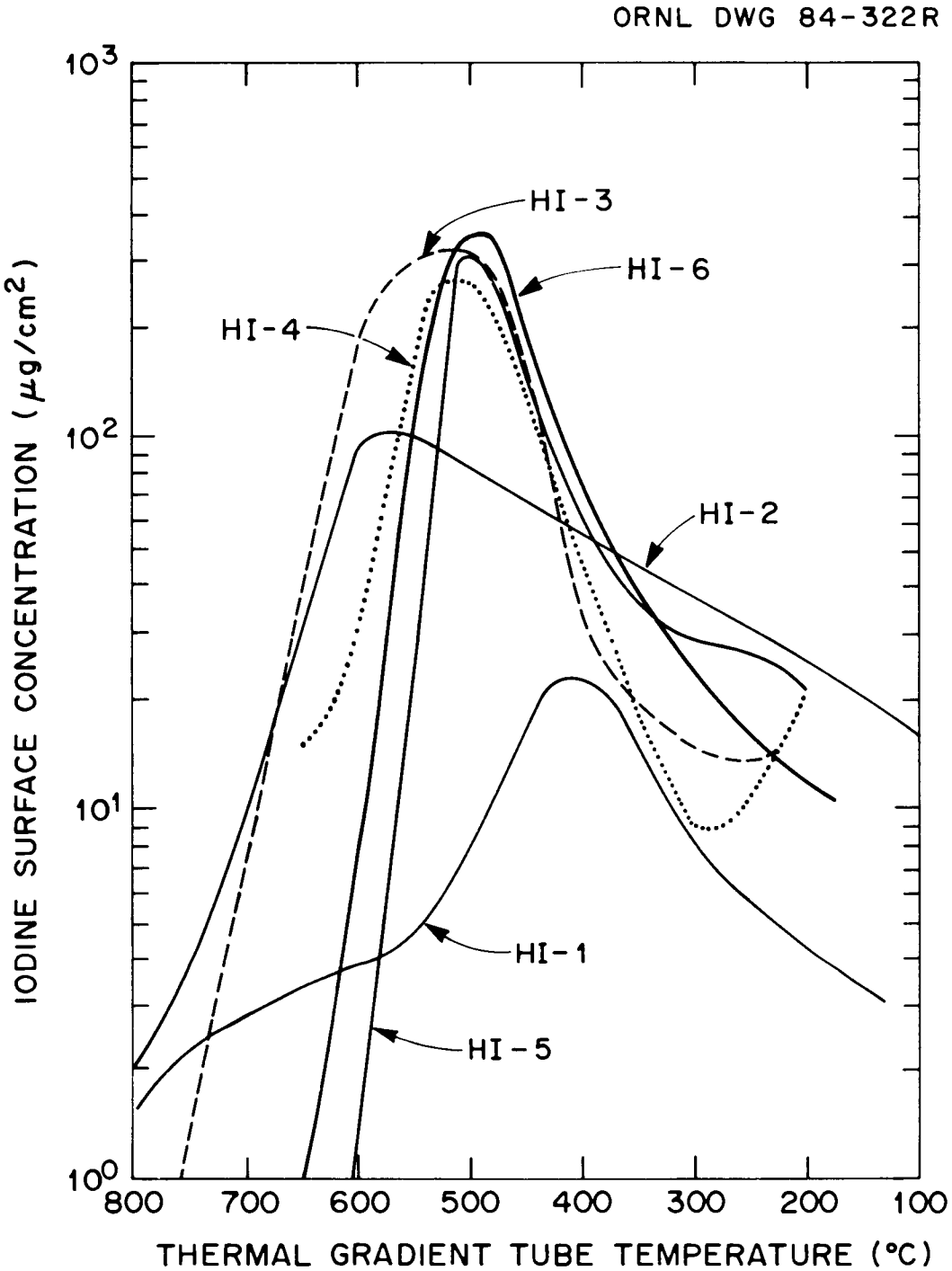

Fig. 13. Deposition behavior of iodine in the TGT liners used in tests HI-1 through HI-6.

water reactor accidents. It was made by adding boric acid and sodium hydroxide to ultra pure water to provide a solution which contained ~3750 ppm boron and ~1000 ppm sodium at a pH of 7.8.

Each section was leached in 200 mL (initially) of the leachant for 23.5 h at 53°C. One milliliter samples were analyzed periodically to follow the leaching rate. Each leach was stirred in the same manner.

Figure 14 shows the percentages of deposited ^{134}Cs removed from the sections with time. Cesium was more readily removed from section 3 than section 1. After 15 min leaching, 67% of the cesium had been removed from section 3 compared with 35% from section 2. After 23.5 h, 99.5% had been removed from section 3 and 90% from section 1. These data show that the reaction products of cesium and stainless steel components, when formed in steam at higher temperatures, are more difficult to remove with PWR cooling water at 53°C than those that formed at lower temperatures in steam. The reason for this may be that larger fractions of the reaction products may be more deeply embedded at higher temperature. Also, at the lower temperature, a portion of the cesium may be condensed rather than reacted with the stainless steel surface. If so, this may initially account for the faster leach rate for section 3. Species of cesium like CsI and CsOH are very soluble in water.

4.2.7 Results from Analysis of Aerosol Sampler

The aerosol sampler apparatus used in test HI-5 was also used in this test. It was positioned between the TGT and filter package. The sampler, described previously,[20] was a screw-driven graphite rod that collected the depositing aerosol along a spiral path as it was moved across a 1.3-mm gap in the line (see Fig. 15). After the test, the 0.8-mm-diam rod was analyzed by gamma spectrometry and also by scanning electron microscopy (SEM) with elemental identification by energy-dispersive x-ray analysis (EDX).

Relatively small amounts of the fission products cesium and iodine were collected on the sampler. Only 6.99 μg cesium was collected on the rod. The distribution of cesium along the sampler is shown in Fig. 16. The movement of the rod was started when the fuel temperature reached 1920°C. The deposit indicated at the start was an accumulation of cesium that occurred during heatup. The test was terminated 1 min later, and the cesium concentration declined rapidly thereafter.

Table 14 lists the elemental fractions for particles and general areas at three different locations on the graphite rod (also see Fig. 16). Cesium and iodine were present in each of the general areas. For the particles examined in the 0- and 1-mm locations, both cesium and iodine were present in all but particle No. 8. No cesium and iodine were found in the four particles picked at random at the 4-mm location; three of those particles contained high fractions of zirconium. Impurities S, Si, Sn, Cl, and Fe were found in many of the particles. The source of the impurities S, Si, and Cl, which have been frequently found by SSMS, appears to be the ZrO_2 ceramics in the furnace.

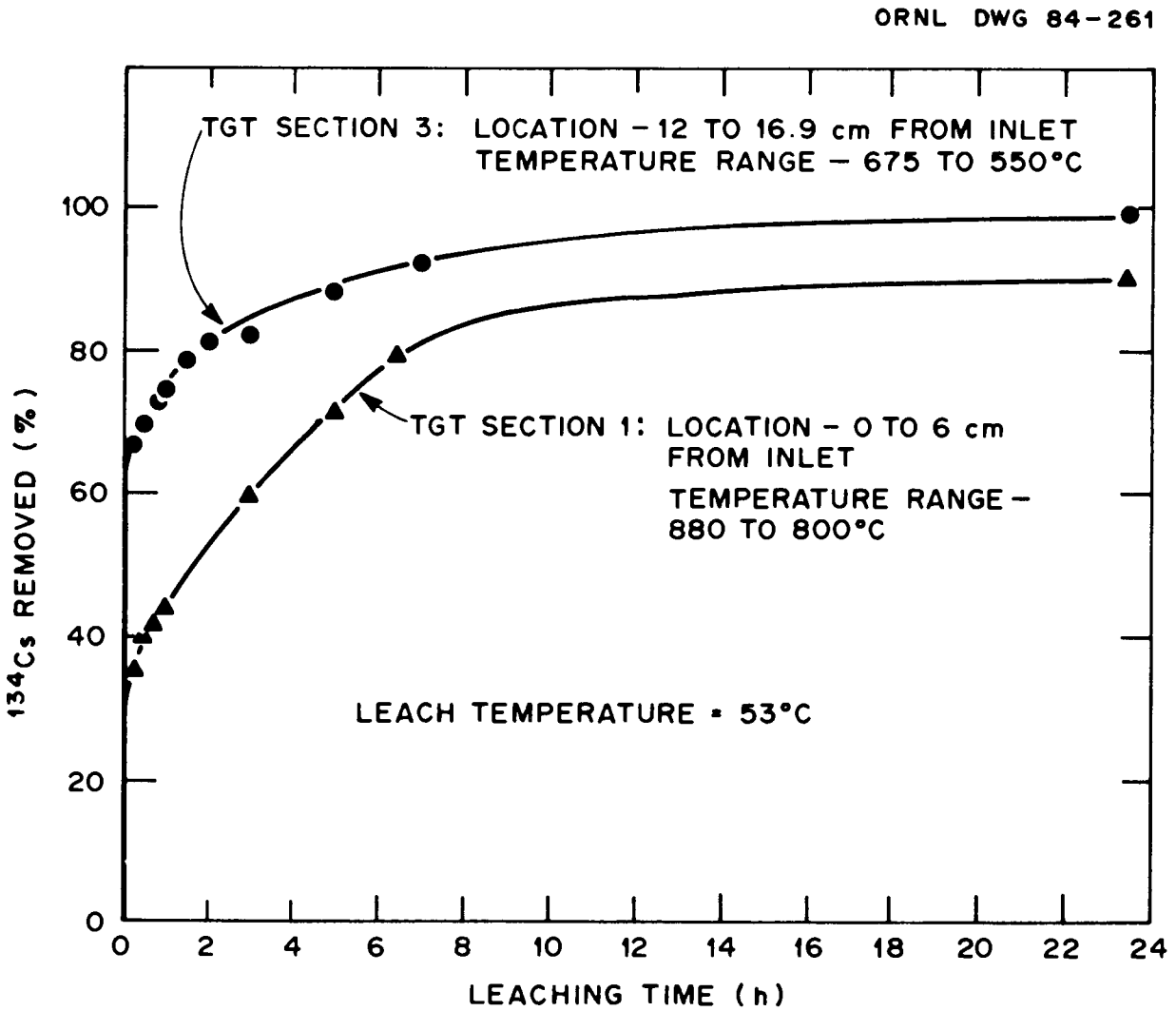

Fig. 14. Rate of removal of ^{134}Cs from stainless steel TGT sections by leaching with a simulant LWR coolant at 53°C.

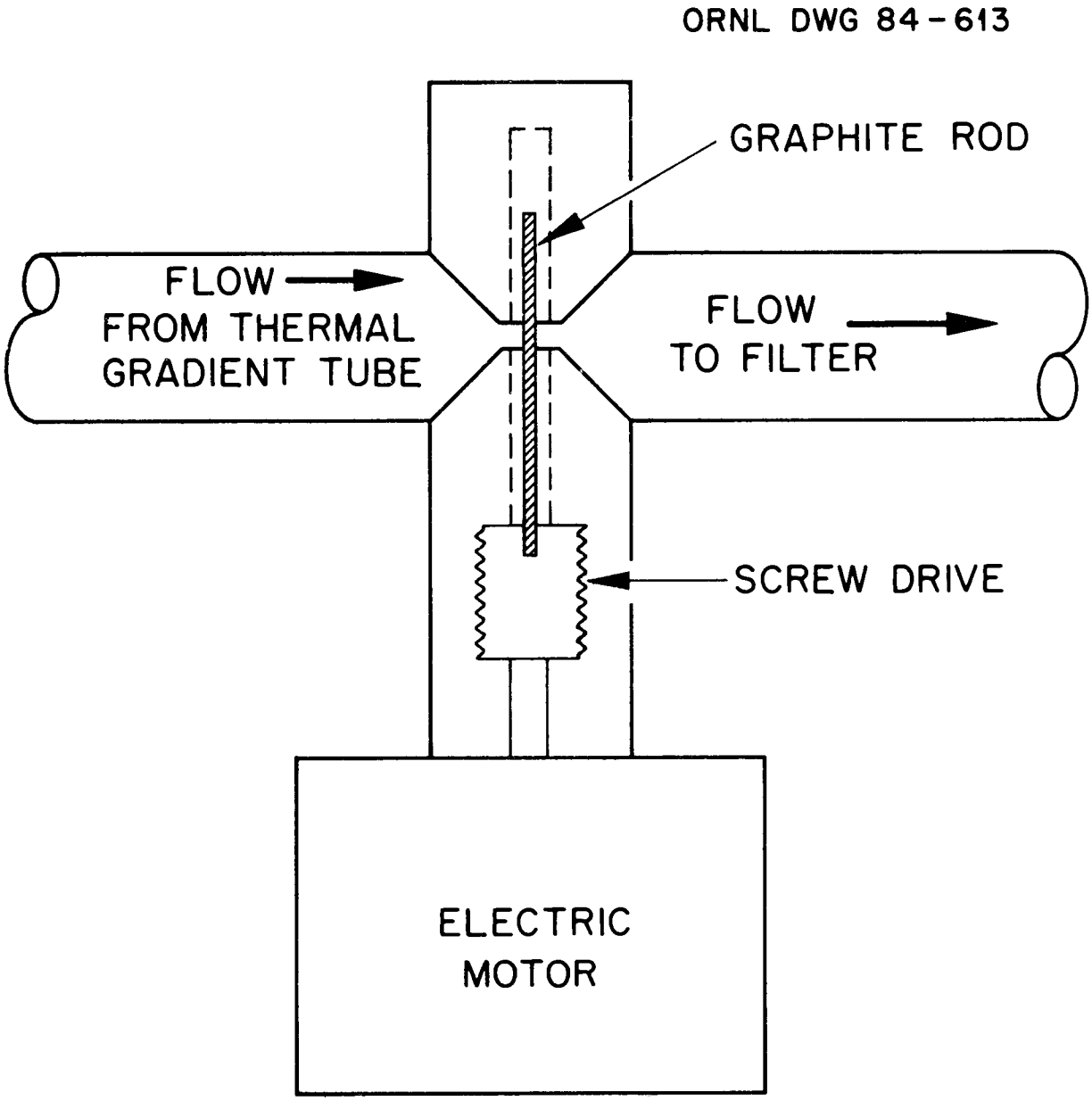

Fig. 15. Miniature aerosol deposition sampler.

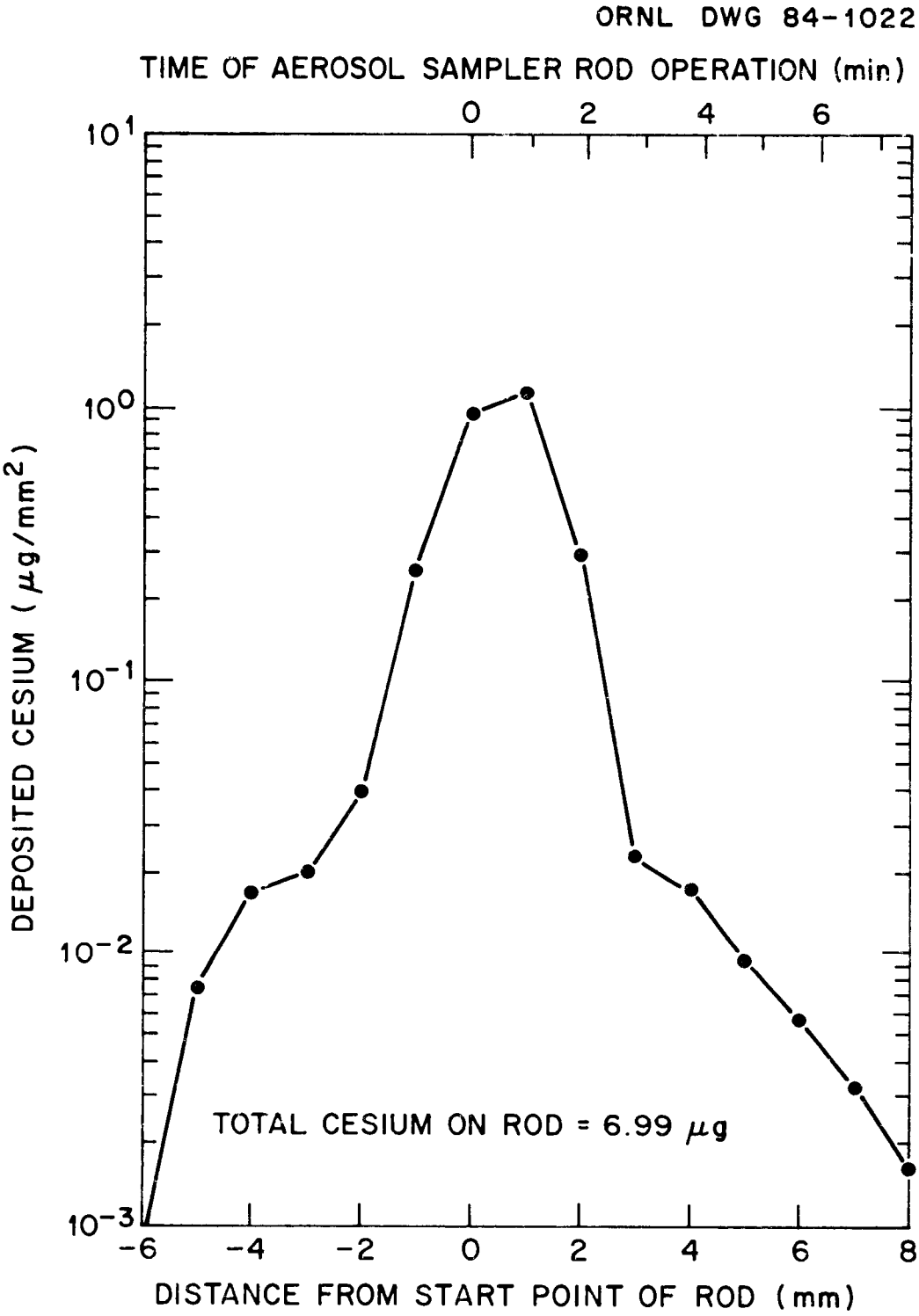

Fig. 16. Distribution of cesium along the graphite sampler.

Table 14. Aerosol sampler data[a] for test HI-6

Particle No.	Distance from start of movement[b] (mm)	Elemental fraction (at. %)							
		Cs	I	Zr	Sn	Si	S	Cl	Fe
1	0	39	14	0	11	14	24	0	0
2	0	45	15	0	7	8	25	0	0
3	0	34	18	0	10	13	25	0	0
4	0	50	15	0	9	5	21	0	0
5	1	37	12	0	9	11	32	0	0
6	1	33	8	0	6	16	37	0	0
7	1	26	9	0	0	21	21	23	0
8	1	14	0	0	0	23	19	43	0
9	4	0	0	84	0	0	0	0	16
10	4	0	0	0	0	33	0	46	21
11	4	0	0	88	0	0	0	0	12
12	4	0	0	53	0	17	0	29	0
General view									
A	0	38	15	0	11	14	24	0	0
B	1	39	12	0	9	12	28	0	0
C	4	31	10	0	0	13	24	22	0

[a]Obtained from SEM/EDX analysis.
[b]Sampler movement was started when the temperature of the fuel specimen reached 1920°C; it was continued for 15 min.

4.2.8 Gamma Spectrometric Analysis of the Fuel Specimen

In order to obtain information about the distribution of fission products as a function of distance along the fuel rod and to obtain an independent measurement of cesium release, direct gamma spectrometric analysis of the fuel specimen was utilized. Pretest measurements included analysis of the entire specimen through 3.81 and 6.35 cm (1.5 and 2.5 in.) of lead to determine the total content of high-energy gamma-emitting fission products (^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{154}Eu); in addition, unshielded measurements at three locations (midlength and near each end) through a 0.4-cm (0.16-in.) window provided a measure of the linear distribution. Similar posttest measurements were made of the entire specimen through 3.81- and 6.35-cm thicknesses of lead, and a scan using the 0.4-cm window with measurements at 1-cm intervals without lead shielding produced a detailed profile of fission product distribution.

These data were used to determine cesium release from the fuel by direct comparison of ^{134}Cs (1365 keV) and ^{154}Eu (1274 keV) values.

Europium-154 was chosen because no significant release of Eu has been observed in the HI test series and because it has a high-energy gamma ray similar to that of ^{134}Cs. Thermodynamic calculations suggest that europium is probably present in LWR fuel as Eu_2O_3.[21] Because of its low volatility, little migration of europium would have occurred during the test. It was necessary to compare the cesium isotope with the reference isotope because of pretest and posttest attenuation differences. After testing, the fuel specimen was placed in additional containment tubes and stabilized in epoxy resin.

Since the gamma-ray energies were relatively high and almost the same, a posttest attenuation correction factor for ^{134}Cs (1365 keV) was determined by dividing the pretest activity of ^{154}Eu (1274 keV) by the posttest value.

The following calculations show how the percentage of ^{134}Cs released was obtained:

- Attenuation correction factor (for epoxy, containment tubing, etc.)

$$F = \frac{\text{Pretest } ^{154}\text{Eu (1274 keV)}}{\text{Posttest } ^{154}\text{Eu (1274 keV)}} = \frac{0.4617 \text{ Ci}}{0.4267 \text{ Ci}} = 1.082 \ ;$$

- % ^{134}Cs released = % cesium released

$$= \frac{[(\text{Pretest } ^{134}\text{Cs}) - (\text{Posttest } ^{134}\text{Cs})\ (F)]}{(\text{Pretest } ^{134}\text{Cs})} \times 100$$

$$= \frac{[(7.885 \text{ Ci}) - (4.452 \text{ Ci})\ (1.082)]}{(7.885 \text{ Ci})} \times 100 = 38.9\% \ .$$

The percentage obtained by counting the individual apparatus components and relating them to the ORIGEN inventory was 33.1 (see Table 9). This has been the normal method of reporting release and is referred to here as method I.

Table 15 lists the percentages ^{134}Cs released as determined by both methods for tests HI-3, -4, -5, and HI-6. Considering the differences in the methods of mesurement, the values obtained by method II agreed fairly well with those obtained by method I.

Figure 17 gives the pretest and posttest longitudinal profiles of the ^{134}Cs (1365 keV)/^{154}Eu (1274 keV) ratios. As is shown, only three pretest measurements were taken — at the middle and near the ends. The ratio at each location was ~17.5, indicating a uniform burnup profile. The more detailed scan taken after the test showed that the ratio varied from ~9 at the inlet end to ~12 at the outlet end of the fuel specimen. This shows that the temperature at the inlet end was higher than it was at the outlet end during the test. Pretest measurements indicated that the gradient may have been ~75°C. The change in ratio shown in Fig. 17 suggests that the percentage cesium released per cm of fuel segment varied from ~48% at the inlet end to ~31% at the outlet end, with the average being ~39%.

Table 15. Comparison of percentage cesium released as determined by two methods

Test No.	Cesium released (%)	
	Method I[a]	Method II[b]
HI-1	1.75	c
HI-2	50.5	c
HI-3	58.8	60.2
HI-4	31.7	35.8
HI-5	20.3	24.5
HI-6	33.1	38.9

[a]Based on gamma counting of apparatus components and ORIGEN determined segment inventory.

[b]Determined by counting fuel rod segment before and after test.

[c]Fuel rod segment was not counted.

4.2.9 Fuel Examination

Because of the reduction in steam flow during the test and the short time at high temperature, the Zircaloy cladding was not completely oxidized. After being cast in epoxy resin, the fuel specimen was cut at ~2-cm intervals; these radial sections are shown in Fig. 18. Severe oxidation is apparent at the inlet end of the specimen (section 1), and cladding melting and/or fracturing is visible in all views except that near the outlet end (section 7). A large cladding fracture appears to extend over more than half the length of the specimen.

Detailed examination of the fuel and cladding microstructure is being conducted at Argonne National Laboratory and will be reported later.

5. COMPARISON OF RELEASE DATA FROM THE HI AND HT TEST SERIES

Two series of tests (HI and HT) have been conducted in the Fission Product Release Program at ORNL where highly irradiated LWR fuel segments have been tested in steam in the temperature range 1300 to 2000°C to determine fission product release under simulated severe accident conditions. In this section, the differences in test parameters and release rate coefficient results for the two test series are discussed; also, preliminary explanations for the differences are given. A more thorough examination and comparison of the results will be provided in the HI test series topical report which is to be published soon.

The importance of the role of an expanded versus a nonexpanded pellet-to-cladding gap on fission product release was examined in these

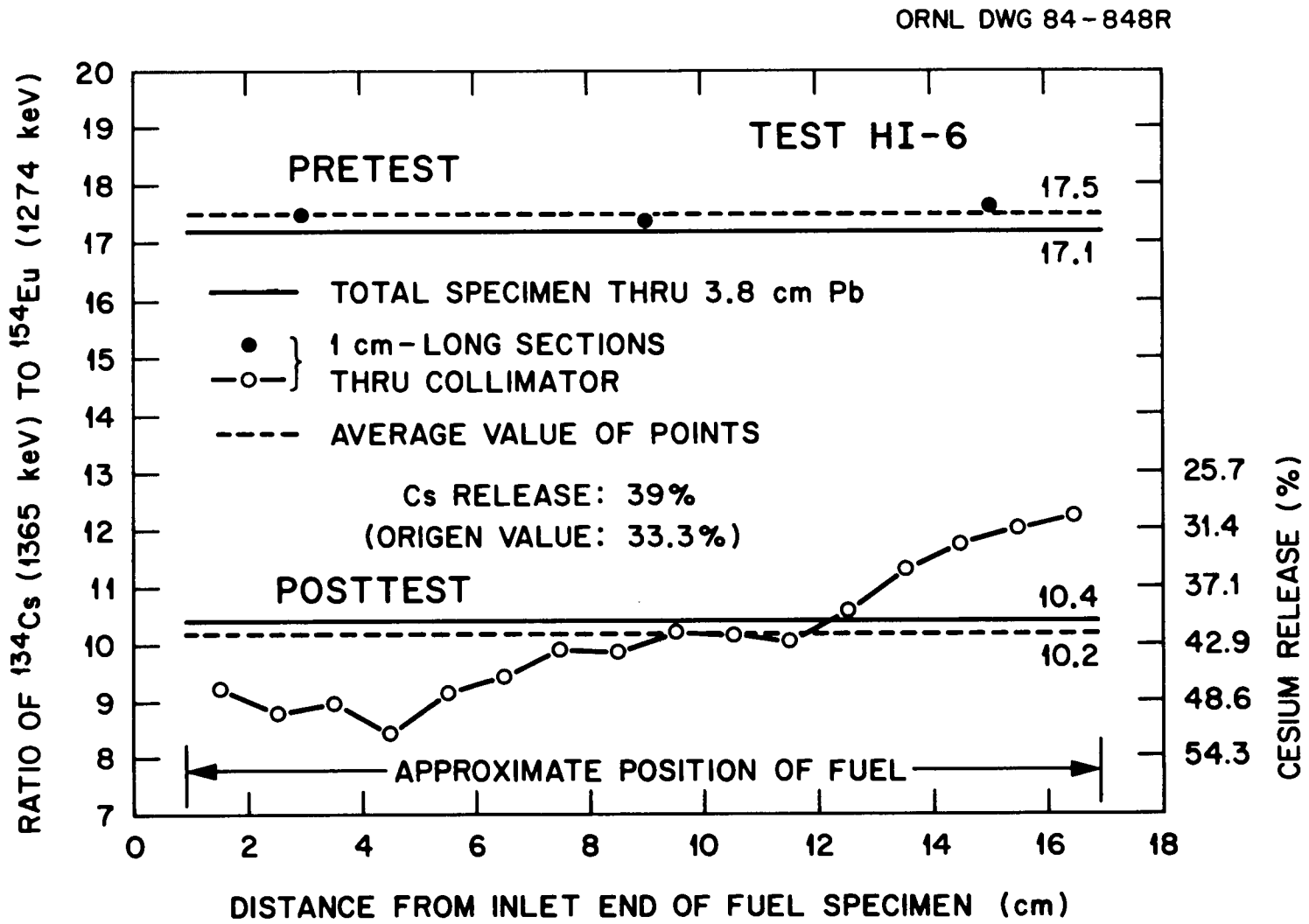

Fig. 17. Comparison of pretest and posttest ratios of ^{134}Cs (1365 keV) to ^{154}Eu (1244 keV) along the fuel rod segment in test HI-6.

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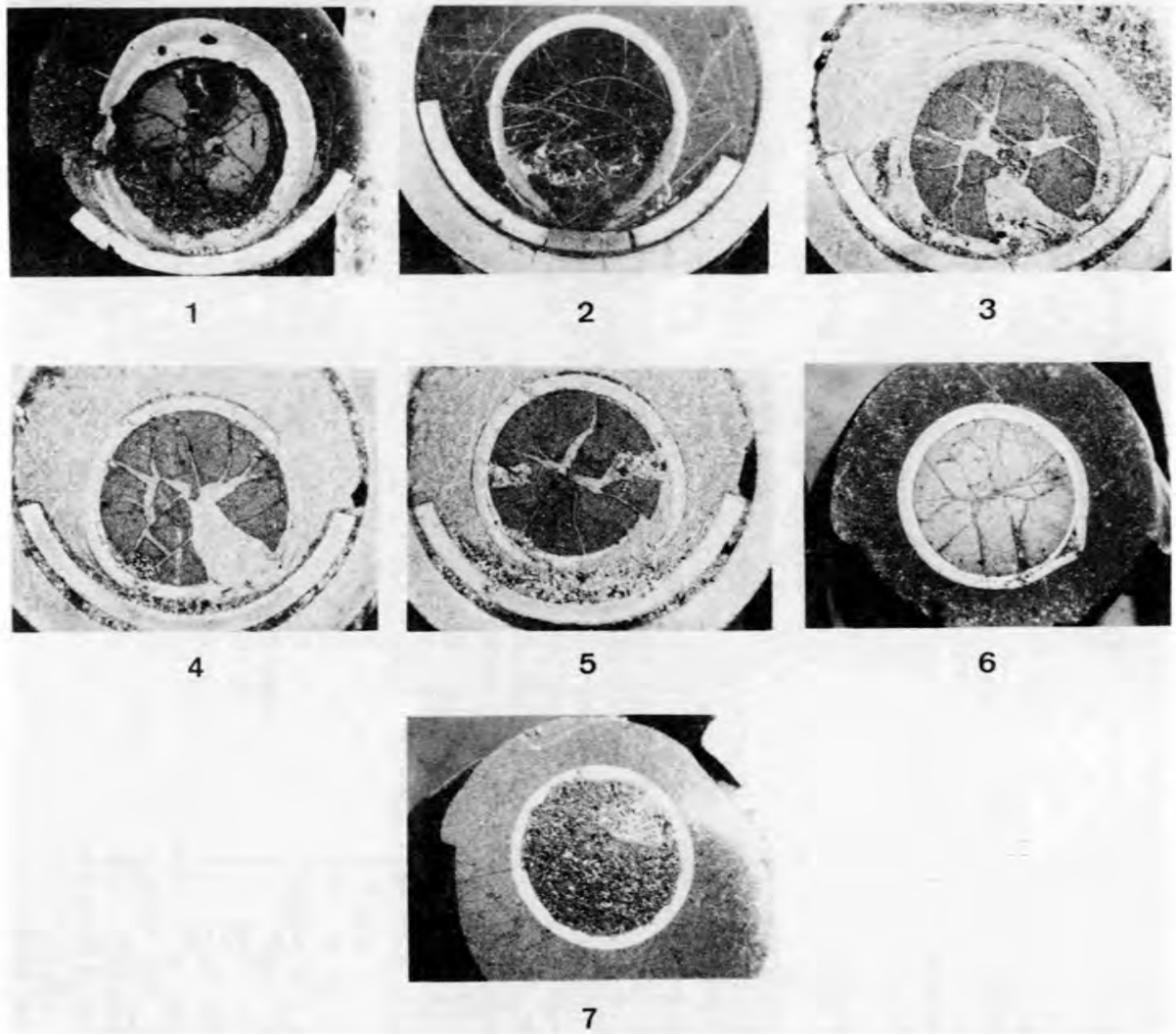

Fig. 18. Radial sections through the fuel specimen after test HI-6. The cuts were made at ~2-cm intervals, with section 1 near the inlet end and section 7 near the outlet end.

two series of tests. This is important because the cladding of individual fuel rods would expand and rupture during the initial heatup period of most severe LWR accidents. The gap width would be the widest at and near the rupture location; along much of the 3.66-m-long rod, the gap width would be little changed from the preaccident state. The Zircaloy cladding of the fuel specimens used in the HT test series was expanded during a separate operation to enlarge the existing gap width to 0.38 mm prior to testing.[4] This was not done in the HI test series. In both test series, a simulated defect was provided by drilling a single 0.159-cm-diam hole through the cladding at the center of each test specimen.

The fuel rod specimens in both test series were heated by induction heating, but the methods of coupling were different. In the HT tests, the Zircaloy cladding of the test specimen was coupled. In the HI tests, either a tungsten or graphite susceptor tube was coupled (see Fig. 3). The latter method enabled the fuel specimen to be heated longer and more isothermally.

The operating conditions for the HI and HT tests are listed in Table 16. The temperatures for several of the HT tests have been changed from those in previous publications as the result of a careful review of all test data and temperature calibration runs. The heatup and cooldown rates of the test specimen for the HT tests were considerably faster than those for the HI tests. On the other hand, with the exception of test HI-6, the fuel specimens in the HI tests were heated at the test temperatures for longer periods of time. The steam–helium flow rate was high in each of the HT tests and three of the HI tests. By using the dependency of temperature on cesium release rates as given in the NUREG-0772 report,[22] the equivalent amount of high-temperature release time was determined for the linear heatup and cooldown periods.

Additional information and fission product release results are listed in Table 17. All the HT tests as well as the first three HI tests were conducted with fuel segments from the same fuel rod that was irradiated in the H. B. Robinson-2 PWR. Tests HI-4 and -6 were conducted with BWR fuel; the segments came from the Peach Bottom-2 and Monticello reactors, respectively. The HI-5 specimen was irradiated in the Oconee unit 1 reactor (PWR). The burnup range for the specimens was between 10 and 40 MWd/kg. The average linear heat rating was from a low of ~175 W/cm for the H. B. Robinson fuel to a high of ~332 W/cm for the Peach Bottom-2 fuel. The large variance in linear heating rating is reflected in the "in reactor" release values for ^{85}Kr, 0.3% for H. B. Robinson and 10.2% for Peach Bottom-2 (~7% rod average).

In order to minimize the effect of different heating times, average release rates were calculated for each test and nuclide using the method in NUREG-0772. The release rate values are given in Table 18 and plotted in Fig. 19. The release rate coefficient curves for Kr, Cs, and I in NUREG-0772 are also shown for comparison. In this calculational comparison, no compensation was made for burnup, linear heat rating, fuel structure before heating, change in release rate with time, and possible fuel fragmentation resulting from rapid heatup and cooldown.

Table 16. Operating conditions for ORNL fission product release tests

Test No.	Temperature (°C)	Heatup rate (°C/s)	Cooldown rate (°C/s)	Time at temperature (min)	Heatup/ cooldown allowance[a] (min)	Steam flow (L/min STP)	Inert gas flow (L/min STP)
HT-1	1325	9.9	12.2	10	0.25	0.72	0.87 (He)
HT-2	1445	11.1	11.1	7	0.25	0.94	0.87 (He)
HT-3	1610	6.7	17.0	3	0.38	1.02	0.87 (He)
HT-4	1400	18.5	11.0	0.33	0.20	0.93	0.79 (He)
HI-1	1375[b]	0.97	0.60	30	3.8	1.00	0.50 (Ar)
HI-2	1700	1.04	1.17	20	2.5	0.99	0.33 (Ar)
HI-3	1975[b]	2.78	1.67	20	1.3	0.30	0.30 (He)
HI-4	1900[b]	1.93	1.58	20	1.6	0.32	0.30 (He)
HI-5	1725[b]	1.00	1.33	20	2.5	0.39	0.40 (He)
HI-6	1950[b]	1.77	2.00	1	1.5	~1.00	0.42 (He)

[a]Equivalent to additional time within 85°C of test temperature.

[b]Revised from previously published temperatures.

Table 17. Data summary for fission product release tests

Test No.	Fuel type	Burnup (MWd/kg U)	Temperature (°C)	Time[a] (min)	Amount released (% of original fuel rod inventory) ^{85}Kr In reactor[b]	^{85}Kr In test	^{137}Cs	^{129}I
HT-1	PWR	28	1325	10.3	0.3	2.1[c]	0.22[c]	0.33[c]
HT-2	PWR	28	1445	7.3	0.3	9.6[c]	9.6[c]	4.7[c]
HT-3	PWR	28	1610	3.4	0.3	16.5[c]	21.9[c]	25.2[c]
HT-4	PWR	28	1400	0.5	0.3	5.6[c]	6.1[c]	3.5[c]
HI-1	PWR	28	1375	33.8	0.3	2.83	1.75	2.04
HI-2	PWR	28	1700	22.5	0.3	51.5	50.5	53.0
HI-3	PWR	25	1975	21.3	0.3	59.0	58.8	35.4
HI-4	BWR	10	1900	21.6	10.2	21.1	31.7	24.7
HI-5	PWR	39	1725	21.5	3.5	16.5	20.3	22.4
HI-6	BWR	40	1950	2.5	2.0	29.6	33.1	24.7

[a]Time includes heatup/cooldown allowance from Table 16.

[b]Measured by rod puncture corrected for individual test segment burnup.

[c]Estimated release from fully heated portion of test segment.

Table 18. Release rate coefficients for fission product release tests

Test No.	Temperature (°C)	Release rate coefficients (fraction/min)		
		^{85}Kr[a]	^{137}Cs	^{129}I
HT-1	1325	2.06×10^{-3}	2.13×10^{-4}	3.21×10^{-4}
HT-2	1445	1.38×10^{-2}	1.38×10^{-2}	6.59×10^{-3}
HT-3	1610	5.30×10^{-2}	7.27×10^{-2}	7.51×10^{-2}
HT-4	1400	1.15×10^{-1}	1.26×10^{-1}	7.13×10^{-2}
HI-1	1375[b]	8.49×10^{-4}	5.22×10^{-4}	6.10×10^{-4}
HI-2	1700	3.22×10^{-2}	3.13×10^{-2}	3.36×10^{-2}
HI-3	1975[b]	4.19×10^{-2}	4.16×10^{-2}	2.05×10^{-2}
HI-4	1900[b]	1.24×10^{-2}	1.77×10^{-2}	1.31×10^{-2}
HI-5	1725[b]	8.72×10^{-3}	8.61×10^{-2}	9.57×10^{-2}
HI-6	1950[b]	1.30×10^{-1}	1.62×10^{-1}	1.13×10^{-2}

[a]Calculated based on the amount of ^{85}Kr in the fuel at the start of the heatup.

[b]Revised from previously published temperatures.

The release rate coefficients obtained in the HI tests for fission products Kr, Cs, and I (Fig. 19) were a factor of 3.2 lower than the values indicated by the NUREG-0772 curves and a factor of 19 lower then the values (dashed line) obtained in the HT tests. Obviously, the dashed line plots through the HT and HI data points, which are shown parallel to the NUREG-0772 curves, were drawn with some judgment, with the HI-2 and HI-6 values discounted for the following reasons. Those values were significantly higher and were similar to the NUREG-0772 values. In both those HI tests, long axial cracks occurred during the tests making it easier for the fission products to escape. Furthermore, it is possible that the exposed surfaces of the fuel pellets were partially oxidized by the steam causing enhanced fission product release. Because of an extremely short test time, the release rate coefficient values for Cs, I, and Kr for test HT-4 were also ignored in plotting the dashed line curve for the HT tests.

We believe the large difference in release rate coefficients for the two test series was caused by two factors: (1) the heatup and cooldown rates of the fuel specimens in the HT tests were considerably faster than in the HI tests (a factor of 6 for heatup and a factor of 9 for cooldown), and (2) the cladding of the fuel specimens in the HT tests was pretest expanded to provide a wider pellet-to-cladding gap (0.38 mm).

Stresses in the fuel set up by the more rapid heatup and cooldown rates in the HT tests and the lack of cladding restraint probably resulted in a larger network of cracks in the fuel than occurred in the HI tests. These cracks provide pathways for the volatile fission products

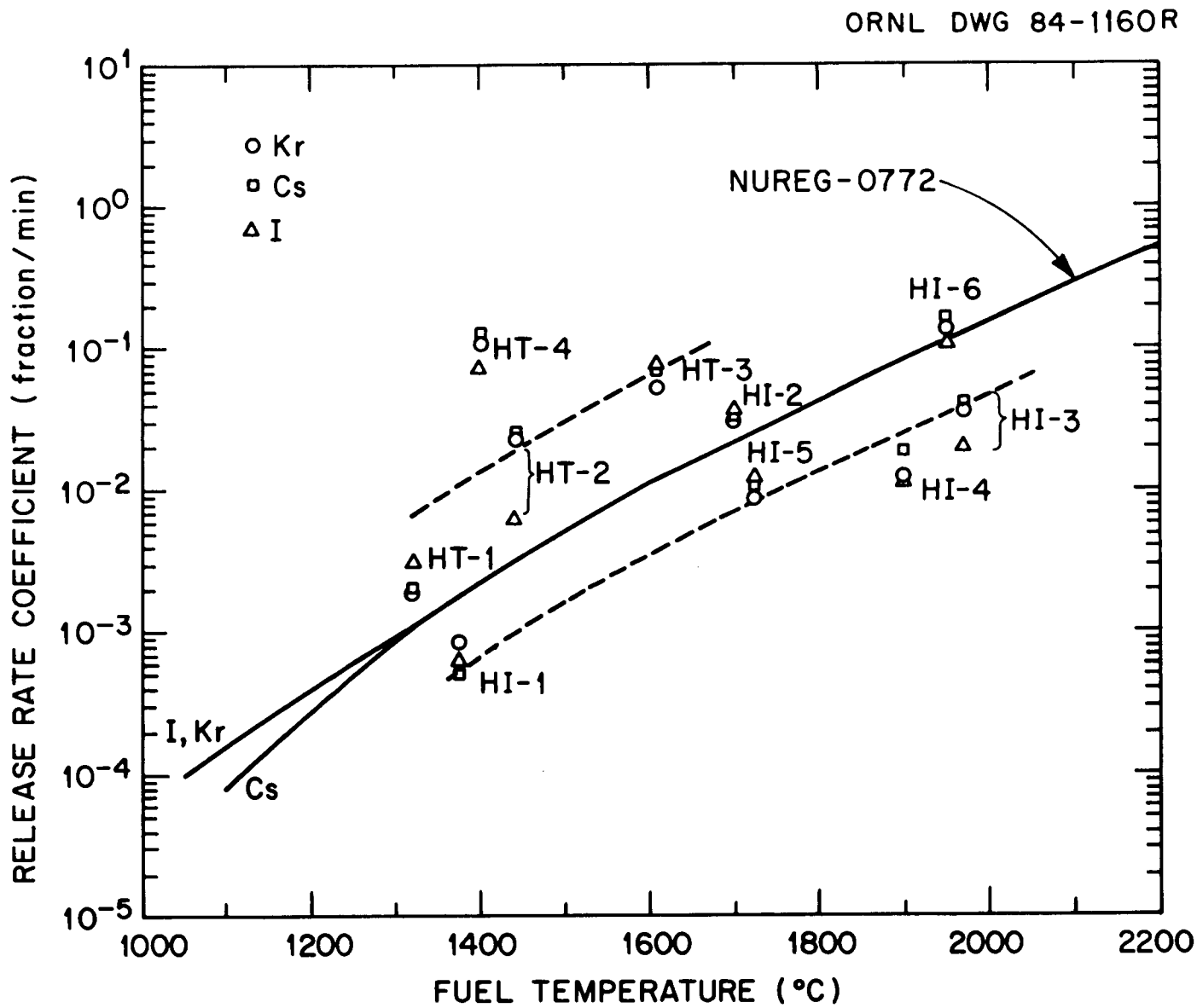

Fig. 19. Comparison of release rate coefficient data from HI and HT tests with curves from NUREG-0772.

in the grain boundaries to escape. (Grain boundaries act as storage areas for volatile fission products during irradiation.[23]) Also, the enlarged pellet-to-cladding gap made it easier for the freed fission products to escape.

At temperatures >1400°C, UO_2 transforms from being a brittle or semibrittle material to being ductile (plastic) and rapidly loses its strength with increased temperature.[21] With decreased strength, there is a weakening of the grain boundaries. Expansion pressure of the volatile fission products in bubbles along the grain boundaries causes cracks to develop. Also, the gas bubbles can coalesce and tunnel along the weakened boundaries to find escape avenues.

In the HT tests, there was considerable difference in the releases in tests conducted at 1300 and 1400°C; there appeared to be a "breakaway" release phenomenon. The weakening of the fuel at 1400°C most likely played a role in that behavior leading to the more significant releases.

6. CONCLUSIONS

This report presents only a limited evaluation and interpretation of the data obtained in test HI-6. The following observations and conclusions should be considered of a preliminary nature:

1. This test concluded the HI test series in which PWR and BWR fuel specimens were tested in a horizontal position. The specimen was heated to 1950°C for ~1 min in an atmosphere of flowing steam (~1.0 L/min) and helium (0.42 L/min). For fission product release calculations in which release during heatup and cooldown is included, it is recommended that a total effective release time of 2.5 min at 1950°C be used.

2. As in previous tests, the total release fractions for Kr, Cs, and I were similar (29.6, 33.1, and 24.7%, respectively), suggesting similar release mechanisms under these conditions. A small difference in time-of-release of Kr and Cs was observed. Similar release fractions for rubidium and bromine are estimated. Rubidium was detected by SSMS and bromine by neutron activation.

3. Significant release fractions for ^{110m}Ag (6.0%) and ^{125}Sb (0.06%) were detected by gamma spectrometry. Most (99.8%) of the released ^{110m}Ag was deposited on the ZrO_2 ceramics at the outlet end of the furnace. About 67% of the detected ^{125}Sb was also found at the inlet end of the ZrO_2 furnace tube liner; the remainder was found on the first 1-cm section at the entrance to the TGT.

4. Pretest and posttest gamma spectrometric analysis of the fuel specimen indicated that 38.9% of the ^{134}Cs was released, which is in fair agreement with the value (33.1%) obtained by comparing the "summation of components" value with the ORIGEN-calculated inventory.

5. The deposition profile for cesium in the stainless steel TGT suggested that a large portion of the cesium reacted with the stainless steel surface but only at the inlet end where the temperature was >600°C during the test and where surface oxidation occurred.

6. The deposition profile for iodine suggested that it deposited as cesium iodide.

7. In a leach study with a simulated LWR coolant solution at 53°C, cesium was found to be significantly more difficult to remove from a stainless steel TGT section where deposition occurred at ~800°C than from one where deposition occurred at ~600°C. (The leach solution contained ~3750 ppm boron and ~1000 ppm sodium with a pH of 7.8.)

8. Because of the short test time at high temperature and because of the reduction in steam flow during the test, the Zircaloy cladding was not completely oxidized. Radial sections of the test specimen which was cast in epoxy resin showed severe oxidation at the inlet end, evidence of melting and/or fracturing in all views examined except near the outlet end, and a large cladding fracture that extended over more than half the length of the specimen.

9. A comparison was made of Cs, I, and Kr release rate coefficients obtained in the HI and HT test series with NUREG-0772 values. The coefficients obtained in the HI tests were factors of 19 and 3.2 times lower than the HT tests and NUREG-0772 values, respectively.

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13. ABSTRACT (200 words or less)

The sixth in a series of high-temperature fission product release tests was conducted for 1 min at 1950°C in a steam–helium atmosphere. The 15.2-cm-long test specimen was a section of fuel rod which was irradiated to 40.3 MWd/kg in the Monticello BWR.

Posttest analyses showed total releases of 29.6% for ^{85}Kr (includes 2% that was released to the plenum during irradiation), 33.1% for ^{137}Cs, 24.7% for ^{129}I, 6.0% for ^{110m}Ag, and 0.06% for ^{125}Sb.

A stainless steel thermal gradient tube was used to examine the retention of fission product cesium by stainless steel. Gamma scans showed that a significant fraction of the released cesium was retained at temperatures >600°C where the surface had been oxidized. Cesium that was released as CsI appeared unaffected by its contact with the stainless steel.

A comparison was made of Cs, I, and Kr release rate coefficients obtained in the HI and HT test series with NUREG-0772 values.

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