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**MARTIN MARIETTA**

**Data Summary Report for Fission  
Product Release Test HI-5**

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

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MARTIN MARIETTA ENERGY SYSTEMS, INC.  
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DEPARTMENT OF ENERGY

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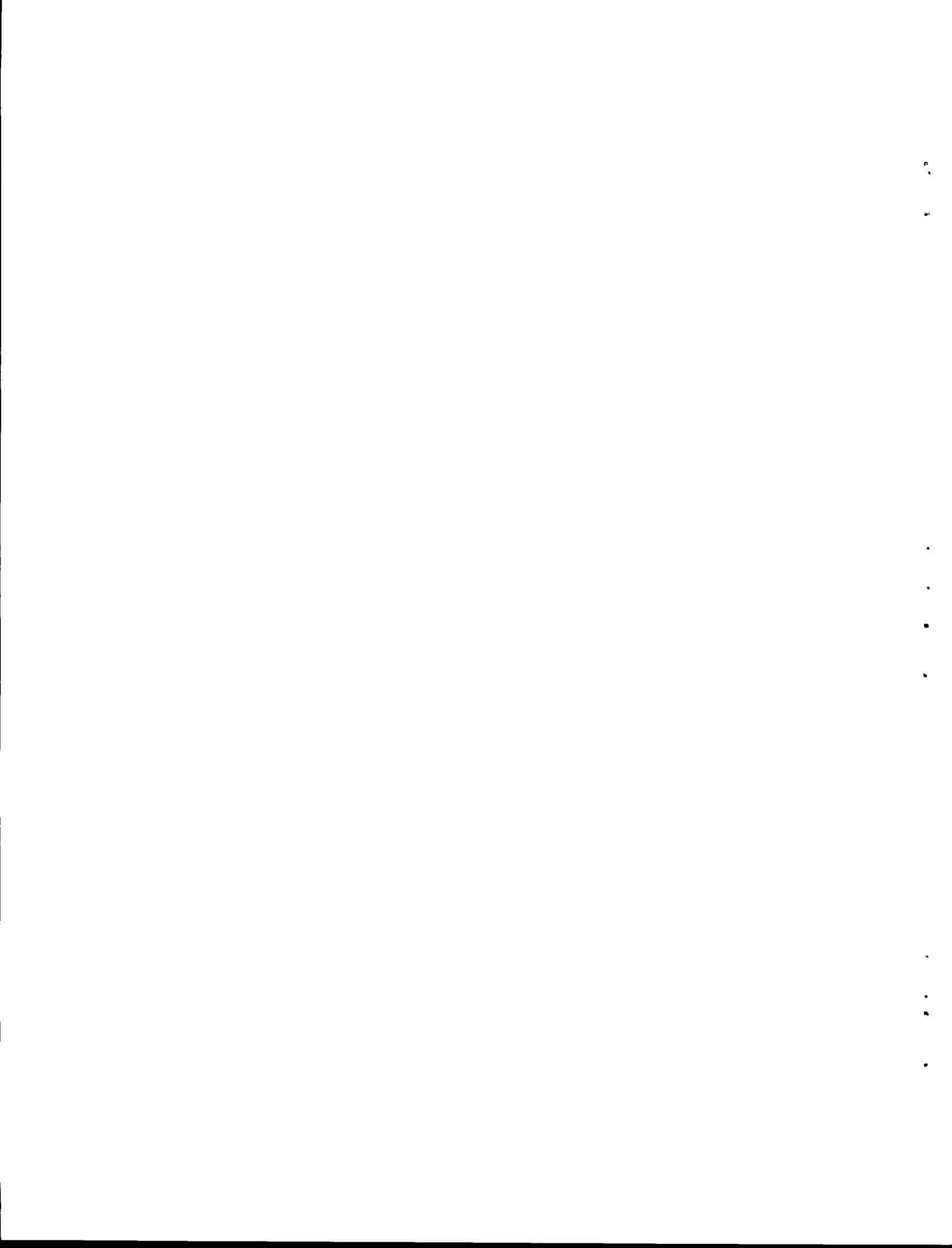
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OAK RIDGE NATIONAL LABORATORY  
Oak Ridge, Tennessee 37831  
operated by  
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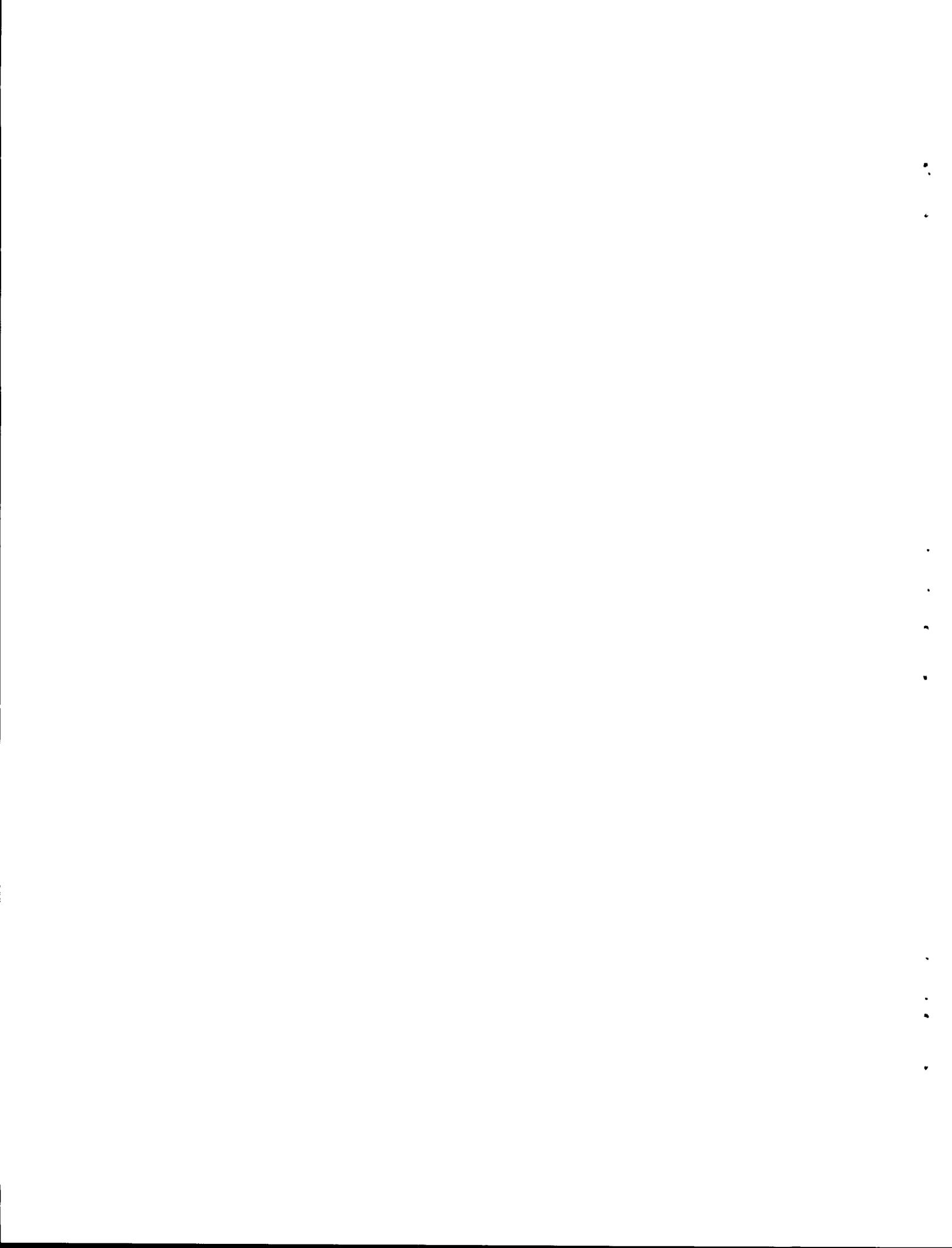


## ABSTRACT

The fifth in a series of high-temperature fission product release tests was conducted for 20 min at 1700°C in flowing steam. The test specimen was a 15.2-cm-long section of a fuel rod from the Oconee 1 PWR which had been irradiated to a burnup of ~38.3 MWd/kg. Using an induction furnace, it was heated under simulated LWR accident conditions in a hot cell.

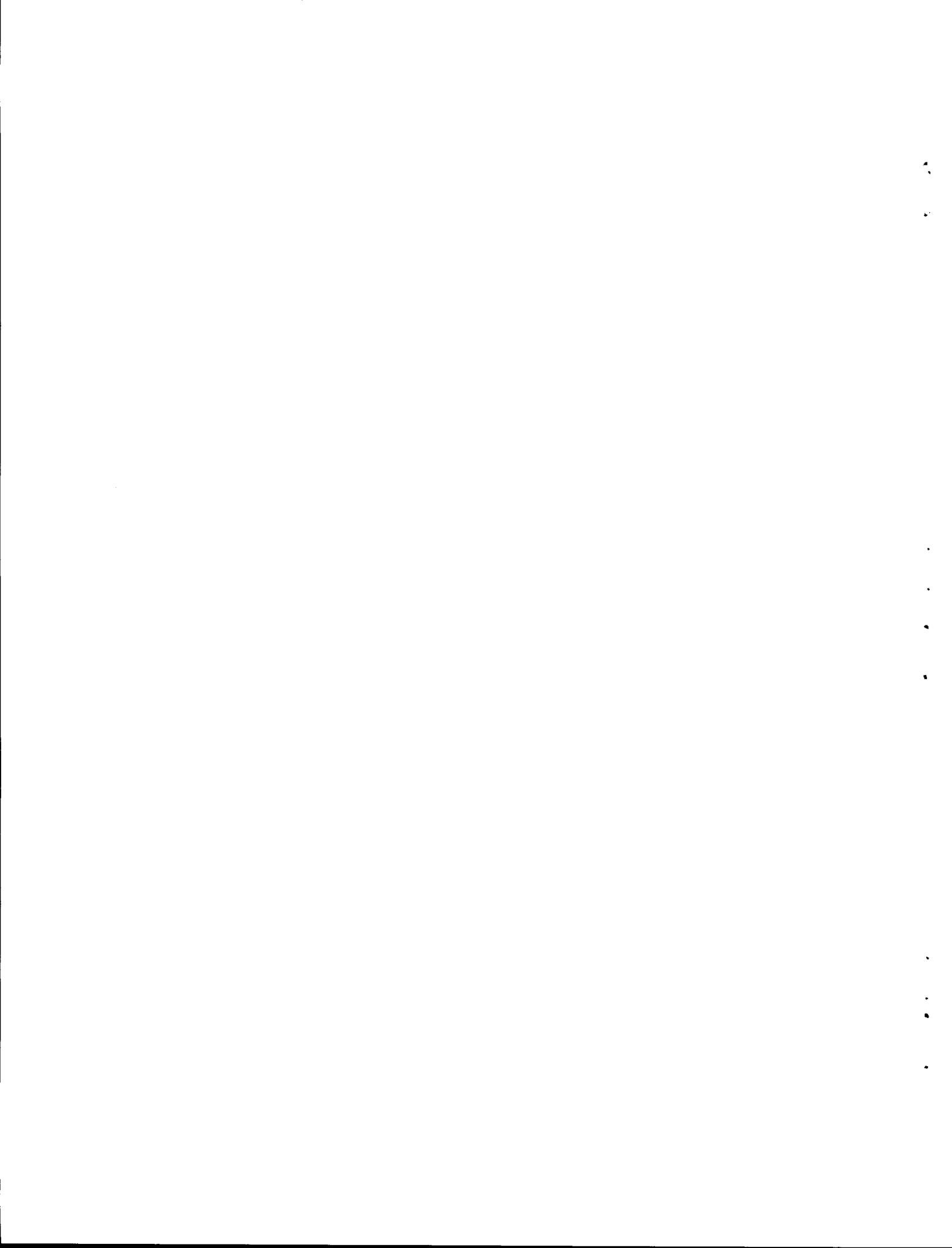
Posttest inspection showed severe oxidation and fragmentation of a portion of the fuel specimen, but no cladding melting was apparent. Based on fission product inventories calculated by ORIGEN, analyses of test components showed total releases from the fuel of 19.9% for  $^{85}\text{Kr}$ , 22.4% for  $^{129}\text{I}$ , 18.0% for  $^{110\text{m}}\text{Ag}$ , and 20.3% for  $^{137}\text{Cs}$ . A smaller fraction of the  $^{125}\text{Sb}$  (0.326%) was released from the fuel, and ~99% of the  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$  was retained in the furnace. Posttest analysis of the fuel specimen indicated a  $^{134}\text{Cs}$  release of 24.5%, which is reasonably good agreement.

This test was conducted at the same temperature as test HI-2 but with the steam flow reduced in order to determine the effect of cladding and fuel oxidation on fission product release. The release of Cs, I, and Kr was less than half that in HI-2, but it is not certain that the degree of oxidation alone was responsible for the difference. The large axial crack that occurred in HI-2 probably was a significant factor.



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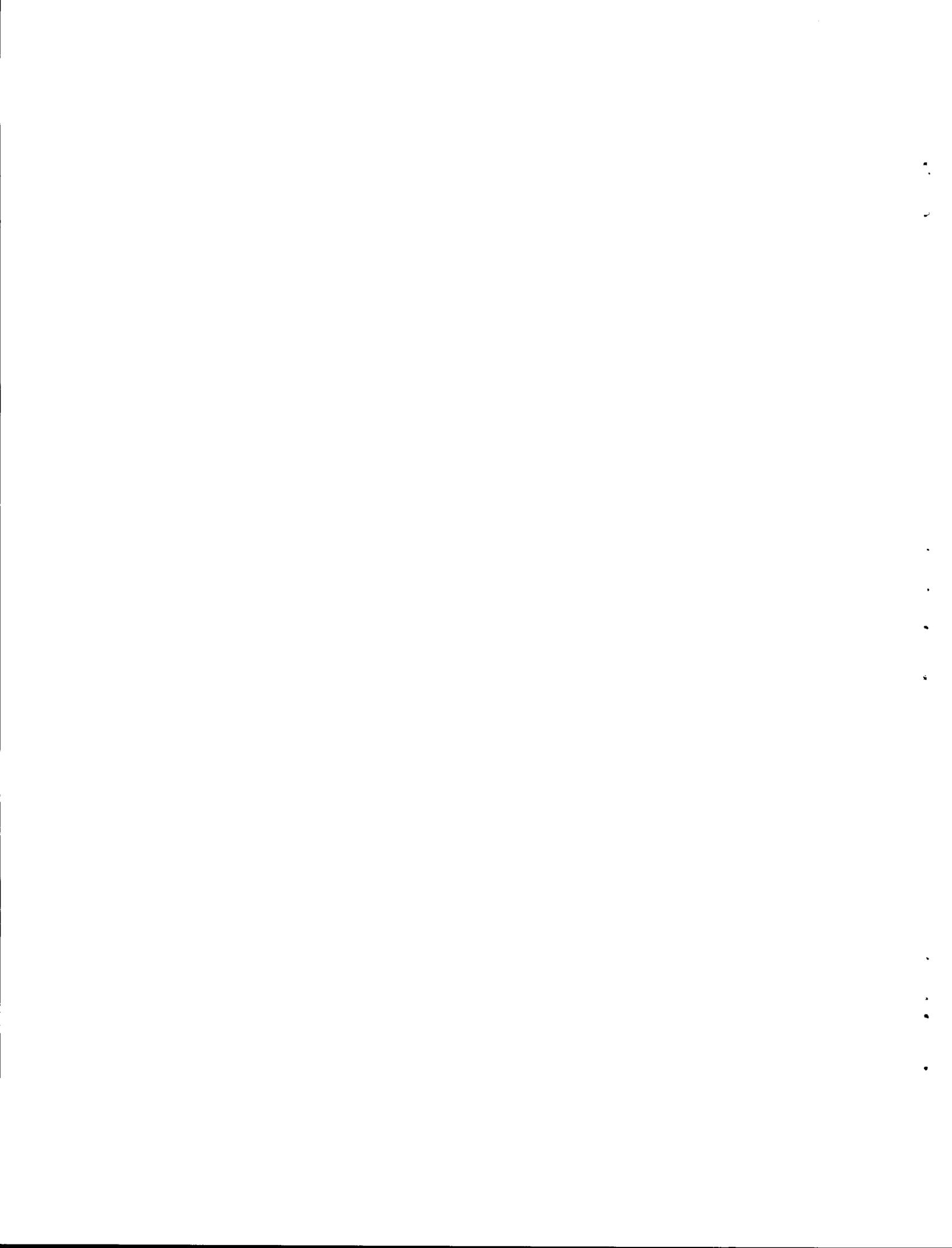
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## FOREWORD

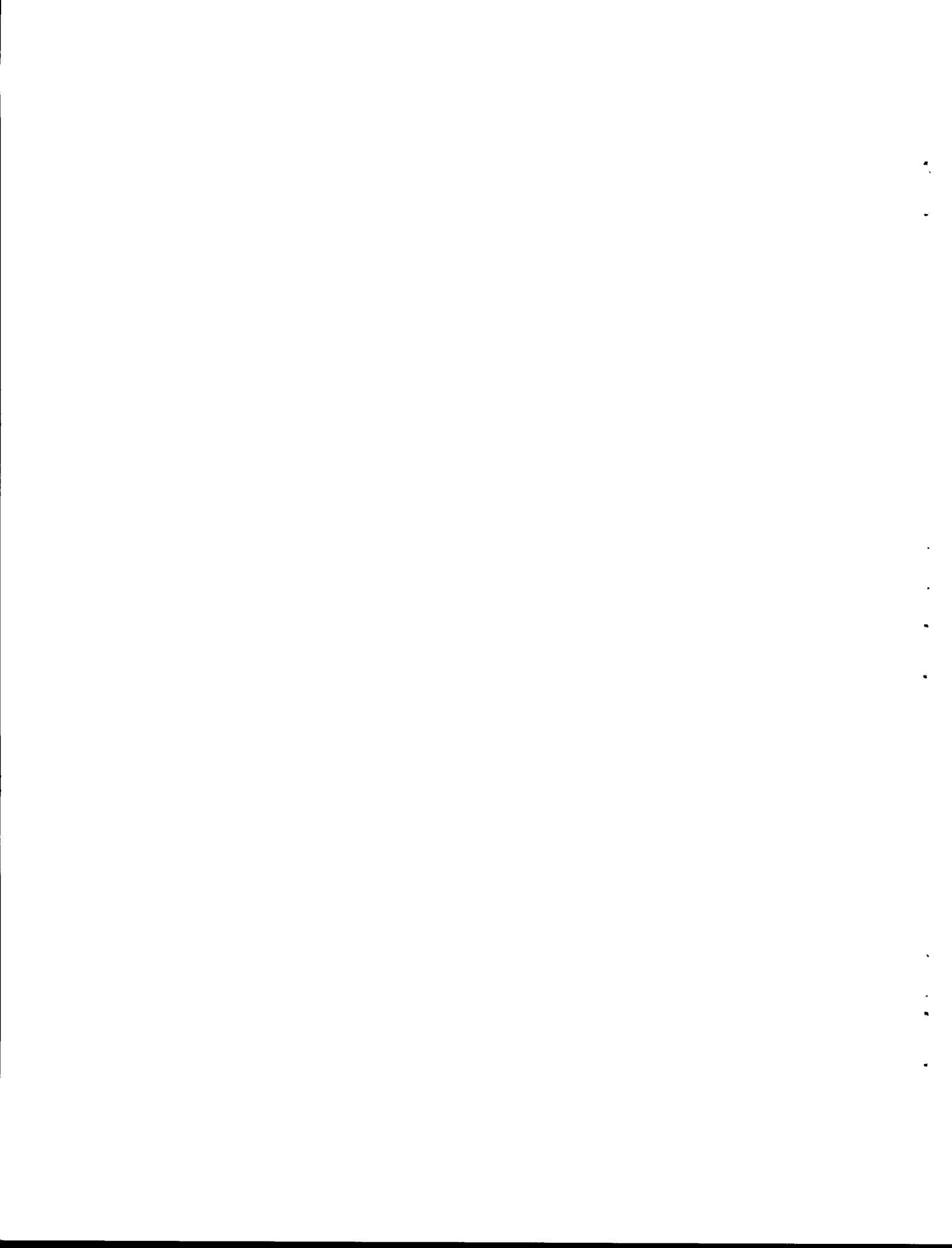
This document is the fifth in a series of reports describing the conduct and results of fission product release testing of commercial LWR fuel. The other reports describing work under this fission product release program are:

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, to be published.



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DATA SUMMARY REPORT FOR FISSION PRODUCT RELEASE FROM TEST HI-5

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

1. EXECUTIVE SUMMARY

The objective of this report is to present a thorough documentation of the observations and results collected for this fission product release test (HI-5). 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 four previous tests in this series and for 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 Oconee 1 PWR from February 1975 to November 1979. 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 posttest analysis procedures are described.

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

1. Total release from the fuel values for  $^{85}\text{Kr}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{129}\text{I}$ , and  $^{137}\text{Cs}$  were 19.9, 18.0, and 22.4, and 20.3%, respectively. In addition, 0.33% of the  $^{125}\text{Sb}$  was released.
2. Of the above released nuclides, all of the  $^{85}\text{Kr}$  and  $^{129}\text{I}$  and about two-thirds of the  $^{137}\text{Cs}$  escaped from the furnace and were found in the collection system. On the other hand, ~99% of the released  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$  were retained in the  $\text{ZrO}_2$ -lined furnace, indicating large differences in mobility and/or reactivity between these elements.
3. The observed behavior of Cs, I, Sb, and Ag in the thermal gradient tube was consistent with previous tests; the apparent chemical species were CsI, CsOH, Sb, Ag, and at least one additional Cs-containing compound.
4. Gamma spectrometric analysis of the fuel specimen before and after the test indicated that 24.5% of the  $^{134}\text{Cs}$  was released. This independent measurement compares reasonably well with the corresponding values based on ORIGEN inventories and the recovery of the volatile elements.
5. Samples of the particulate material were collected and examined/analyzed by SEM/EDX; the results indicated that some of the particles were primarily CsI with a general background of cesium.

Structural (Zr and Si) and impurity (S) elements were generally present also.

6. Although the cladding was embrittled by oxidation and had partially disintegrated, no evidence of melting or fuel/cladding interaction was apparent in the preliminary examination of radial sections.
7. This test was conducted at the same temperature as test HI-2 but with the steam flow reduced in order to determine the effect of cladding and fuel oxidation on fission product release. The release of Cs, I, and Kr was less than half that in HI-2, but it is not certain that the degree of oxidation alone was responsible for the difference. The large axial crack that occurred in HI-2 probably was a significant factor.

Further microstructural and microchemical examinations of the fuel will be conducted at Argonne National Laboratory and will be reported separately.

## 2. INTRODUCTION

This report summarizes data from the fifth test in a series designed to investigate fission product release from LWR fuel in steam throughout the temperature range of 1400 to  $\sim 2400^{\circ}\text{C}$ .<sup>1</sup> Earlier tests, conducted under similar conditions at temperatures of 500 to  $1600^{\circ}\text{C}$ , have been reported by Lorenz et al.<sup>2-5</sup> 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 light water reactors (LWRs). The primary objectives of this program are:

1. to determine fission product release from discharged LWR fuel at temperatures up to and including fuel melting ( $\sim 2400^{\circ}\text{C}$ );
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  $\text{UO}_2$  containing a range of fission product elements) will also be investigated, but only at test temperatures  $>2000^{\circ}\text{C}$ . As in the first four tests, all remaining tests will be conducted in a flowing mixture of steam and helium (or argon) at atmospheric pressure; steam concentrations will be varied to simulate different accident sequences or core locations.

Test temperatures in the existing induction furnace are limited by material properties to a maximum of  $\sim 2000^{\circ}\text{C}$ . Higher-temperature tests are planned but will require the replacement of the currently used  $\text{ZrO}_2$  ceramics with  $\text{ThO}_2$ . In addition, the existing fission product collection and analysis system will be expanded to provide broader sampling capability with the higher-temperature furnace.

This report provides a brief description of test HI-5 and a tabulation of all the results obtained. As noted in the previous data summary reports,<sup>6-9</sup> thorough data evaluation and correlation will be included in a subsequent topical report covering the first series of fission product release tests at temperatures up to  $2000^{\circ}\text{C}$ .

### 3. TEST DESCRIPTION OF TEST HI-5

The matrix of tests planned for this project includes temperature ( $1700$  to  $2400^{\circ}\text{C}$ ) as the dominant variable; steam flow rate ( $0.3$  to  $1.5$  L/min) and time ( $1$  to  $30$  min), both of which affect the extent of oxidation, are secondary variables. The objective of this particular test was to obtain release data at  $1700^{\circ}\text{C}$  for  $20$  min in steam flowing at  $0.3$  L/min. The steam flow rate in test HI-2, also at  $1700^{\circ}\text{C}$ , was  $1.0$  L/min. In addition, a miniature aerosol sampler was included in the collection apparatus for the first time.

#### 3.1 FUEL SPECIMEN DATA

The test specimen was a  $15.2$ -cm-long section of rod 08639 from a  $15 \times 15$  fuel assembly (serial No. 1D13), which operated in the Oconee Unit 1 reactor from Feb. 11, 1975, to Nov. 22, 1979.<sup>10</sup> Details of the irradiation and of the characteristics of this particular specimen are listed in Table 1; calculated fission product inventories for the specimen are shown in Tables 2 and 3.

An axial scan of the total gamma radioactivity along the fuel rod is shown in Fig. 1. Reductions in burnup resulting from the flux depression caused by the grid spacers are apparent. This gamma scan is dominated by  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  but also includes the emissions from such long-lived, gamma-emitting nuclides as  $^{106}\text{Ru}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{125}\text{Sb}$ ,  $^{144}\text{Ce}$ , and  $^{154}\text{Eu}$ . The total gas release (Kr and Xe) from the entire fuel rod was reported to be  $3.35\%$ .<sup>10</sup> 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  $20$  and  $120$  in. The test specimens were cut from section 13, as indicated in Fig. 1. Thus, we calculate that  $\sim 4.1\%$  of the krypton and xenon was released from the test specimen during irradiation.

In addition to the test HI-5 fuel specimen, three short samples ( $1$  to  $2$  cm long) were cut from section 13 for metallographic examination and chemical analysis.

Table 1. Data for fuel specimen used in test HI-5

---

Fuel rod identification	Rod 08639 Assembly 1D13 Oconee Unit 1 (PWR)
Irradiation data	
Period	Feb. 11, 1975 to Nov. 22, 1979 (four cycles)
Assembly burnup <sup>a</sup>	~40 MWd/kg U
Specimen data	
Length	15.2 cm
Fuel loading	107.47 g UO <sub>2</sub> (94.73 g U)
Total weight	133.02 g
Weight of Zircaloy	25.55 g
Gas release during irradiation	3.35% (from whole rod) 4.1% (from specimen)

---

<sup>a</sup>The reported values varied between 38 and 42 MWd/kg U depending on measurement method. (Values taken from G. M. Bain and W. A. McInteer, Data Package for High Burnup Fuel Samples, LRC 4063, Lynchburg Research Center, Babcock and Wilcox Co., January 1983.)

Table 2. Amounts of principal fission and activation product elements in Oconee-1 fuel after 603 days of decay:<sup>a</sup> specimen HI-5

Element	Amount in fuel rod, 2.216 kg of initial heavy metal (g)	Amount in <sup>b</sup> HI-5 specimen (mg)
Se	0.1419	6.065
Br	0.0542	2.317
Kr	0.9087	38.84
Rb	0.8554	36.56
Sr	2.101	89.80
Y	1.127	48.17
Zr <sup>c</sup>	492.5	2.105 × 10 <sup>4</sup>
Mo	8.514	363.9
Tc	1.955	83.56
Ru	5.832	249.3
Rh	1.118	47.79
Pd	3.780	161.6
Ag	0.2056	8.788
Cd	0.3123	13.35
In	0.0055	0.0236
Sn	8.142	348.0
Sb	0.0678	2.898
Te	1.244	53.17
I	0.6098	26.07
Xe	13.64	583.0
Cs	6.718	287.2
Ba	3.884	166.0
La	3.092	132.2
Ce	6.137	262.3
Pr	2.829	120.9
Nd	10.13	433.0
Pm	0.1795	7.67
Sm	2.018	86.26
Eu	0.4146	17.72
Gd	0.2967	12.68
Total fission and activation products	386.0	1.65 × 10 <sup>4</sup>
U	2105	8.998 × 10 <sup>4</sup>
Pu	21.68	926.7
Total actinides	2129	9.1 × 10 <sup>4</sup>

<sup>a</sup>Calculated by C. W. Alexander using the ORIGEN computer program, with a burnup of 38.3 MWd/kg and a 603-d decay to July 15, 1981.

<sup>b</sup>Original uranium content of 15.2-cm fuel specimen was 94.72 g; initial enrichment was 3.2% <sup>235</sup>U. Thus, the fuel specimen was 94.72 g/2216 g = 4.27% of a fuel rod.

<sup>c</sup>Calculated amounts in the Zircaloy cladding.

Table 3. Principal radionuclides (including activation products) and selected stable nuclides in Oconee-1 fuel after 603 days of decay:<sup>a</sup> specimen HI-5

Nuclide	Amount in fuel rod, 2.216 kg of initial heavy metal		Amount in HI-5 specimen <sup>b</sup>	
	(g/rod)	(Ci/rod)	(mg)	(mCi)
<sup>54</sup> Mn	$9.35 \times 10^{-8}$	$7.24 \times 10^{-4}$	$3.997 \times 10^{-6}$	0.031
<sup>60</sup> Co	$1.80 \times 10^{-4}$	0.2036	$7.694 \times 10^{-3}$	8.703
<sup>65</sup> Zn	$4.798 \times 10^{-6}$	0.0395	$2.051 \times 10^{-4}$	1.688
<sup>83</sup> Kr	0.0970	0.0	4.146	0.0
<sup>84</sup> Kr	0.2884	0.0	12.33	0.0
<sup>85</sup> Kr	0.0497	19.52	2.124	834.4
<sup>86</sup> Kr	0.4707	0.0	20.12	0.0
<sup>90</sup> Sr	1.235	168.5	52.79	7202
<sup>106</sup> Ru <sup>e</sup>	0.0996	333.3	4.257	$1.425 \times 10^4$
<sup>110m</sup> Ag <sup>d</sup>	$3.780 \times 10^{-4}$	1.795	0.01616	76.72
<sup>125</sup> Sb	0.020	20.66	0.8549	883.1
<sup>129</sup> I	0.4622	$8.16 \times 10^{-5}$	19.76	0.0034
<sup>131</sup> Xe	1.029	0.0	43.98	0.0
<sup>132</sup> Xe	2.856	0.0	122.1	0.0
<sup>134</sup> Xe	3.756	0.0	160.5	0.0
<sup>136</sup> Xe	5.950	0.0	254.3	0.0
<sup>134</sup> Cs	0.1671	216.2	7.14	9233
<sup>137</sup> Cs	2.880	250.6	128.1	$1.071 \times 10^4$
<sup>144</sup> Ce	0.121	386.2	5.172	$1.651 \times 10^4$
<sup>147</sup> Pm	0.1794	166.4	7.668	7113
<sup>154</sup> Eu	0.0960	25.92	4.103	1108
Total fission products	87.32	2752	3732	$1.176 \times 10^5$
<sup>235</sup> U	13.37	$2.89 \times 10^{-5}$	571.5	$1.235 \times 10^{-3}$
<sup>236</sup> U	9.53	$6.17 \times 10^{-4}$	407.3	0.0264
<sup>238</sup> U	2082	$7.0 \times 10^{-4}$	$8.899 \times 10^5$	0.0299
<sup>238</sup> Pu	0.499	8.553	21.35	365.6
<sup>239</sup> Pu	11.31	0.703	483.4	30.05
<sup>240</sup> Pu	5.66	1.291	241.9	55.18
<sup>241</sup> Pu	2.84	293.1	121.4	$1.253 \times 10^5$
Total actinides	2129	324.1	$9.10 \times 10^4$	$1.39 \times 10^4$

<sup>a</sup>Calculated by C. W. Alexander using the ORIGEN computer program, with a burnup of 38.3 MWd/kg and a 603-d decay to July 15, 1981.

<sup>b</sup>Original uranium content of 15.2-cm fuel specimen was 94.72 g; initial enrichment was 3.2% <sup>235</sup>U. Thus, the fuel specimen was 94.72 g/2216 g = 4.272% of a fuel rod.

<sup>c</sup>Sum of fission product (99.97%) and activation product (0.030%).

<sup>d</sup>Sum of fission product (99.9%) and activation product (0.01%).

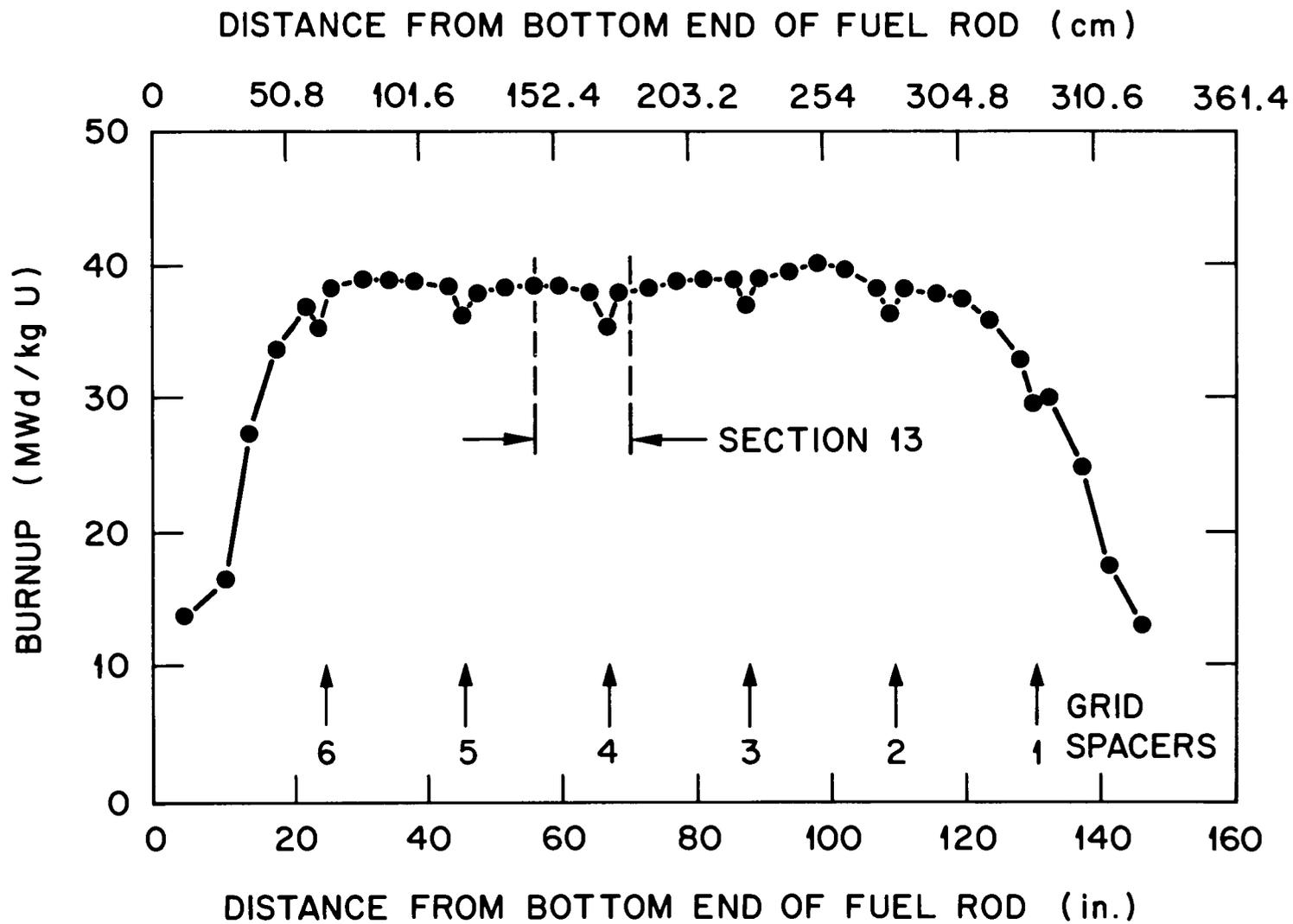


Fig. 1. Gross gamma scan of fuel rod 08639 from Ocone 1. The test HI-5 specimen was cut from section 13.

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  $\text{UO}_2$  fuel during subsequent handling. 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. 2.

### 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,<sup>2-4</sup> fuel rod burst experiments,<sup>11</sup> and molten fuel tests.<sup>12</sup> The graphite susceptor was protected from steam oxidation by a blanket of helium gas that leaked through joints in the  $\text{ZrO}_2$  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 Fig. 4. The fission product collection system included a platinum thermal gradient tube,\* 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. 4, prior to reaching the cooled charcoal. Instrumentation included two Pt/Rh thermocouples and an optical pyrometer for temperature measurement of the test specimen, three NaI(Tl) radiation detectors connected to a multichannel analyzer, and conventional electrical and gas flow instruments. A data acquisition system (Fig. 5) 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 4.

### 3.3 TEST CONDITIONS AND OPERATING PROCEDURE

The test was conducted under the operating conditions listed in Table 5. 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 assemblies 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  $\sim 300^\circ\text{C}$  and purged with helium. Also, all connecting lines to the furnace—thermal gradient tube—filter pack assemblies were preheated to at least  $125^\circ\text{C}$  to prevent steam condensation during the test.

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\*Hereafter in this report, thermal gradient tube will be abbreviated frequently to TGT.

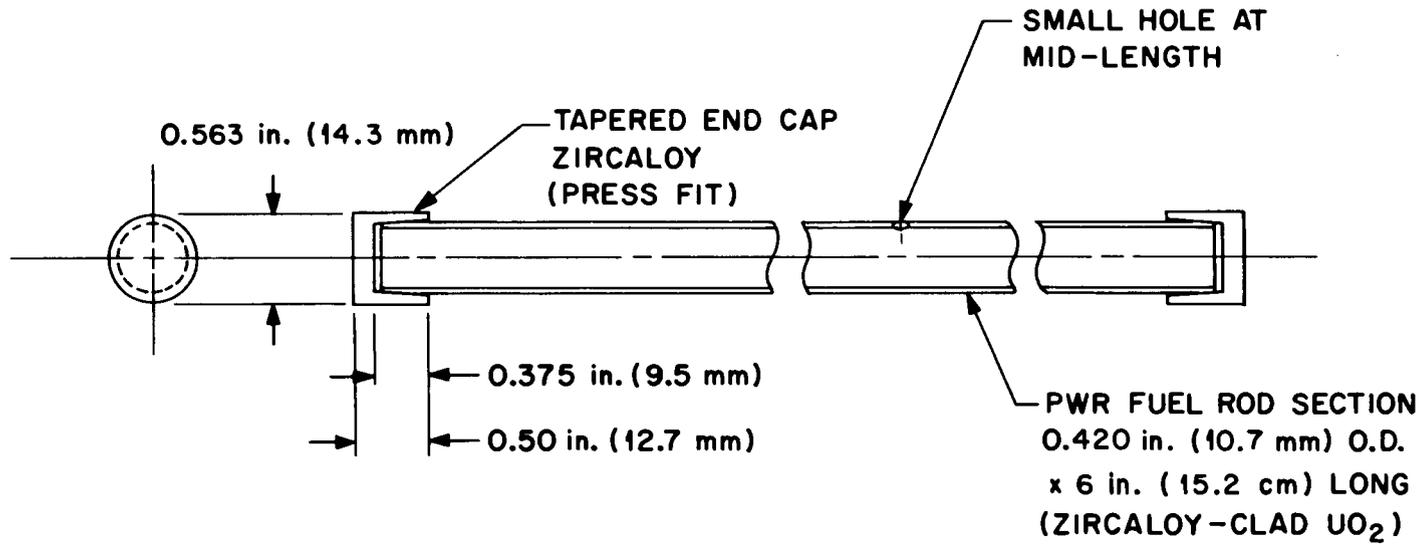


Fig. 2. Fuel specimen for fission product release studies.

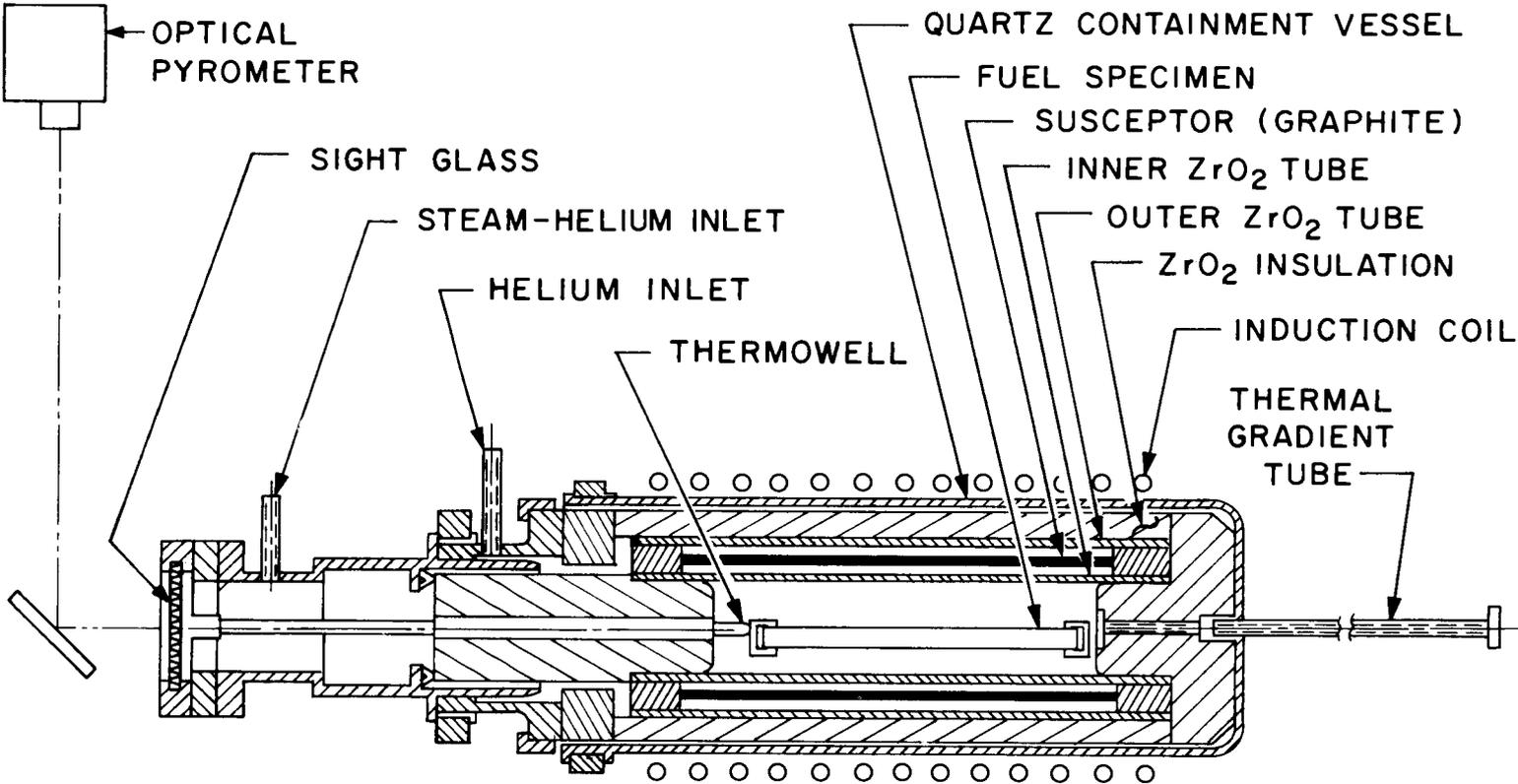


Fig. 3. Fission product release furnace.

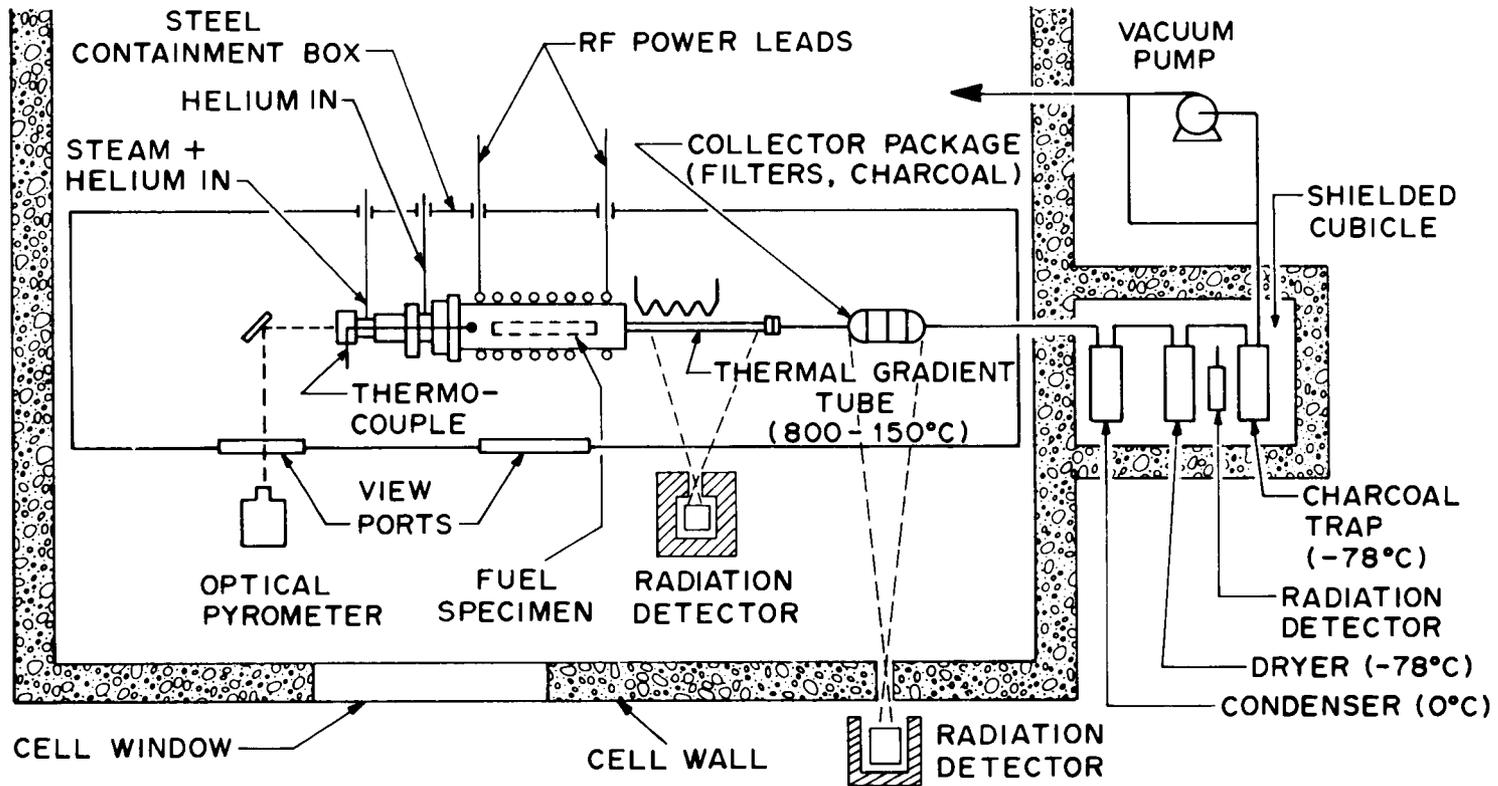


Fig. 4. Fission product release and collection system.

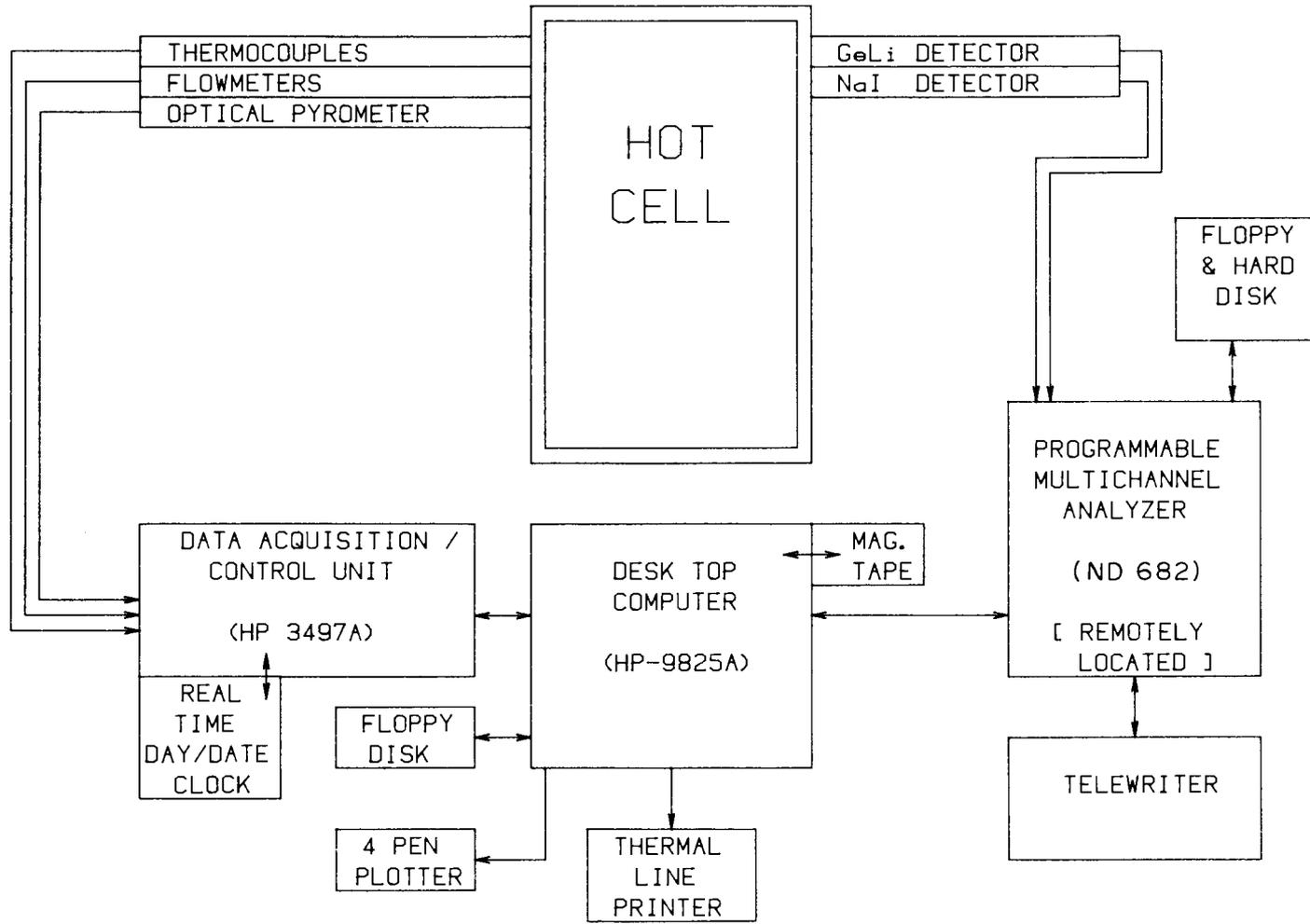


Fig. 5. Data acquisition and processing system for fission product release test.

Table 4. Summary of significant changes in apparatus and test conditions for the five HI tests

Component/condition	Test HI-1	Test HI-2	Test HI-3	Test HI-4	Test HI-5
Furnace thermocouple	Pt-10% Rh vs Pt (bare)	Pt-10% Rh vs Pt (bare)	W-5% Re vs W-26% R (ZrO <sub>2</sub> thermowell)	W-5% Re vs W-26% Re (ZrO <sub>2</sub> thermowell)	Pt-10% Rh vs Pt (bare)
Susceptor	Tungsten	Tungsten	Graphite	Graphite	Graphite
Pretreatment of fibrous ZrO <sub>2</sub> insulator	None	Zirconyl nitrate (one coat)	Zirconyl nitrate (one coat)	Zirconyl nitrate (two coats)	Zirconyl nitrate (one coat)
Thermal gradient tube liner	Platinum	Platinum/gold	Platinum	Platinum	Platinum
Thermal gradient tube length, cm	30.5	35.6	35.6	35.6	35.6
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 steel	4.6-mm ID × 190-mm long stainless steel
Entrance cone to filter pack	Teflon	Teflon	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
Dryer and cold charcoal temperature, °C	-78	-78	-195	-195	-78
Inert gas	Argon	Argon	Helium	Helium + 0.05% H <sub>2</sub>	Helium
Aerosol sampler	None	None	None	None	Graphite rod, screw-driven

Table 5. Operating data for test HI-5

Specimen temperature at start of heating ramp	270
Heatup rate	1.1°/s (66°/min)
Test temperature	1700°C ± 50
Time at test temperature	20 min
Nominal flow rate data: <sup>a</sup>	
He purge to thermocouple	0.10 L/min
He purge to graphite susceptor	0.15 L/min
He to steam generator	0.15 L/min
Steam into system	0.41 L/min
Volume flow data: <sup>b</sup>	
He purge to thermocouple	7.02 L
He purge to susceptor	9.87 L
He to steam generator <sup>c</sup>	9.45 L
H <sub>2</sub> generated <sup>d</sup>	11.0 L
CO generated <sup>e</sup>	3.30 L

<sup>a</sup> Measured by mass flow meters, L at 20°C and 1-atm absolute pressure.

<sup>b</sup> Measured by totalizers on mass flow meters during the 50 min of steam flow into apparatus, operating at room temperature = 23°C.

<sup>c</sup> Absolute pressure in furnace during test was 0.1044 MPa (783 mm Hg).

<sup>d</sup> At atm = 0.0994 MPa (745.5 mm Hg).

<sup>e</sup> Based on 1.77-g weight loss from graphite susceptor during test, assuming all carbon converted to CO.

The furnace thermocouple indicated slight temperature overshoot (to  $\sim 1750^{\circ}\text{C}$ ) at the beginning of the test; this probably damaged the thermocouple, since it failed  $\sim 4$  min later, as may be seen from Fig. 6, and was lost for the remainder of the test. The optical pyrometer, however, operated reliably throughout the test period.

### 3.4 POSTTEST DISASSEMBLY AND SAMPLE COLLECTION

After the test was completed, the apparatus was disassembled. Initially, the filter assembly and the liner of the TGT were removed and transferred to another hot cell to avoid potential contamination from fuel handling. Secondly, the fuel specimen was removed from the furnace; in order to minimize disturbance of the severely damaged fuel, only the  $\text{ZrO}_2$  was touched. The specimen and boat were pushed into a clear plastic tube to limit contamination of the hot cell during visual inspection and photography. The fuel specimen, shown in Fig. 7, appeared to be heavily oxidized only on the inlet end. The central part of the specimen was severely fractured, but both ends were relatively intact. Since the steam flow rate was low during the test, the limited oxidation of the cladding was not surprising. The test period (20 min) was not long enough to allow complete oxidation; therefore, the outlet end was under a reducing hydrogen blanket during much of the test period. Consequently, much of the Zircaloy cladding should have hydrided first, then oxidized later.

As in previous experiments, the  $\text{ZrO}_2$  and the fuel specimen were cast in epoxy resin to preserve the relative sample integrity during handling and transfer to another hot cell. This assembly was analyzed for radioactive fission product content by gamma spectrometry. It was also gamma-scanned at 1-cm intervals to determine fission product distribution along the specimen. These data were compared with pretest data (Sect. 4.2.7). The fuel specimen was then transferred to the High Radiation Level Examination Laboratory, where it was cut into radial sections for detailed inspection (Sect. 4.2.8). 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 6.

The filters and TGT liner were weighed before and after the test. These data showed that the TGT and glass wool prefilter gained 0.04 and 0.11 g, respectively.

Because of the high level of radioactivity, the platinum TGT liner was gamma-ray analyzed through 2.54 cm of lead. It was also gamma-scanned without the lead shielding through a 1.0-cm slit at 1.0-cm intervals to determine the distribution of radioactivity (primarily  $^{137}\text{Cs}$ ) with temperature (see Sect. 3.2.4). Subsequently, the TGT was cut into 11 sections for individual analysis. A graphite electrode smear sample from each of these sections and from the glass wool prefilter was taken for spark-source mass spectrometric (SSMS) analysis.

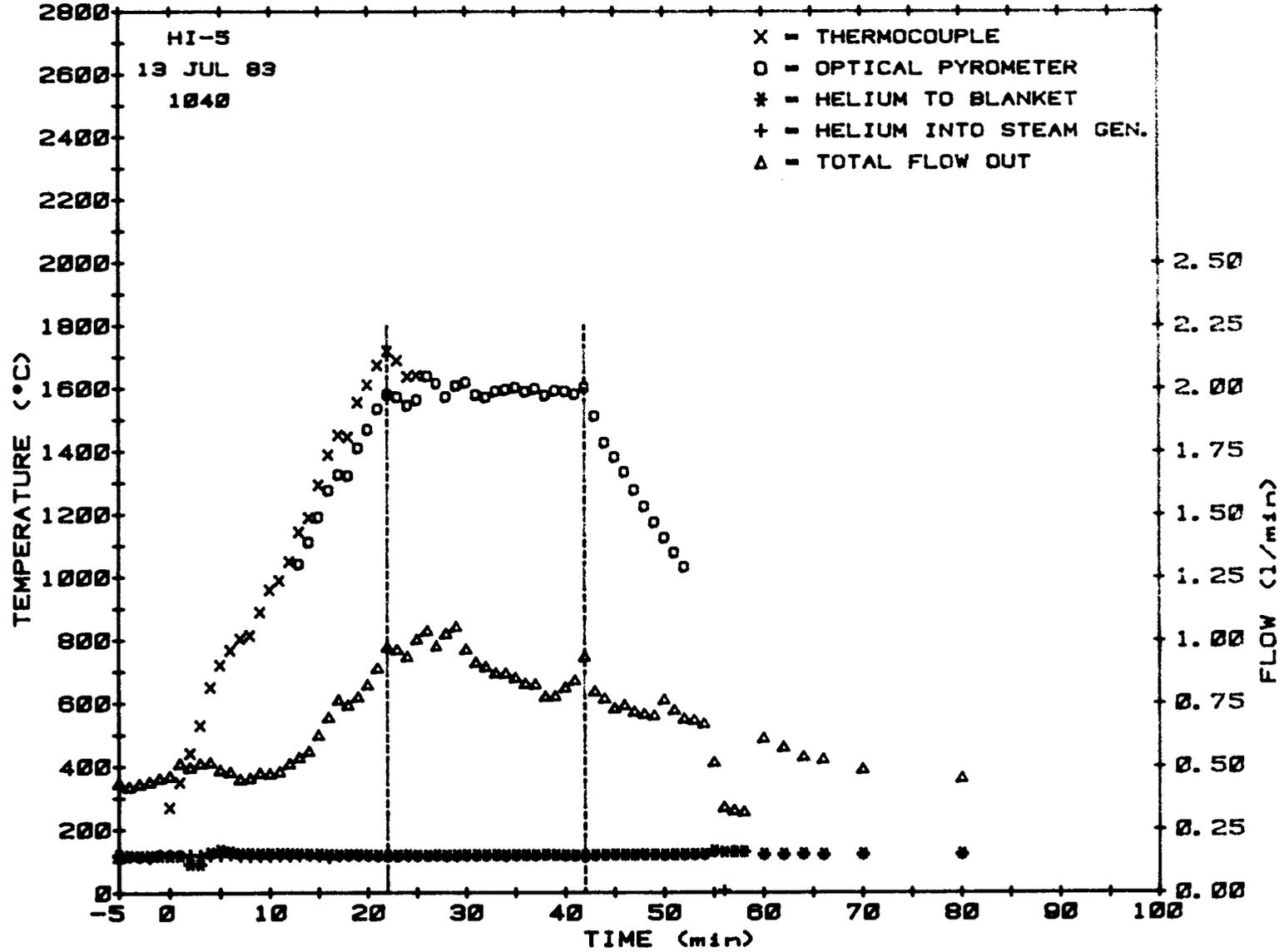


Fig. 6. Temperature and flow history for test HI-5.

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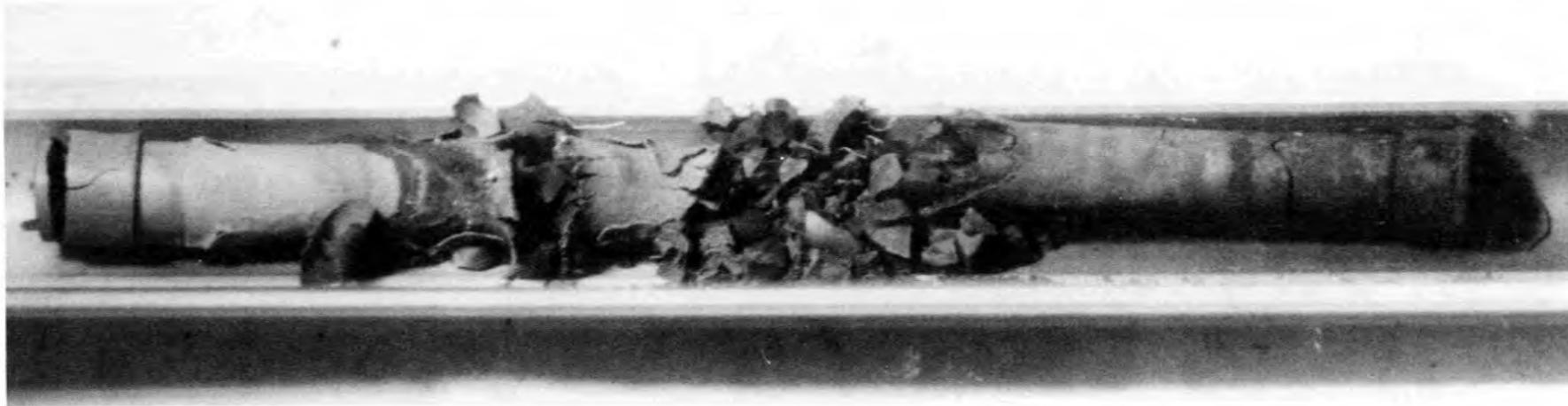


Fig. 7. Posttest appearance of fuel specimen from test HI-5. Note heavily oxidized (white) area at steam inlet and (left) as opposed to less oxidized (dark) area at outlet end (right).

Table 6. Analytical techniques for fission product release analysis

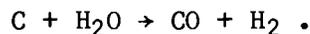
Technique	Time	Location	Elements
Gamma spectrometry	Pretest, posttest	Fuel specimen	Long-lived, high energy fission products — Ru, Ag, Sb, Cs, Ce, Eu
	On-line	Thermal gradient	Cs, Kr
	Posttest	Furnace components, thermal gradient tube, filters	Ru, Ag, Sb, Cs, Ce, Eu
Activation analysis	Posttest	Charcoal, 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

Each section of the TGT platinum liner and each of the filters were analyzed by gamma spectrometry before and after leaching successively with basic ( $\text{NH}_4\text{OH} + \text{H}_2\text{O}_2$ ) and acidic ( $\text{HNO}_3 + \text{HF}$ ) solutions. The quartz furnace vessel, the quartz thermal gradient tube housing, 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 and of the charcoal from the filter assembly.

#### 4. TEST RESULTS

##### 4.1 TEST DATA

The temperature and flow history of the entire test is presented in Fig. 6. These are uncorrected data; posttest temperature calibrations showed that the temperature indicated by the optical pyrometer was  $\sim 150^\circ\text{C}$  lower than the actual temperature at the inlet end of the 15.2-cm-long fuel specimen. There was an axial temperature gradient of  $\sim 75^\circ$ , with  $T_{\text{max}}$  near the inlet end and  $T_{\text{min}}$  at the outlet end of the specimen. Test operating conditions are summarized in Table 5. The helium flow rates were determined from mass flowmeters, and the steam flow rate was based on pretest calibrations of the steam generator and on the amount of water collected in the condenser and dryer. All weight loss from the graphite susceptor was assumed to result from the reaction with steam:



Using the data shown in Table 5 and the reported conversion factors for the measurement of CO and  $\text{H}_2$  in the mass flowmeters, we calculate that  $\sim 70\%$  of the 25.5 g of zirconium in the fuel specimen cladding and end caps should have been converted to  $\text{ZrO}_2$ . Based on subsequent observations of the fuel specimen, however, this value appears to be high. A more accurate measure of the extent of oxidation should be obtained from microstructural examination being conducted at Argonne National Laboratory.

The release histories of  $^{85}\text{Kr}$  to the cold charcoal traps and of the  $^{137}\text{Cs}$  to the TGT and the filters are shown in Fig. 8. These values were determined as relative rates during the test, and quantitative posttest 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 curves, significant release of both  $^{85}\text{Kr}$  and  $^{137}\text{Cs}$  occurred during cooling.

##### 4.2 POSTTEST DATA

Gamma-ray spectrometric analysis of the disassembled apparatus components revealed that the principal radioactive species present were  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ . 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 activation analysis for  $^{129}\text{I}$ , is

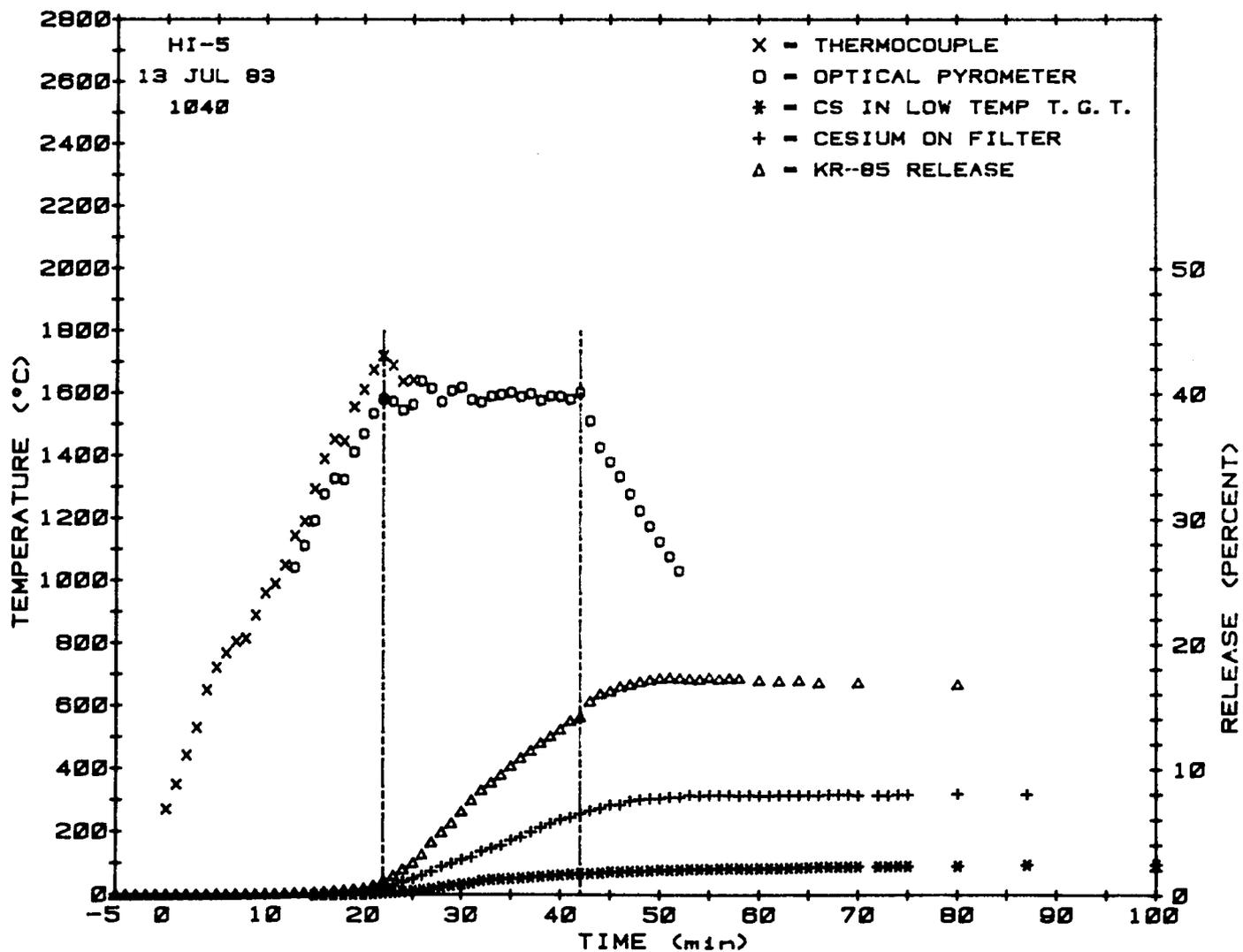


Fig. 8. Collections of krypton and cesium as functions of temperature and time in test HI-5.

presented in Table 7. Note that the krypton and cesium releases are similar, 19.9 and 20.3% respectively, a usual observation in this series of tests. The krypton value includes an estimated 4.1% of the specimen 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.

Table 7. Distribution and fractional release of fission products in test HI-5

Test component or collector	Temperature or range (°C)	Fraction of fission product inventory <sup>a</sup> found (%)				
		<sup>85</sup> Kr	<sup>137</sup> Cs	<sup>129</sup> I	<sup>125</sup> Sb <sup>b</sup>	<sup>110m</sup> Ag <sup>b</sup>
Furnace	1000-1750	0	7.68	0.135	0.323	17.79
Thermal gradient tube	200-810	0	4.71	7.36	0.003	0.046
Aerosol sampler	150	0	0.050	0.100	0	0
Filters	140	0	7.81	14.74	10 <sup>-5</sup>	0.175
Hot charcoal	140	0	0	0.017	0	0
Cold charcoal	-78	19.9 <sup>c</sup>	0	0	0	0
Totals		19.9 <sup>c</sup>	20.25	22.35	0.326	18.01

<sup>a</sup>Inventory based on ORIGEN calculations by C. W. Alexander.

<sup>b</sup>Large amounts of <sup>134</sup>Cs and <sup>137</sup>Cs interfered with analyses of <sup>125</sup>Sb and <sup>110m</sup>Ag prior to leaching. In addition, 98% of Sb and Ag found in the furnace was associated with the ZrO<sub>2</sub> outlet end plugs. A small amount of <sup>154</sup>Eu (0.021% of inventory) was found on these same ZrO<sub>2</sub> components.

<sup>c</sup>About 4.1% of the <sup>85</sup>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.

The apparent release of antimony was low compared to cesium and iodine, probably because the cladding was not completely oxidized. The cesium radioactivity interfered with the detection of antimony, so some released antimony may not have been detected. Antimony appears to be retained by unoxidized Zircaloy,<sup>8,9</sup> thereby limiting the release rate from the furnace. The released antimony was found on ZrO<sub>2</sub> surfaces near the

inlet end of the test specimen; posttest examination revealed that only the inlet end was heavily oxidized.

#### 4.2.1 Results from Gamma Spectrometry

Table 8 provides the detailed results of gamma spectrometric analysis for  $^{137}\text{Cs}$ . Approximately 38% of the released cesium remained inside the  $\text{ZrO}_2$ -lined furnace, primarily on components at the outlet end. Of the  $^{137}\text{Cs}$  that escaped the furnace, less than half deposited on the platinum TGT, and the remainder deposited on the filters. No cesium was found beyond the filter package. The distribution of cesium throughout the test apparatus is illustrated in Fig. 9; the iodine distribution, as determined by component leaching and activation analysis for  $^{129}\text{I}$ , is shown for comparison. About ten times more cesium was released than iodine, in good agreement with the ORIGEN value of 11.1 for the fuel inventories (Table 2). The largest concentrations of cesium were found on the zirconia outlet end plug and the glass wool prefilter. The deposit on the plug indicates a possible reaction (perhaps the formation of cesium zirconate, and the cesium on the filter probably was transported there as an aerosol).

Significant amounts of  $^{110m}\text{Ag}$  and  $^{125}\text{Sb}$  were found on the furnace components and on some sections of the TGT. (In general, the analyses for  $^{110m}\text{Ag}$  and  $^{125}\text{Sb}$  were possible only after removal of a large fraction of the  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  by leaching.) These data are summarized in Table 9. Although some 17% of the silver and 0.28% of the antimony were released from the fuel, very small fractions escaped from the furnace. The behavior of these elements in the TGT will be discussed in more detail in Sect. 3.2.5.

#### 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  $^{129}\text{I}$  to  $^{130}\text{I}$ , 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 in the  $\text{NH}_4\text{OH} + \text{H}_2\text{O}_2$  solution. Small aliquots of the solutions were chemically treated to remove cesium, loaded onto ion exchange media, irradiated, and 12.4-h  $^{130}\text{I}$  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 10. The total fraction of iodine released, 22.35%, was a minimum value. Since iodine cannot be detected directly, it has to 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. In this test, however, the good agreement in the fractional release values for Kr, I, and Cs indicates that no significant amount of iodine was missed. Iodine that collected on the glass fiber filters probably was transported there by being deposited on particulate material. Very small amounts of the iodine ( $\sim 0.01\%$ ) were collected on the

Table 8. Fractional release and distribution of cesium in test HI-5

Location	Temperature (°C)	Cesium found in each location			
		Amount ( $\mu\text{Ci } ^{137}\text{Cs}$ )	(mg total Cs)	Percentage of specimen inventory <sup>a</sup>	Percentage of released
<b>Furnace components</b>					
Inlet end components	~1000	5.973+1 <sup>b</sup>			
Outer ZrO <sub>2</sub> tube	~1700	2.242+1			
Fibrous ZrO <sub>2</sub> insulator	800-1600	8.471+1			
Graphite susceptor	1800	1.240+2			
Inner ZrO <sub>2</sub> tube	1700	1.559+4			
Outlet end components	~1000	5.750+4			
ZrO <sub>2</sub> outlet end plugs	1000-1700	7.038+5 <sup>c</sup>			
Miscellaneous debris	800-1700	1.254+4			
Quartz vessel	~800	3.215+4			
Total		8.219+5	2.204+1	7.68	37.9
<b>Thermal gradient tube</b>					
Quartz tube	200-800	1.072+4			
Pt segment 1		7.443+1			
Pt segment 2		1.313+2			
Pt segment 3		1.006+2			
Pt segment 4		1.742+3			
Pt segment 5		3.156+4 <sup>c</sup>			
Pt segment 6		2.779+5 <sup>c</sup>			
Pt segment 7		8.390+4 <sup>c</sup>			
Pt segment 8		4.122+4 <sup>c</sup>			
Pt segment 9		3.285+4 <sup>c</sup>			
Pt segment 10		2.426+4 <sup>c</sup>			
Pt segment 11		1.753+4 <sup>c</sup>			
Total		5.045+5	1.353+1	4.71	23.3
Aerosol sampler	140	5.364+3	1.433-1	0.050	0.25
<b>Filter package</b>					
Entrance tube	140	1.074+3			
Entrance cone		4.250+2			
Glass wool prefilter		8.215+5 <sup>c</sup>			
First HEPA filter		1.323+4			
Second HEPA filter		2.271+1			
Heated charcoal		2.72-3			
Miscellaneous parts		1.086+2			
Total		8.364+5	2.242+1	7.81	38.6
Total all components		2.168+6	5.813+1	20.25	100

<sup>a</sup>Based on a burnup of 38.3 MWd/kg, the test fuel specimen contained 287.1 mg total cesium and 10.71 Ci <sup>137</sup>Cs, decay corrected to July 15, 1981. (ORIGEN calculation by C. W. Alexander, ORNL.)

<sup>b</sup>Exponential notation: 5.973+1 = 5.973 × 10<sup>1</sup>, etc.

<sup>c</sup>Analyzed through 2 to 5 cm of lead because of high radioactivity and corrected for attenuation.

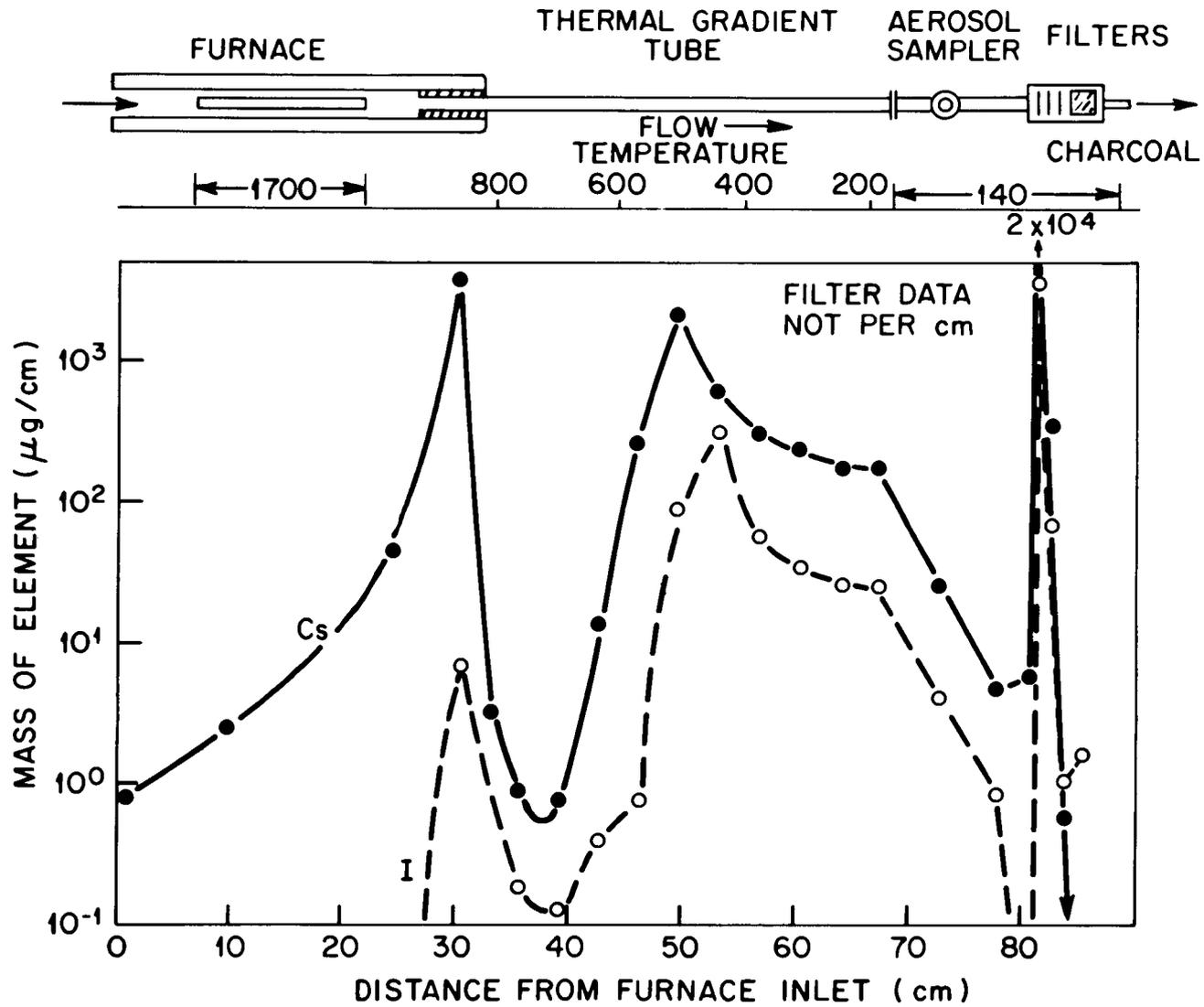


Fig. 9. Distribution of cesium and iodine in test HI-5 apparatus.

Table 9. Fractional release and distribution of silver and antimony found by gamma spectrometry in test HI-5

Location	Temperature (°C)	Silver			Antimony			
		( $\mu\text{Ci } ^{110\text{m}}\text{Ag}$ )	( $\mu\text{g Ag}$ ) <sup>a</sup>	Percentage of inventory	( $\mu\text{Ci } ^{125}\text{Sb}$ )	( $\mu\text{g Sb}$ ) <sup>b</sup>	Percentage of inventory	
<b>Furnace components</b>								
Inlet end components	~1000		1.775		6.966			
ZrO <sub>2</sub> insulator tubes	800-1600		15.59		3.004			
Graphite susceptor	1800		9.414		ND			
Outlet end components	~1000		264.4		ND			
ZrO <sub>2</sub> outlet end plugs	1000-1700	13350			2817			
Miscellaneous debris	800-1700		13.90		26.51			
Quartz vessel	~800		ND		ND			
<b>Total</b>		13650	1563	17.79	2854	9.367	0.323	
<b>Thermal gradient tube</b>								
Pt segment 1	200-800		0.970		0.6893			
Pt segment 2			5.340		9.6893			
Pt segment 3			8.152		10.07			
Pt segment 4			2.200		4.136			
Pt segment 5			ND		ND			
Pt segment 6			ND		ND			
Pt segment 7			5.201		ND			
Pt segment 8			4.136		ND			
Pt segment 9			2.892		ND			
Pt segment 10			3.661		ND			
Pt segment 11			2.995		ND			
<b>Total</b>			35.55	4.071	0.046	24.58	0.0807	0.003
Filter package	140	134 <sup>c</sup>	15.3	0.175	0.0653	2 × 10 <sup>-4</sup>	7 × 10 <sup>-6</sup>	
<b>Total all components</b>		13820	1582	18.01	2879	9.448	0.326	

<sup>a</sup>Based on inventory data: 8.787 mg Ag and 76.72 mCi <sup>110m</sup>Ag in specimen.

<sup>b</sup>Based on inventory data: 2.898 mg Sb and 883.1 mCi <sup>125</sup>Sb in specimen.

<sup>c</sup>Detected only after leaching.

Table 10. Fractional release and distribution of iodine in test HI-5  
(results of activation analysis for  $^{129}\text{I}$ )

Location	Temperature (°C)	Iodine found at each location		Percentage of specimen inventory <sup>a</sup>	Percentage of released	Ration of cesium to iodine ( $\mu\text{g Cs}/\mu\text{g I}$ )
		Amount ( $\mu\text{g }^{129}\text{I}$ )	( $\mu\text{g total I}$ )			
Furnace components						
Quartz vessel	~800	4.75	6.27			137
ZrO <sub>2</sub> outlet end plug	1000-1700	17.90	23.63			800
Fibrous ZrO <sub>2</sub> insulator	800-1600	4.06	5.32			0.43
Total <sup>b</sup>		26.68	35.22	0.135	0.60	625
Thermal gradient tube 200-800						
Quartz tube (all)		8.50	11.22			25.6
Segment 1		0.024	0.032			62.6
Segment 2		0.056	0.074			47.6
Segment 3		0.033	0.044			61.3
Segment 4		1.10	1.45			32.2
Segment 5		2.10	2.77			305
Segment 6		245.9	324.6			23.0
Segment 7		816.9	1078			2.09
Segment 8		159.0	209.9			5.26
Segment 9		93.75	123.8			7.11
Segment 10		75.02	99.03			6.57
Segment 11		49.14	64.87			7.25
Total		1452	1917	7.36	32.91	7.05
Aerosol sampler	140	19.71	26.02	0.100	0.45	5.51
Filter package 140						
Entrance tube		3.93	5.19			5.55
Entrance cone		0.006	0.008			1400
Glass wool prefilter		2854	3767			5.85
First HEPA filter		53.28	70.33			5.04
Second HEPA filter		0.77	1.02			0.60
Charcoal		3.27	4.32			$1.7 \times 10^{-5}$
Total		2915	3847	14.76	66.04	5.83
Total all components		4413	5825	22.35	100	9.98

<sup>a</sup>Based on an average burnup of 38.3 MWd/kg, the test specimen contained 19.76 mg of  $^{129}\text{I}$  and 26.06 mg of total iodine; decay corrected to July 15, 1981.

<sup>b</sup>Small amounts of iodine probably were present on other furnace components that were not leached for iodine analysis.

<sup>c</sup>In addition, 1.07  $\mu\text{g}$  of bromine was found on the charcoal; this value corresponds to 0.046% of the specimen inventory compared with 0.017% of the iodine inventory found on the charcoal.

second HEPA filter and the heated charcoal. Only the most volatile and penetrating forms of iodine, such as organic iodides and elemental iodine, are expected to penetrate the first HEPA filters.

The heated charcoal was also analyzed for bromine by activation analysis. The results showed  $\sim 1.07 \mu\text{g}$  bromine, which corresponds to  $\sim 0.047\%$  of the calculated specimen inventory (Table 2). Any bromine released from the test specimen would be expected to behave in a manner similar to that observed for iodine but with somewhat greater volatility.

The distribution of cesium and iodine throughout the test apparatus is illustrated in Fig. 9.

#### 4.2.3 Results of Spark-Source Mass Spectrometry (SSMS) Analysis

Elemental analyses were obtained by SSMS for graphite electrode smear samples at 11 locations along the platinum TGT and one from the glass wool filter. Following the sampling, the amount of  $^{137}\text{Cs}$  in each sample was determined by gamma spectrometry. These values were then used as the standards in determining mass values for the fission products, structural materials, and impurity elements that were detected on the samples.

Table 11 summarizes the results obtained by SSMS analysis. These data are divided into three groups: (1) fission products; (2) special materials, such as cladding (Zr and Sn) and apparatus structural materials (Zr, Mg, Ca, and Pt); and (3) other materials (impurities in the system). The values for the TGT were obtained by making the assumption that each smear sample was representative of all the deposited material in a 1-cm length of the TGT at the sampling location. The SSMS results for a smear sample were multiplied by the detected amount of  $^{137}\text{Cs}$  on that section (from gamma measurements) and divided by the known amount of  $^{137}\text{Cs}$  in the smear sample from SSMS measurement to obtain the mass of a particular element on that section of the TGT liner. The data for the glass prewool filter were treated in a similar fashion, but the SSMS values were multiplied by the total mass of  $^{137}\text{Cs}$  on the filter. This method of analysis should be considered only semiquantitative in nature; the accuracy is estimated to be about a factor of 3. However, it does provide information about those elements that are transported to the collection train but cannot be measured by gamma spectrometry or neutron activation analyses.

Selected data from Table 11 are plotted in Figs. 10-12 to illustrate the distribution along the TGT. The profiles for the three groups of deposited materials are illustrated in Fig. 10 along with the total material profile. The largest fraction of the fission products deposited at temperatures  $< 600^\circ\text{C}$ . In contrast, the largest fractions of the special materials and other materials deposited at temperatures  $> 600^\circ\text{C}$ . Cesium, cadmium, and rubidium were the major fission product metals that deposited in the TGT; iodine and bromine were the major fission product nonmetals. The cesium profile (Fig. 11) was determined by gamma spectrometry. There are two profiles for iodine. One was determined by neutron

Table 11. Results of spark-source mass spectrometric analyses of samples from test HI-5

Element	Mass of element found (mg/cm)											Filter-1 (mg total)
	Thermal gradient tube sample											
	1	2	3	4	5	6	7	8	9	10	11	
Distance from inlet (cm)	0.3	1.0	4.7	8.2	11.7	15.3	18.8	22.3	25.8	19.5	33.1	
<b>Fission products</b>												
Cs (R) <sup>a</sup>	0.003	0.001	0.001	0.013	0.238	2.10	0.651	0.311	0.241	0.179	0.177	22.0
Cd (R)						2.22	0.642	0.224	0.876	0.103	0.250	0.144
I (R)						0.015	0.321	0.224	0.175	0.044	0.010	0.124
Rb (R)				<0.011	0.272	0.741	0.019	0.013	0.009	0.0004	0.012	0.824
Br (R)					0.007	0.001	0.013	0.011	0.005	0.003	0.001	0.103
Ag (R)								0.0002	0.004	0.001		
Te						0.007	<0.002	0.002		0.001		
Ba			0.002			0.001				<0.0001	0.001	
Total	0.003	0.001	0.003	0.024	0.517	5.08	1.65	0.785	1.310	0.330	0.451	23.2
<b>Special materials<sup>b</sup></b>												
Zr (N)	0.007	0.030	0.009	0.030	0.027	0.001	0.001		0.0002	0.0004	0.003	0.412
Sn (N)			0.001			0.007	0.001	0.016	0.044	0.030	0.003	0.620
Mg					0.007	0.005	0.003	0.045	0.026	0.007	0.001	0.206
Ca	0.020	0.006	0.001	0.110	0.027	0.002	0.001	0.001	0.0003	0.0001	0.001	0.144
Total	0.027	0.036	0.011	0.140	0.061	~0.015	0.006	0.062	0.071	0.038	0.008	1.48
<b>Other materials<sup>c</sup></b>												
S	0.068	0.045	0.001	0.020	0.122	0.593	0.128	0.067	0.088	0.059	0.100	4.42
Zn	0.055	0.015	0.002	0.020	0.054	0.007	0.003	0.007	0.007	0.004	0.002	0.185
Cl	0.034	<0.015	0.001	0.221	0.027	0.007	0.006	0.007	0.004	0.003	0.003	0.412
Na	0.002	0.002	0.0002	0.387	0.136	0.0595	0.002	0.011	0.018	0.007	0.010	2.06
Fe	0.068	0.045	0.003	0.221	0.136	0.004	<0.001	0.005	<0.0004	<0.0004	0.013	0.412
K	0.003	0.002	0.0002	0.033	0.011	0.037	0.001	0.001	0.0002	0.0003	0.002	0.412
Al	0.041	0.005	0.002	0.039	0.003	0.002	<0.001	0.002	0.0003	0.0001	0.002	0.412
Cu	0.003	<0.0003		0.011	0.001	0.002				0.0003	0.001	0.206
Cr	0.006	0.005	0.0002		0.004	0.001	<0.001	0.0002	0.0002		0.001	0.021
Mn	<0.001					0.005	<0.003	<0.005		<0.0003	0.003	0.021
Pb			0.001			0.002		0.001		0.001	0.002	0.062
Ni	0.682	0.090	0.001	0.006	0.136	0.022	0.001	0.005		<0.0004	0.008	0.062
Total	0.963	0.244	0.011	1.000	0.630	0.741	0.147	0.110	0.118	0.076	0.147	8.68
Total all materials	0.993	0.261	0.025	1.164	1.21	5.84	1.80	0.957	1.50	0.444	0.606	33.36

<sup>a</sup>(R) denotes radiogenic isotopic distribution, (N) denotes natural isotopic distribution.

<sup>b</sup>Material known to be part of the system, such as fuel, cladding, and apparatus structural materials.

<sup>c</sup>Apparently impurities in system.

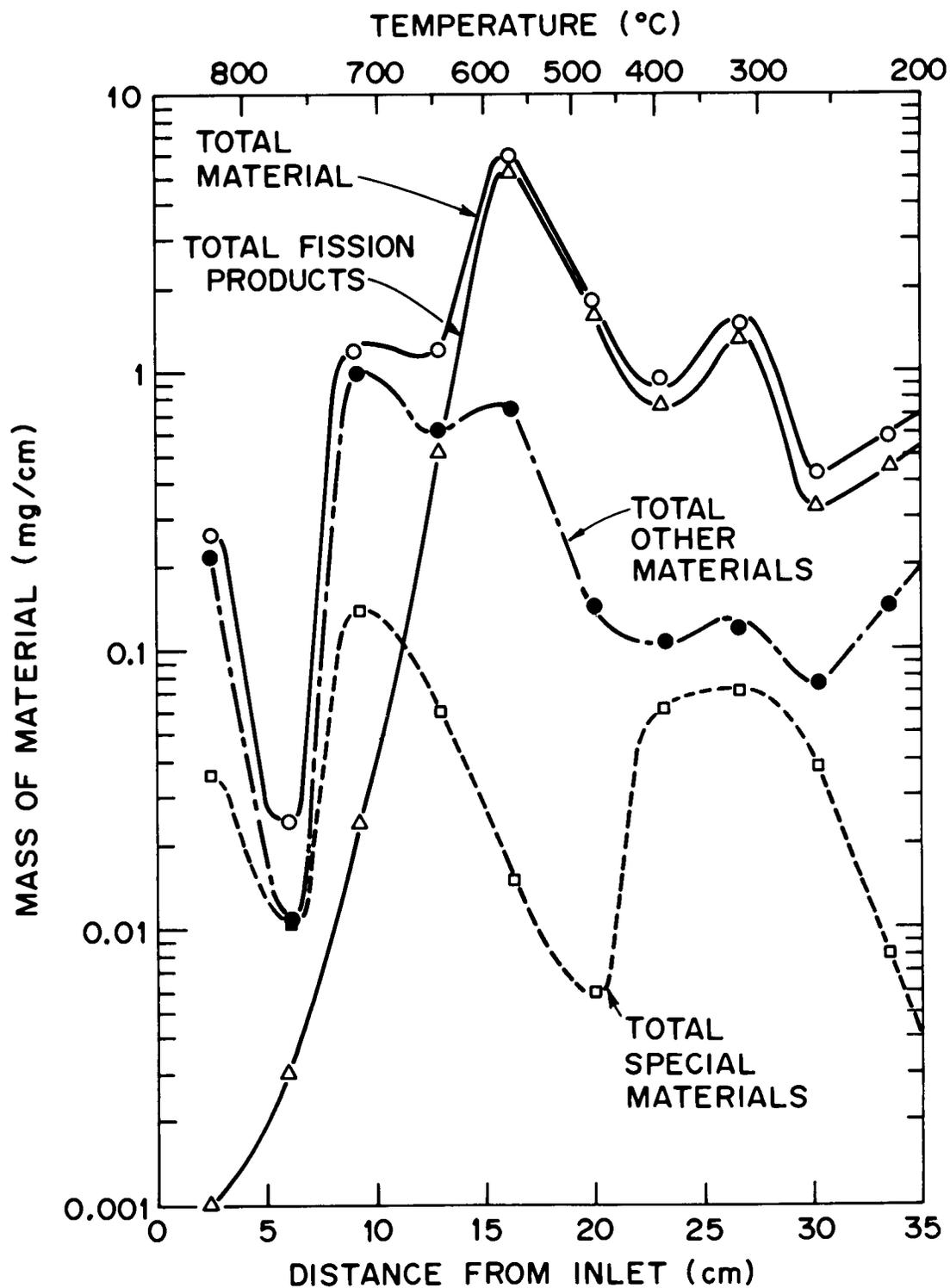


Fig. 10 Distribution of materials deposited in the TGT, as determined by SSMS.

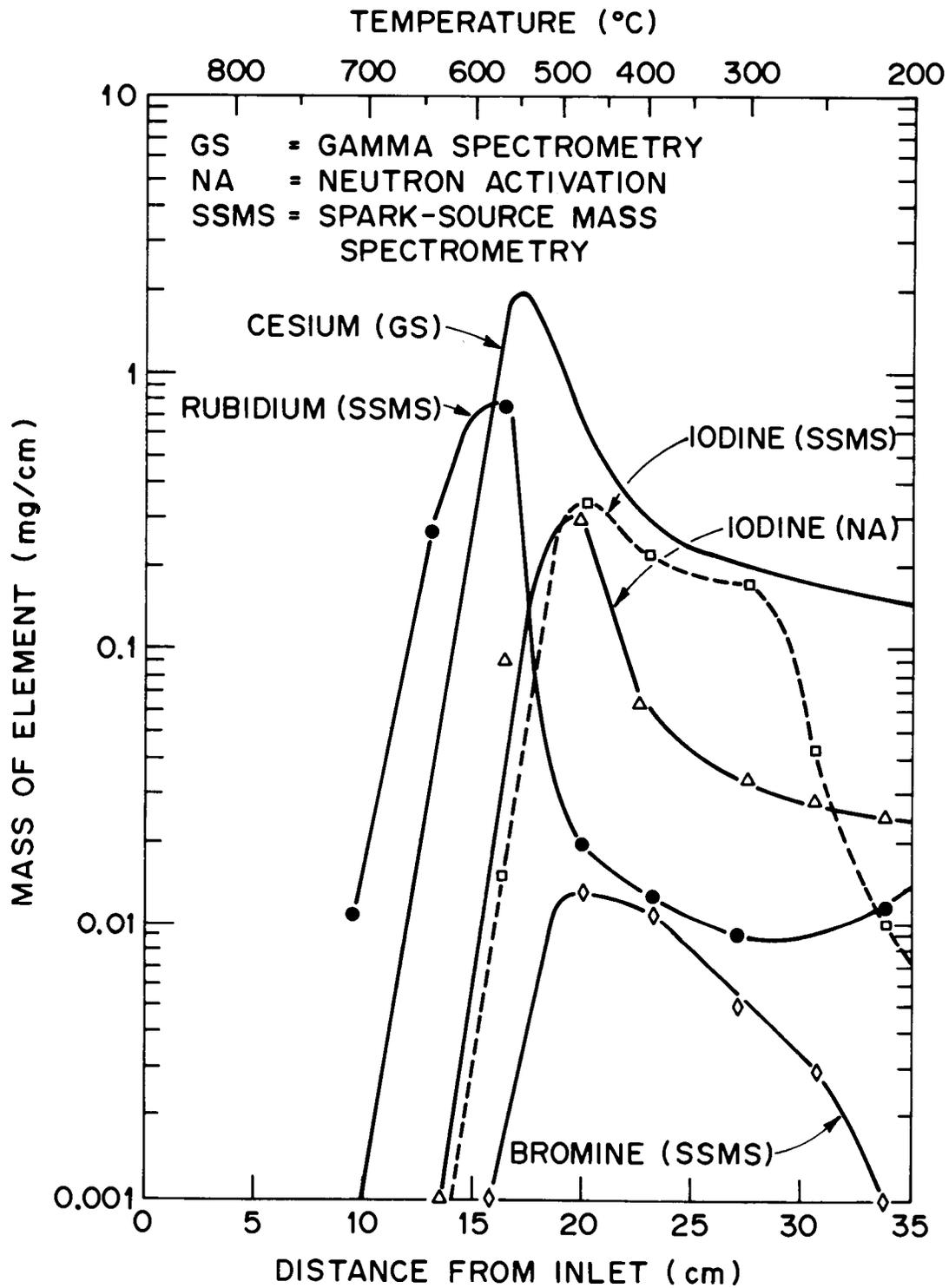


Fig. 11. Distribution of fission product elements found by SSMS in the TGT.

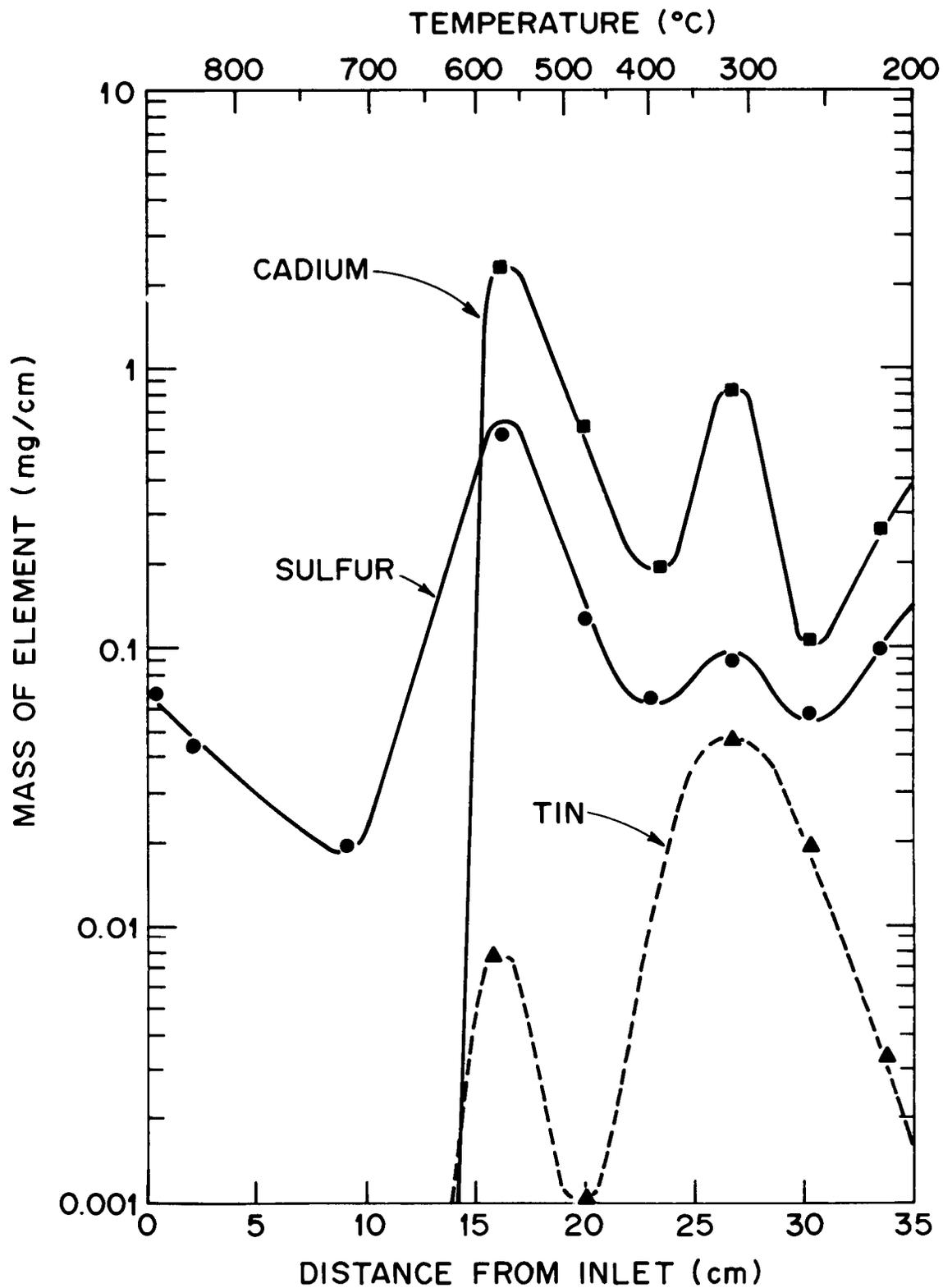


Fig. 12. Distributions of Cd, S, and Sn in the TGT, as determined by SSMS.

activation and the other by SSMS. The profiles for rubidium and bromine were determined by SSMS. Considering that SSMS is only a semiquantitative method of analysis, the profile obtained for I, Br, and Rb appear to be quite good. The deposition peaks are located, relative to cesium, where they were expected.

Table 12 shows the fractions of test specimen inventories of certain fission products found in the TGT and on the glass wool prefilter. Similar percentages of the chemical analogs rubidium and cesium and the chemical analogs bromine and iodine were found. The percent iodine determined by SSMS was only 22% lower than the value determined by neutron activation analysis.

Table 12. Fission product release values based on spark-source mass spectrometry

Fission products	Fraction of specimen inventory found (%) <sup>a</sup>
Cs	12.3 <sup>b</sup>
Rb	13.0
I	11.5 (14.8) <sup>c</sup>
Br	11.0
Cd	100
Ag	0.2 <sup>d</sup>
Te	<0.02
Ba	<0.003

<sup>a</sup>Includes only deposits in the TGT and on the glass wool prefilter.

<sup>b</sup>Cesium value was determined by gamma spectrometry and used as a standard to determine the others.

<sup>c</sup>Value determined by neutron activation.

<sup>d</sup>Gamma counting showed that most of the released Ag deposited on (reacted with) ZrO<sub>2</sub> components at the outlet end of the furnace. None of the furnace components were included in the SSMS analyses.

Profiles for cadmium, sulfur, and tin are shown in Fig. 12. Peak deposits for all three elements occurred at ~575°C as was the case for cesium and rubidium (Fig. 11). Downstream at ~325°C, on the other hand, only the elements Sn, Cd, and S (also some Mg) were present in that peak. Furthermore, the deposit was yellowish in color which suggests the possibility of cadmium and/or tin sulfides, both of which are yellow.

(Magnesium sulfide is reddish brown.) The mechanism of deposition is not certain.

#### 4.2.4 Mass of Material Collected in the Thermal Gradient Tube and on the Filters

As observed in previous tests in this series,<sup>7-9</sup> deposits of significant mass were found in the TGT and on the filters. The filters are shown in Fig. 13. The masses of these deposits from test HI-5 are compared below with those from test HI-4, which operated at similar steam flow conditions.

Test	Temperature (°C)	Mass collected (mg)		Calculated aerosol concentration <sup>a</sup> (g/m <sup>3</sup> )	
		TGT	Filters	At furnace temperature	At 150°C
HI-4	1850	30	70	0.73	3.8
HI-5	1700	40	106	1.20	5.6

<sup>a</sup>Assumes test time plus 5 min (25 min for these tests) for aerosol production time.

Most of the material deposited in the TGT probably resulted from vapor condensation, whereas that on the filters was largely aerosol. The total masses collected were used to calculate the average aerosol and vapor concentration at furnace temperature and at 150°C during the tests. The concentrations at furnace temperature may be analogous to the vapor plus aerosol concentration existing within a reactor core during a severe accident, and those at 150°C might be comparable to conditions within the containment building following an accident. As noted in Sect. 4.2.3, atypical impurity elements (especially sulfur) constitute a significant fraction of the aerosol mass in these tests.

#### 4.2.5 Thermal Gradient Tube Results

The TGT was a quartz tube of 0.4-cm internal diameter, lined with platinum foil to provide an "inert" deposition surface. The platinum formed a tube ~0.38 cm in diameter and 35.6 cm long. This tube cooled the hot gas entering it from the furnace; the heat was rejected by conduction radially in the gas and then through the platinum liner, a small gas gap, and the quartz tube.

The temperature profile along the TGT was measured by 12 thermocouples on the outside of the quartz tube, four of them controlled the

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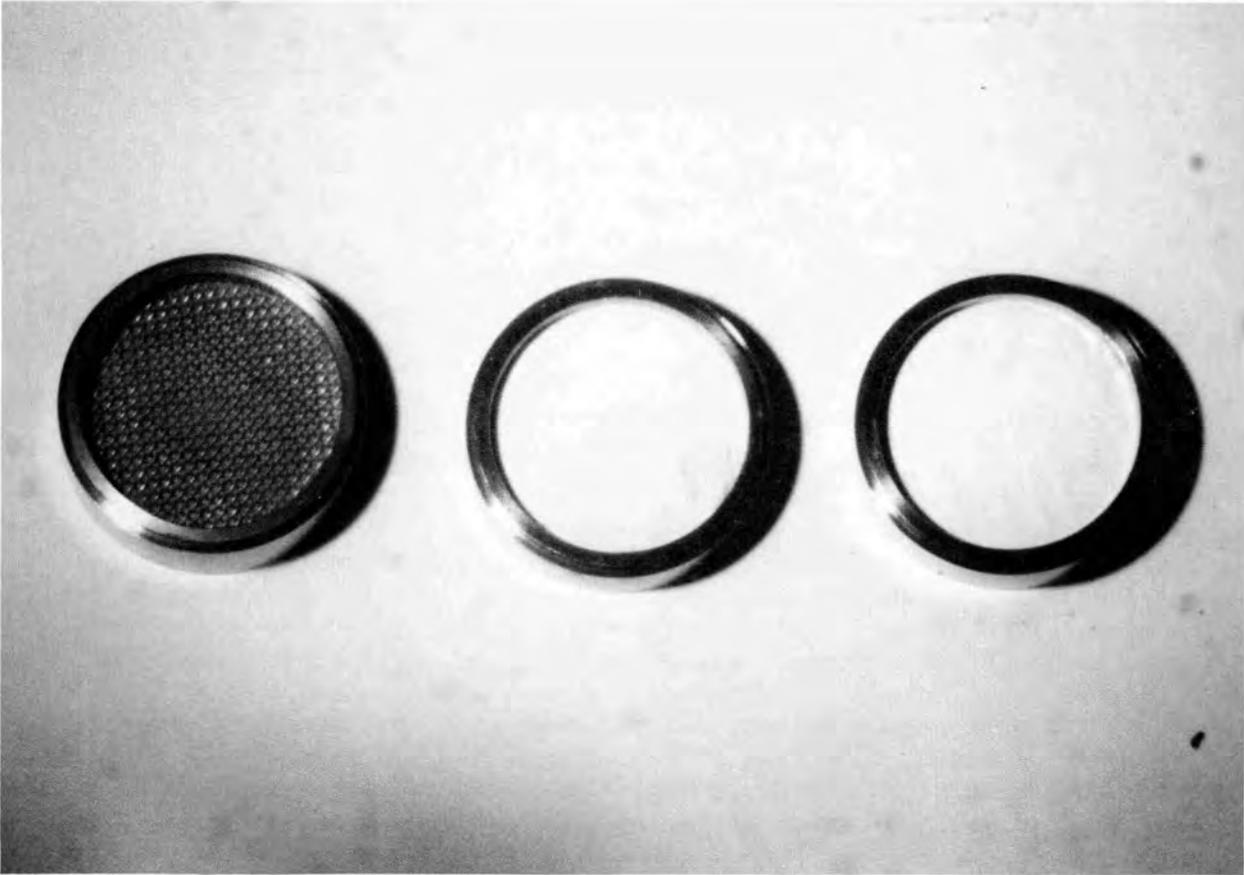


Fig. 13. Filters from test HI-5, with prefilter (covered by screen) on left. Note the absence of visible deposits on the HEPA filters.

four electric heaters that preheated the tube and maintained the profile during the test. Automatic controllers kept the temperature drift below  $\pm 3^\circ\text{C}$  except at the inlet to the tube. The inlet temperatures fluctuated because the temperature and composition of the gas coming from the furnace varied. Figure 14 shows the average profile during the 20 min that the fuel was at test temperature.

Appendix 1 of ref. 8 contains a simple calculation of the radial temperature distribution in the TGT. Applying such methods to the conditions of test HI-5, we calculate:

when thermocouple temperature =  $x^\circ\text{C}$ ,  
 inside surface of the platinum =  $x + 5^\circ\text{C}$ , and  
 center gas temperature =  $x + 13^\circ\text{C}$ .

Table 13 contains Cs, I, Ag, and Sb data from the TGT sections. The cesium profile was also measured by gamma scanning the intact platinum liner. As shown in the comparison in Fig. 15, there was excellent agreement. Spark-source mass spectrometry detected iodine and silver, and Table 14 compares the results with gamma spectrometry, indicating adequate agreement. The SSMS cannot detect antimony at the low levels observed in test HI-5 (0.1  $\mu\text{g}$  in total on the entire TGT).

The cesium profile is plotted against temperature in Fig. 16. The peak occurs in the range of 510 to 600 $^\circ\text{C}$ ; the gamma scan places it more precisely at 570 to 580 $^\circ\text{C}$ . Of the cesium that entered the TGT, 38% deposited and 62% escaped and was trapped downstream, mainly on the glasswool prefilter.

Figure 16 also shows the iodine profile plotted against temperature; it peaks in the 420 to 510 $^\circ\text{C}$  range — lower than the cesium peak. Of the iodine that entered the TGT, 33% deposited and 67% escaped and was trapped on the glasswool prefilter.

Figure 17 shows how the  $^{137}\text{Cs}$  deposits responded to basic and acidic leaching. As observed in the previous four tests in the HI series, two cesium forms are apparent. At TGT temperatures above 460 $^\circ\text{C}$  (0 to 20 cm), 20 to 60% of the cesium was insoluble in  $\text{NH}_4\text{OH}/\text{H}_2\text{O}_2$  mixtures, but below 460 $^\circ\text{C}$  (20 to 35 cm), <0.4% of the cesium was insoluble. As expected,  $^{134}\text{Cs}$  behaved exactly like  $^{137}\text{Cs}$ . The peak on section 4 (9.5 cm) probably was caused by a fuel or cladding particle that dropped off during basic leaching, causing a high apparent decontamination factor.

Figure 18 shows the antimony profile indicated by measurements of the individual TGT sections. Cesium dominated the gamma activity of the sections; only sections 2 and 3 showed any antimony before cesium had been removed by leaching. Data from sections 1 to 4 show that leaching left the antimony untouched. The test HI-5 antimony profile resembles profiles from tests HBU-11, HI-1, and HI-2.<sup>13</sup>

Figure 19 presents the silver profile, which resembles the profile in test HI-2. The leaching data indicate that  $\text{NH}_4\text{OH}/\text{H}_2\text{O}_2$  does not dissolve

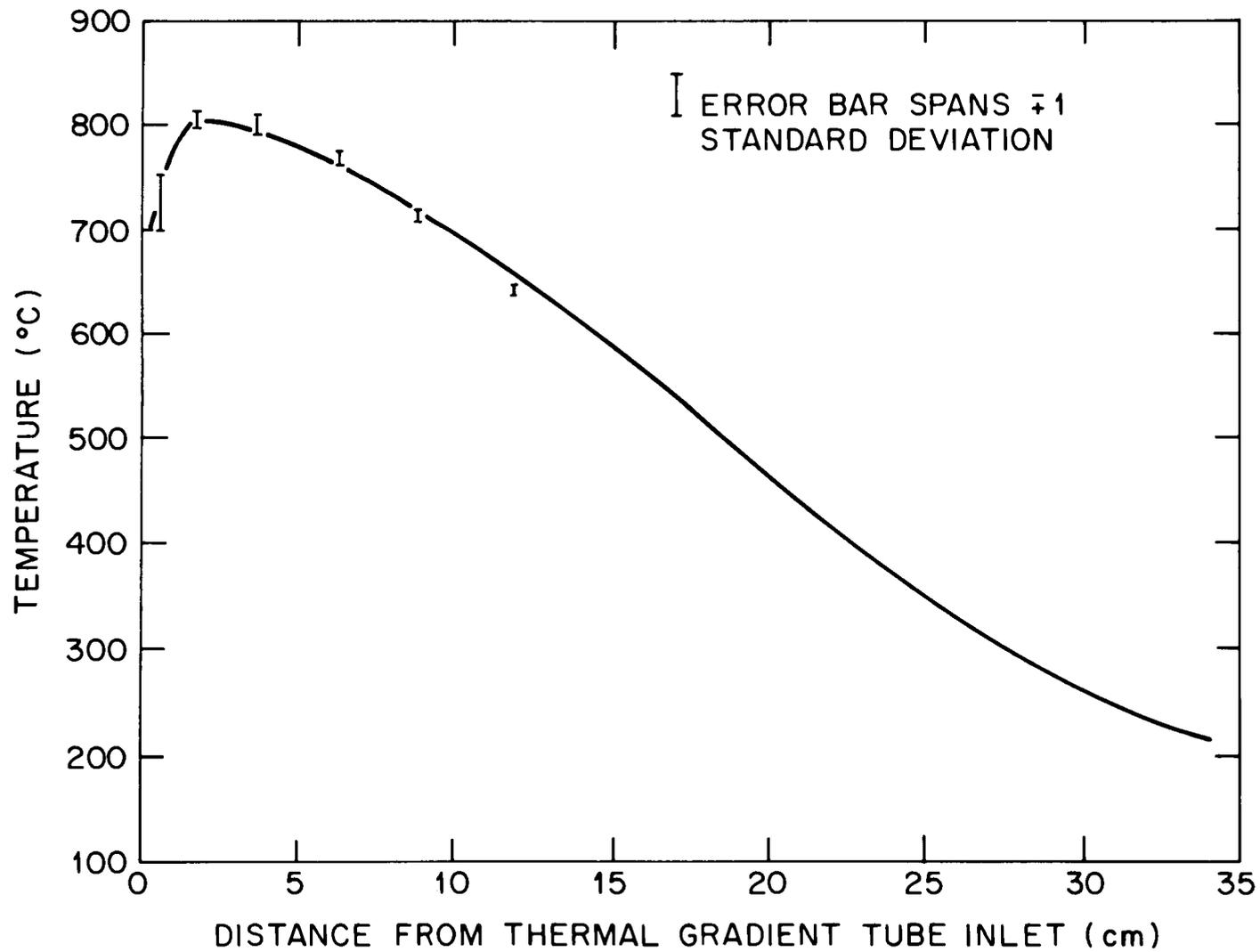


Fig. 14. Temperature distribution along thermal gradient tube in test HI-5.

Table 13. Fission products on the thermal gradient tube in test HI-5

Position (cm)	Temperature range <sup>a</sup> (°C)	<sup>137</sup> Cs <sup>b</sup> (mCi)	Cesium (μg/cm <sup>2</sup> )	<sup>129</sup> I <sup>c</sup> (μg)	Iodine (μg/cm <sup>2</sup> )	<sup>110m</sup> Ag <sup>d</sup> (μCi)	Silver (μg/cm <sup>2</sup> ) <sup>e</sup>	<sup>125</sup> Sb (μCi)	Antimony (μg/cm <sup>3</sup> ) <sup>e</sup>
0-0.6 <sup>f</sup>	705-735	0.074	3.3	0.020	0.049	1.0	0.18 ± 0.07	0.7	3.4 ± 0.6
0.6-4.4	735-810	0.13	0.90	0.060	0.018	5.4	0.16 ± 0.01	9.7	7.8 ± 0.6
4.4-7.9	810-745	0.10	0.75	0.030	0.011	8.2	0.26 ± 0.02	10.0	9 ± 1
7.9-11.5	745-680	1.70	13.0	1.1	0.40	2.2	0.07 ± 0.02	4.1	3.5 ± 0.4
11.5-15.1	680-600	32.0	230	2.1	0.75				
15.1-18.7	600-510	280	2000	250	88				
18.7-22.2	510-420	84.0	630	820	300	4.3	0.13 ± 0.06		
22.2-25.8	420-335	41.0	300	160	57	4.1	0.13 ± 0.01	1.1	0.9 ± 0.3
25.8-29.5	335-275	33.0	230	94	33	2.9	0.09 ± 0.01	0.8	0.7 ± 0.5
29.5-33.2	275-230	24.0	170	75	26	3.7	0.11 ± 0.01	0.8	0.7 ± 0.5
33.2-25.8	230-210	18.0	180	49	24	3.0	0.13 ± 0.01	0.3	0.34 ± 0.2

<sup>a</sup> Estimated temperature at inside surface of platinum TGT.

<sup>b</sup> By gamma spectrometry before leaching.

<sup>c</sup> By neutron activation analysis.

<sup>d</sup> By gamma spectrometry, after leaching had removed >90% of the <sup>134</sup>Cs and <sup>137</sup>Cs.

<sup>e</sup> Assuming 1.018 cm<sup>2</sup> surface area per cm length of TGT.

<sup>f</sup> Measured lengths, corrected for wrinkling during cutting of platinum tube.

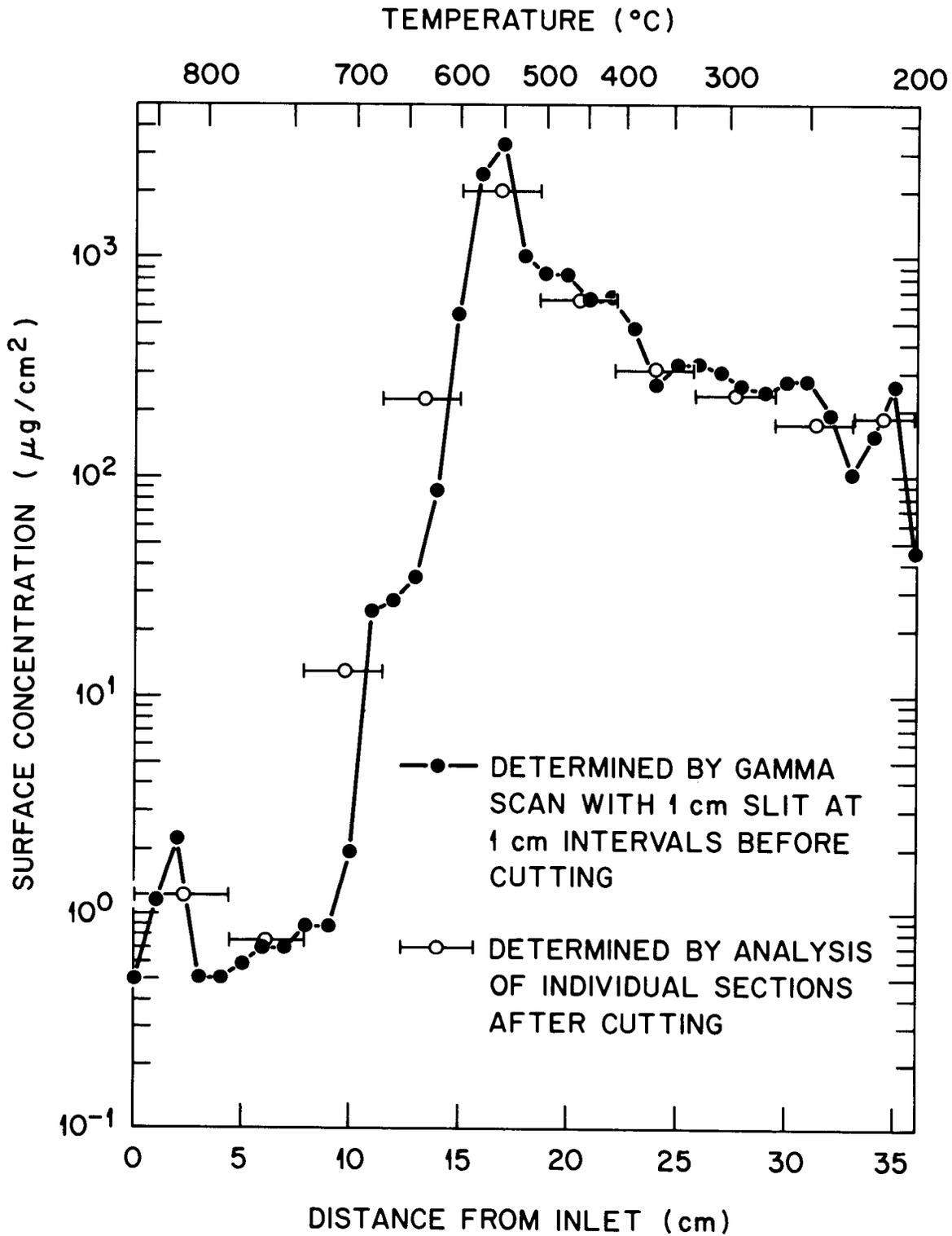


Fig. 15. Cesium profile on test HI-5 thermal gradient tube before and after cutting into sections.

Table 14. Iodine and silver in thermal gradient tube of test HI-5; comparison of results from different analytical methods

Thermal gradient tube section	Iodine ( $\mu\text{g}/\text{cm}^2$ )		Silver ( $\mu\text{g}/\text{cm}^2$ )	
	NAA <sup>a</sup>	SSMS <sup>b</sup>	GS <sup>c</sup>	SSMS
1	0.049	ND <sup>d</sup>	0.18	ND
2	0.018	ND	0.16	ND
3	0.011	ND	0.26	ND
4	0.40	ND	0.070	ND
5	0.75	ND	ND	ND
6	88	15.0	ND	ND
7	300	320	0.13	ND
8	57	220	0.13	0.20
9	33	170	0.090	4.0
10	26	44	0.11	0.7
11	24	10	0.13	ND
Filter <sup>e</sup>	(3900)	(120)	(15)	ND

<sup>a</sup>NAA = neutron activation analysis.

<sup>b</sup>SSMS = spark-source mass spectrometry.

<sup>c</sup>GS = gamma spectrometry.

<sup>d</sup>ND = not detected.

<sup>e</sup>Values for filter are  $\mu\text{g}$  total, not  $\mu\text{g}/\text{cm}^2$ .

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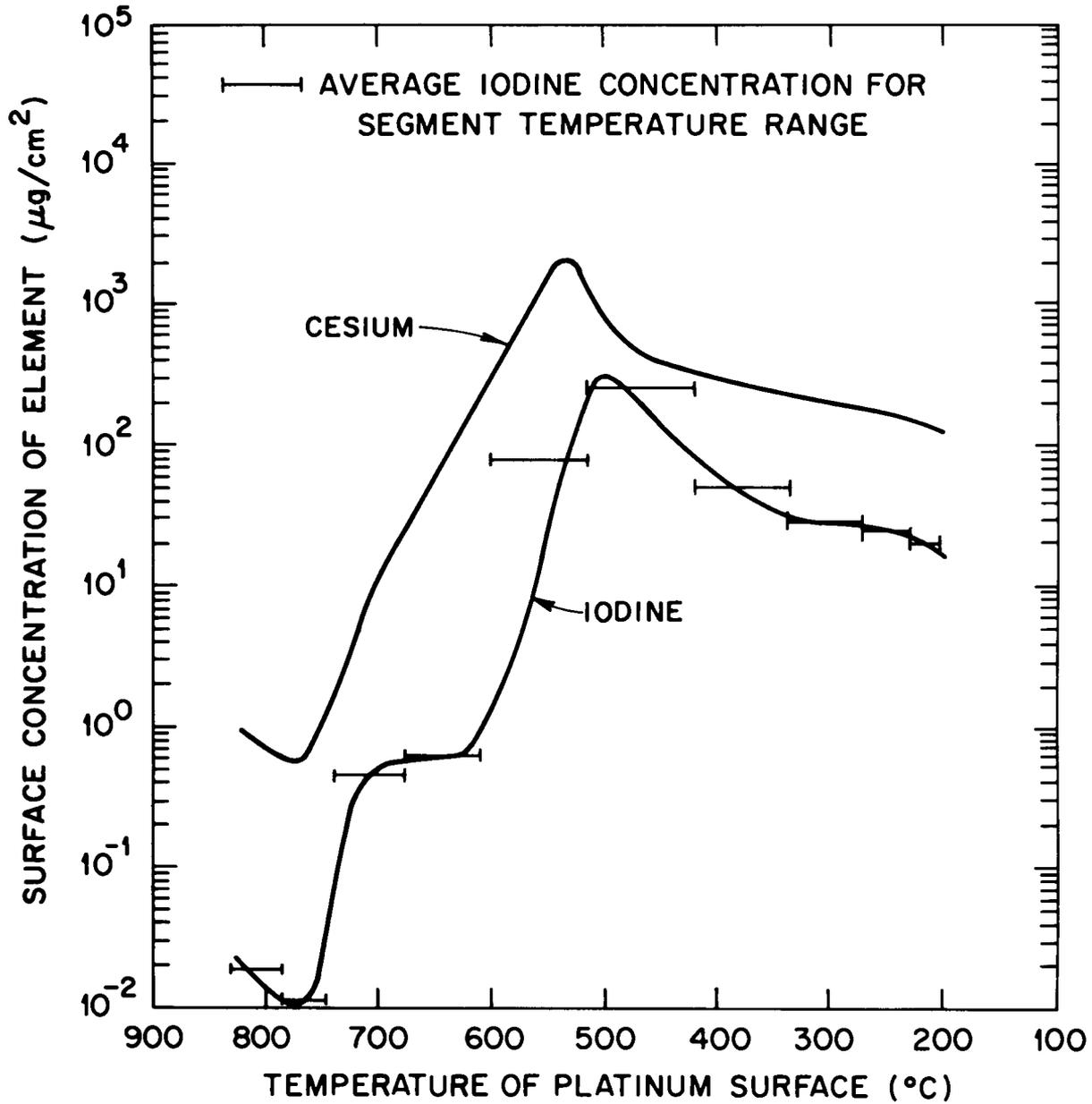


Fig. 16. Cesium and iodine profiles on the thermal gradient tube in test HI-5.

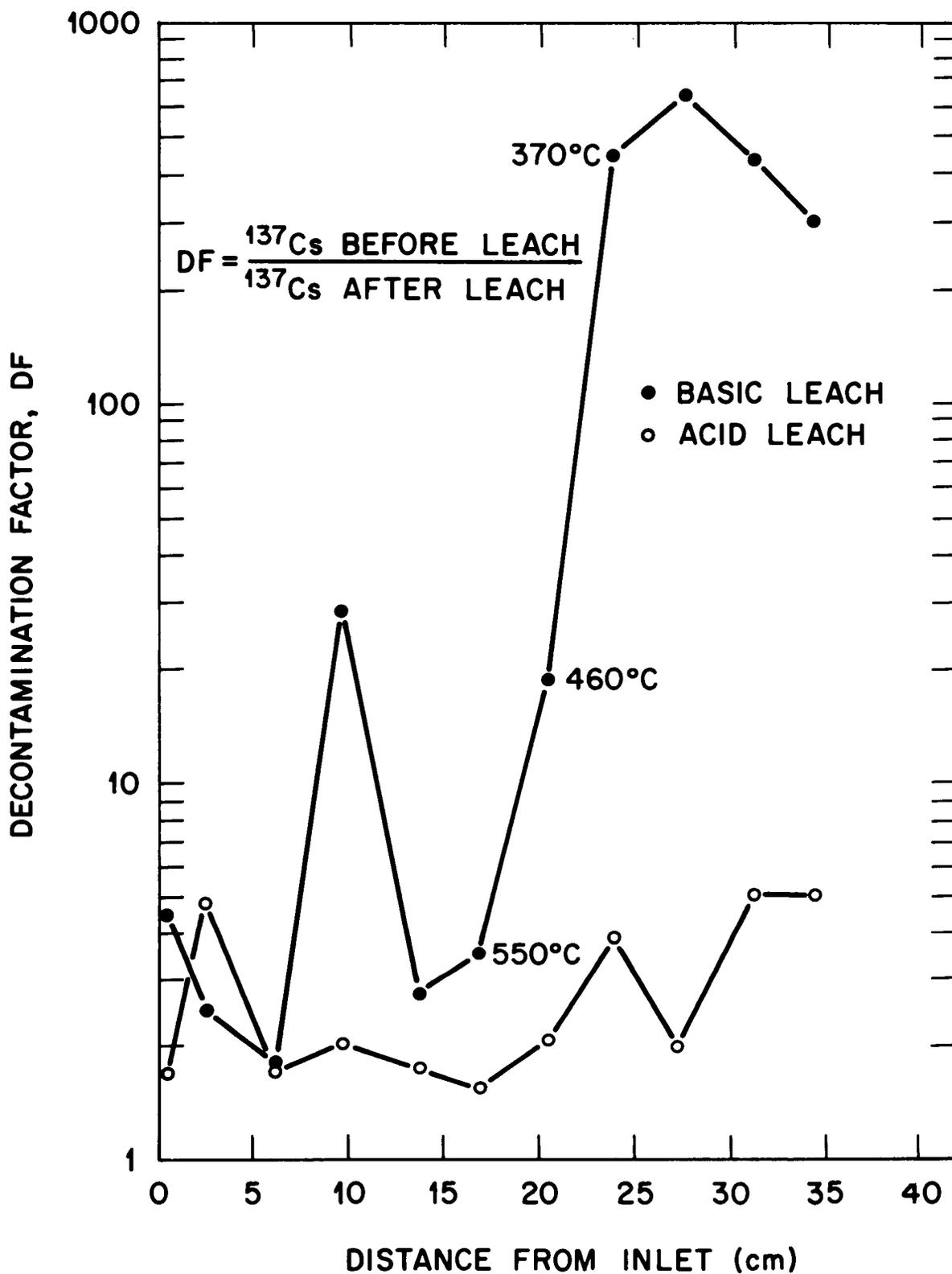


Fig. 17. Solubility of  $^{137}\text{Cs}$  deposited in the TGT of test HI-5 when exposed to basic and acidic leaches.

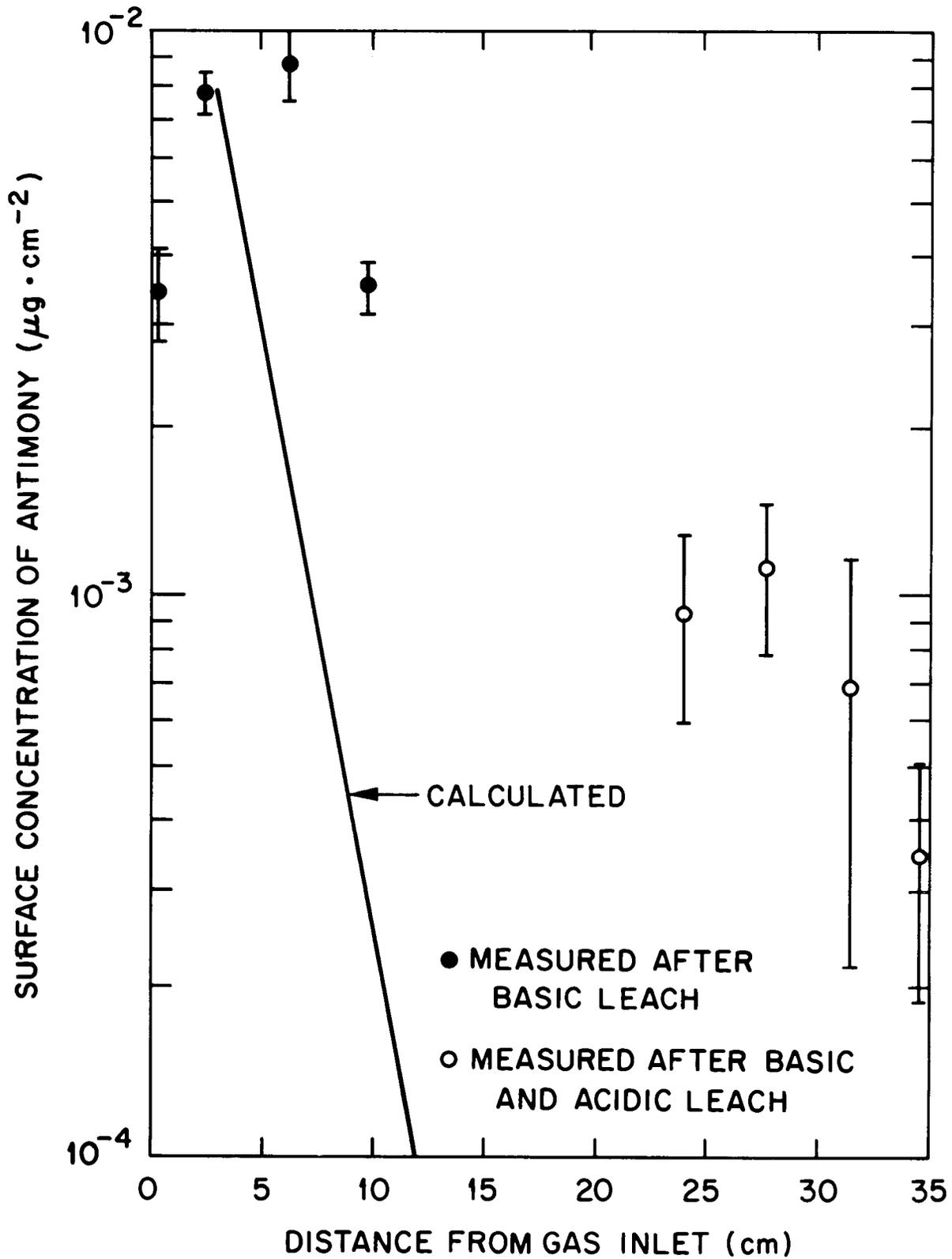


Fig. 18. Distribution of antimony in the TGT in test HI-5.

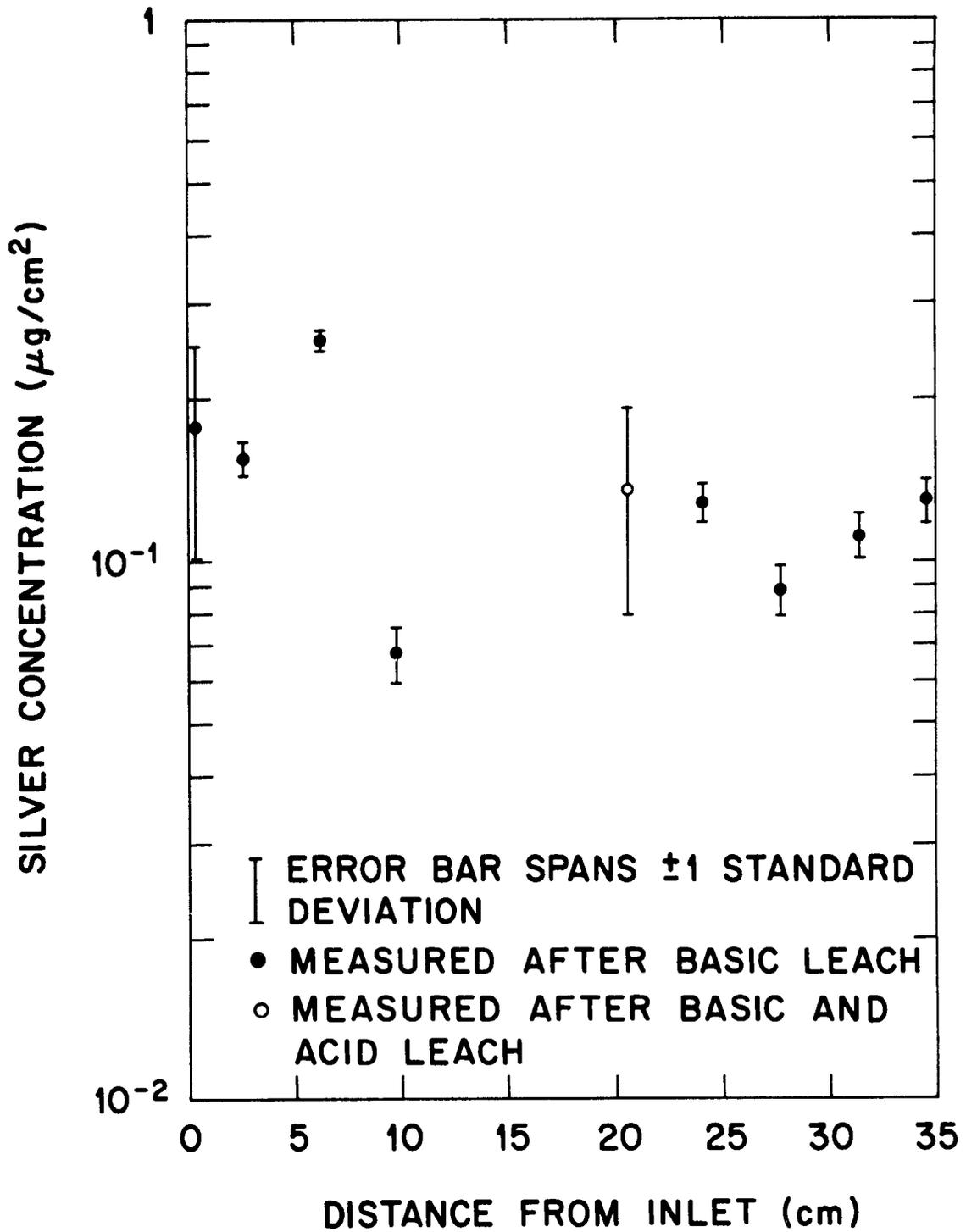


Fig. 19. Distribution of silver in the TGT of test HI-5.

silver (but basic leach data are available only for the sections of the TGT above 740°C). The acidic leach (4 M HNO<sub>3</sub>/0.25 M HF) removed 20% of the silver deposited at 700°C, and at a deposition temperature of 200°C or less, ~100% is removed.

Small amounts of <sup>106</sup>Ru and <sup>154</sup>Eu were detected when the intact platinum liner of the TGT was scanned with a Ge(Li) detector. The <sup>106</sup>Ru profile was flat and very close to background. The <sup>154</sup>Eu profile rose to six times background at 20 to 30 cm from the gas inlet, equivalent to ~0.05 µg Eu/cm<sup>2</sup>, and then decreased to background farther along the tube. However, SSMS detected no ruthenium or europium on the TGT.

#### 4.2.5.1 Behavior of cesium and iodine

During passage through the TGT, the concentrations of cesium- and iodine-containing vapors decrease. Two mechanisms could cause such a decrease in vapor concentration: deposition onto the platinum wall or condensation to form aerosols in the gas. As seen in Fig. 16, both the cesium and iodine profiles peaked near midlength and then declined toward the outlet end of the TGT. Table 15 compares data from the first five tests in the HI series. The third column in this table, iodine profile gradient, contains values which indicate the slope, or the rate of decreases, in the vapor concentration for iodine species. Gas phase diffusion should limit the rate of deposition; if deposition was the dominant mechanism (as opposed to condensation to form aerosol), the values in the last column of Table 15,

$$\frac{\text{Gradient} \times \text{flow}}{D},$$

would be constant. Since these values vary from 0.2 to 0.6, it appears that deposition was not dominant. Another characteristic of a diffusion-limited process is that the gaseous and surface concentrations should drop in step with each other. In test HI-5, the gaseous concentration (including particulate-associated iodine) dropped by a factor of only 1.2, whereas the surface concentration dropped by a factor of 15. Thus, we conclude that condensation of iodine-containing species onto aerosols (or chemical reaction with aerosols) is an important process in these tests. These results corroborate the large amount of iodine (66% of that released from the fuel, as shown in Tables 7 and 10) found on the filters.

During test HI-5, at least 0.71 mol of mixed helium, steam, hydrogen, and CO passed down the TGT while the fuel specimen was at maximum temperature. (Total gas flow was increased by the hydrogen and carbon monoxide produced during limited oxidation of the graphite susceptor by steam.) According to the TGT and filter deposition data, the gas carried  $2.7 \times 10^{-4}$  mol Cs atoms and  $4.5 \times 10^{-5}$  mol I atoms; if CsOH and CsI were the major species, the average partial pressures were:

$$\begin{aligned} &6.4 \times 10^{-5} \text{ bar CsI,} \\ &3.1 \times 10^{-4} \text{ bar CsOH.} \end{aligned}$$

Table 15. Factors affecting the surface concentration gradient of the downstream side of iodine peaks in the thermal gradient tube of test HI-5

Test	Gas flow rate <sup>a</sup> (L/min)	Iodine profile gradient (decades/cm)	Gradient × flow	D <sub>CsI</sub> <sup>b</sup> (cm <sup>2</sup> /s)	Gradient × flow <sup>c</sup> D
HI-1	2.1	0.05	0.11	0.2	0.6
HI-2	1.9	0.04	0.08	0.2	0.3
HI-3	1.0	0.20	0.20	0.5	0.4
HI-4	1.1	0.14	0.15	0.8	0.2
HI-5	1.2	0.07	0.08	0.4 <sub>to</sub> 0.7 <sup>d</sup>	0.1 to 0.2

<sup>a</sup> At 150°C.

<sup>b</sup> Calculated diffusion coefficient of CsI in gas at 150°C; variation occurred because of changing mixture of gases (Ar or He, H<sub>2</sub>O, H<sub>2</sub>, and CO) during the tests.

<sup>c</sup> Calculated using more significant figures than shown in this table.

If susceptor oxidation produced significant amount of CO, the lower values apply.

On-line gamma spectrometry of the TGT showed that <sup>137</sup>Cs deposited at a constant rate, indicating that the partial pressure of cesium compounds was also constant.

The equilibrium vapor pressure of CsI over pure CsI depends on temperature; two expressions were found in the literature:

$$\log_{10} P = 6.243 - 9699/T \text{ (ref. 14),} \quad (1)$$

$$\log_{10} P = 17.47 - 3.52 \log_{10} T - 9678/T \text{ (ref. 15),} \quad (2)$$

with P in bar and T in K. Under the conditions of our test, these two equations may be used to calculate that CsI should begin to deposit in the range 580 to 660°C. This result is in reasonable agreement with the iodine profile in Fig. 16. Therefore, iodine behavior is consistent with its release as CsI, which condenses on the platinum and also collects as an aerosol component on the filters downstream.

Pure CsOH is more volatile than pure CsI. Based on the conditions of test HI-5, and on two expressions for CsOH vapor pressure,

$$\log_{10} P = 5.316 - \frac{6651}{T} \quad (\text{ref. 16}), \text{ and} \quad (3)$$

$$\log_{10} P = 4.752 \pm 0.22 - \frac{6611 \pm 397}{T} \quad (\text{ref. 17}), \quad (4)$$

with P in bar and T in K, we calculated that CsOH would not deposit in the TGT until the platinum temperature fell to the range 440 to 630°C, depending on the concentration. Consequently, the large fraction of the cesium deposited above this temperature range must be combined in another compound. As in test HI-4, SSMS detected more than enough sulfur to form such compounds as Cs<sub>2</sub>S, Cs<sub>2</sub>SO<sub>3</sub>, and Cs<sub>2</sub>SO<sub>4</sub>. Therefore, cesium deposition in both tests could have been strongly influenced by sulfur, an impurity previously found to be released from the furnace ceramics. Conversely, there is no indication that the behavior of iodine was affected by extraneous materials.

#### 4.2.5.2 Behavior of antimony

The relevant data for antimony behavior in the TGT of four fission product release tests are presented in Table 16.

Table 16. Factors affecting the gradient of the antimony profile in the thermal gradient tubes of fission product release tests

Test	Gas flow (L/min at STP)	Antimony profile gradient (decades/cm)	Gradient × flow	$D_{Sb_2}^a$ (cm <sup>2</sup> /s)	$\frac{\text{Gradient} \times \text{flow}}{D}$
HBU-11	1.67	0.083	0.138	2.09	0.066
HI-1	1.44	0.100	0.144	1.73	0.083
HI-2	1.33	0.101	0.135	2.18	0.062
HI-5	0.80	0.042	0.034	3.76 to 5.35	0.006 <sub>a</sub> to 0.009 <sub>b</sub>

<sup>a</sup>Calculated diffusion coefficient of Sb<sub>2</sub> in gas at 1000°C.

<sup>b</sup>If susceptor oxidation produced significant amount of CO, the lower values apply.

The antimony profile for test HI-5 (Fig. 18) is similar in shape to the profiles for the previous tests HBU-11, HI-1, and HI-2; it differs, however, in two aspects. Firstly, the amount of antimony was 2 to 10 times less than in test HI-1, and secondly, the profile is relatively flat, as indicated by the profile gradient in Table 16.

The reason for the relatively flat profile is unclear. Low levels of antimony are difficult to measure, but this is unlikely to cause an apparently flat profile. We considered the possibility that the antimony might have been present as a species with a low diffusion coefficient, such as an aerosol. The very small amount of  $^{125}\text{Sb}$  found on the filter, however, indicates that very little antimony existed in the particulate form (see Table 9), thereby negating this possibility.

#### 4.2.5.3 Behavior of silver

As shown in Fig. 19, the silver profile is relatively flat, indicating that silver was present as a species with very low gas diffusivity, such as aerosol particles. The inertness of the deposit to the basic leach solution indicates that the silver was not present as  $\text{AgCl}$ , which is soluble in ammoniacal solutions. However, the deposit dissolved slightly in  $\text{HNO}_3/\text{HF}$ , which indicates that metallic silver (but not  $\text{Ag}_2\text{S}$ ) may have been present.

The behavior of silver in this test was similar to that observed previously in test HI-2.<sup>7</sup> The flat profile suggests that it was released as the element but quickly condensed to a metallic aerosol or deposited onto other aerosol particles. Note that four times as much silver was carried to the filter as condensed (or otherwise deposited) in the TGT, as shown in Tables 7 and 9.

#### 4.2.6 Results from Analysis of Aerosol Sampler

Although the initial plans for these tests<sup>1</sup> had included the use of a miniature cascade impactor for the classification and analysis of aerosols, subsequent studies indicated that such a tool would be impractical in these tests, especially at higher temperatures where aerosol generation would be very heavy. As an alternative, an entirely different type of aerosol sampler was developed and used for the first time in test HI-5. This instrument, described previously,<sup>18</sup> uses a screw-driven graphite rod to collect deposits on a spiral path as it crosses a 1.3-mm gap in the gas/aerosol stream between the TGT and the filter package (see Fig. 20). After the test, the graphite rod (0.8 mm in diameter) 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 fission product cesium and iodine were collected on the sampler and its housing: 0.25% and 0.45%, respectively of the  $^{137}\text{Cs}$  released from the fuel specimen. The distribution of  $^{137}\text{Cs}$  along the sampler rod, shown in Fig. 21, indicates a gradual increase in  $^{137}\text{Cs}$  deposition during the test. The peak beyond the end of the test (and beyond the end of rod movement) cannot represent test behavior; it probably resulted from pickup of deposits from the gap and/or the housing during disassembly. The locations of specific particles and general areas examined by SEM/EDX are shown at the top of Fig. 21, and the elemental fractions are listed in Table 17. As shown, cesium was present at all locations, both in specific particles and as background, and the

ORNL DWG 84-613

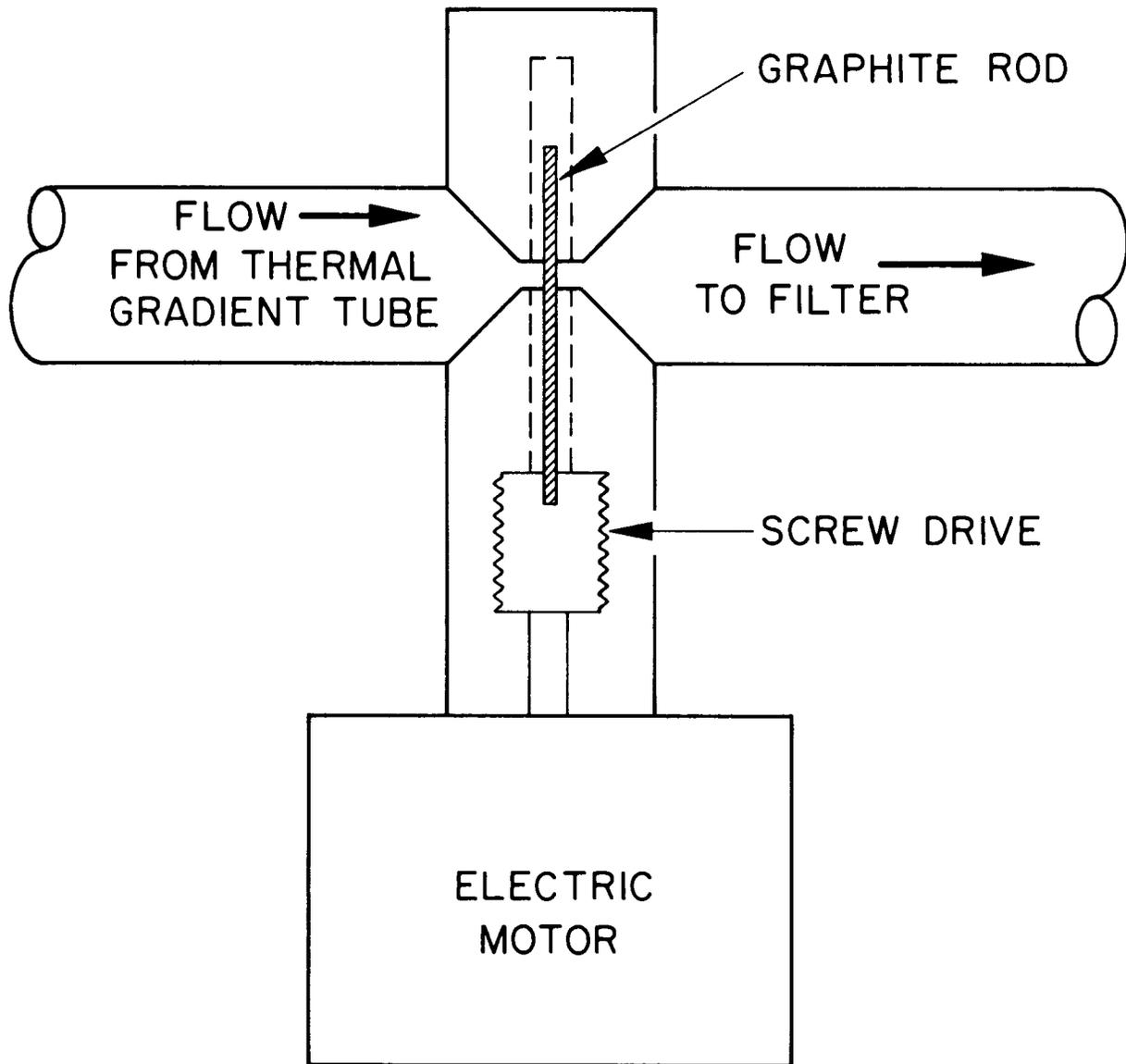


Fig. 20. Miniature aerosol deposition sampler.

ORNL DWG 84-1025

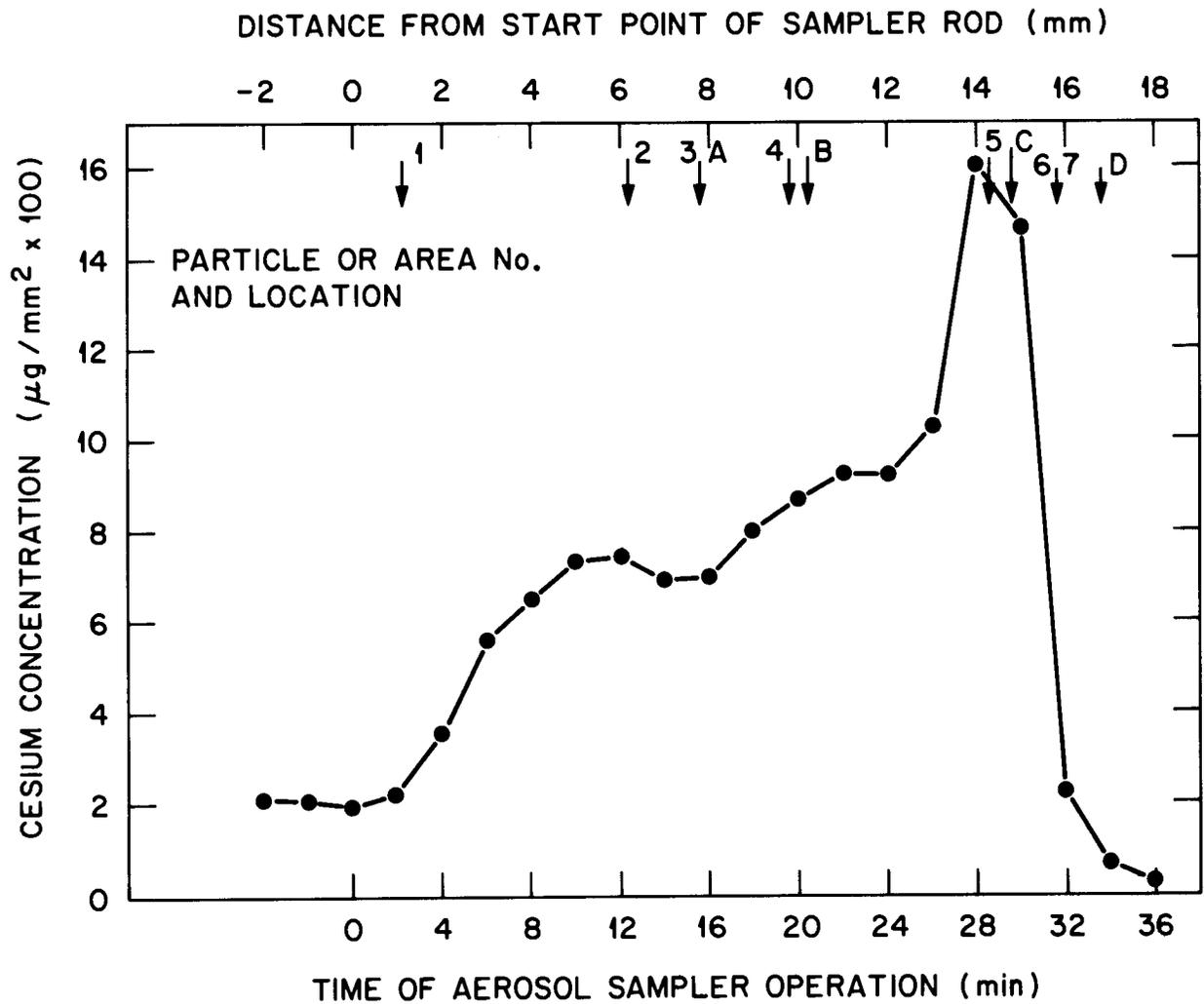


Fig. 21. Distribution of cesium and locations of SEM/EDX analyses along deposition sampler.

Table 17. Aerosol sampler data<sup>a</sup> for test HI-5

Particle No.	Distance from start of movement <sup>b</sup> (mm)	Approximate size or range (μm)	Elemental fraction (% of total)									
			Cs <sup>c</sup>	I	Cd	Zr	Fe	Si	S	Cu	K	Zn
1	1.1	30 × 30	20	12	0	0	Trace	30	27	11	0	0
2	6.2	15 × 15	25	14	0	0	6.0	28	27	0	0	0
3	7.8	3 × 11	35	0	0	51	14.0	0	0	0	0	0
4	9.8	29 × 29	37	21	0	0	Trace	20	18	5.1	0	0
5	14.3	65 × 102	24	0	31	0	0	18	20	0	0	7.2
6	15.8	65 × 97	21	0	0	0	0	34	30	0	14	0
7	15.8	34 × 49	21	0	0	0	0	34	30	0	14	0
General view												
A	7.8	1 to 13	37	0	0	0	8.1	30	25	0	0	0
B	10.2	1 to 45	38	0	0	0	0	16	30	16	0	0
C	14.8	2 to 6	42	0	0	0	0	29	30	0	0	0
D	16.8	1 to 13	38	0	0	0	11	24	27	0	0	0

<sup>a</sup>Obtained from SEM/EDX analysis.

<sup>b</sup>Sampler movement was started at ~1600°C, 2 min before test temperature was attained, and continued for 27 min, until 5 min after start of cooldown.

<sup>c</sup>Because cesium (a principal fission product) was favored at all locations, the other elements were compared to it for evaluation.

impurities silicon and sulfur were found in most places. Many small individual particles and scattered agglomerates 30 to 100  $\mu\text{m}$  in diameter were examined. Many of the particles had a snowflake-like appearance and contained both cesium and iodine. Although the cesium content was invariably greater than the iodine content (Table 17), the presence of CsI primary particles in a general background of cesium is suggested by the Cs/I ratios of particles 1, 2, and 4. The only additional fission product element found was cadmium, in particle 5 (Table 17); the zirconium found in particle 3 could have originated from fission but probably came from the Zircaloy cladding or the  $\text{ZrO}_2$  furnace ceramics. The source of impurities silicon and sulfur, which have frequently been found by SSMS, very likely was the  $\text{ZrO}_2$  ceramics in the furnace. The other, less frequent impurities, listed in Table 17, probably originated from the furnace ceramics also, and, consequently, have little significance in these tests.

#### 4.2.7 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 ( $^{106}\text{Ru}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$ , and  $^{154}\text{Eu}$ ); 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 of the entire specimen through 3.81- and 6.35-cm thicknesses of lead were made, and a scan using the 0.4-cm window with measurements at 1-cm intervals produced a detailed profile of fission product distribution.

These data were used to determine cesium release from the fuel by direct comparison of  $^{134}\text{Cs}$  (1365 keV) and  $^{154}\text{Eu}$  (1274 keV) values. Europium-154 was chosen because it was not released during the test and because it has a high-energy gamma ray similar to that of  $^{134}\text{Cs}$ . Thermodynamic calculations suggest that europium is probably present in LWR fuel as  $\text{Eu}_2\text{O}_3$ .<sup>19</sup> Because of its low volatility, little migration of europium would have occurred during the test.

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

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

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

$$F = \frac{\text{Pretest } ^{154}\text{Eu} (1274 \text{ keV})}{\text{Posttest } ^{154}\text{Eu} (1274 \text{ keV})} = \frac{0.5098 \text{ Ci}}{0.4818 \text{ Ci}} = 1.058$$

- %  $^{134}\text{Cs}$  released = % cesium released

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

$$= \frac{[(9.204 \text{ Ci}) - (6.567 \text{ Ci}) (1.058)]}{(9.204 \text{ Ci})} \times 100 = 24.5\%$$

The percentage obtained by counting the individual apparatus components and relating them to the ORIGEN inventory was 20.3% (see Table 8).

Table 18 lists the percentages of  $^{134}\text{Cs}$  released as determined by both methods for tests HI-3, -4, and -5. The values obtained by method II agreed fairly well with those obtained by method I. The values obtained by method II were only 7 and 13% higher in tests HI-3 and -4 but 21% higher in test HI-5.

Table 18. Comparison of percentage cesium released as determined by two methods

Test	Cesium released (%)	
	Method I <sup>a</sup>	Method II <sup>b</sup>
HI-1	1.75	<i>c</i>
HI-2	50.5	<i>c</i>
HI-3	58.8	60.2
HI-4	31.7	35.8
HI-5	20.3	24.5

<sup>a</sup>Based on gamma-ray counting of apparatus components and ORIGEN-determined segment inventories.

<sup>b</sup>Determined by gamma-ray counting of fuel rod segments before and after tests.

<sup>c</sup>Fuel rod segment was not counted.

The fuel rod segment used in test HI-5 was also scanned before and after the test. Figure 22 shows the pretest and posttest scan profiles of the  $^{134}\text{Cs}$  (1365 keV)/ $^{154}\text{Eu}$  (1274 keV) ratios. Only three locations were measured before testing; counts per centimeter of fuel were taken after the test. The average percentage release for the posttest points was  $25.7 \pm 5\%$ , with the higher values being at the inlet end and the lower ones at the outlet end. These values compare well with the whole rod measurement of 24.5%, which is believed to be more accurate. Considering that there was a temperature gradient along the fuel specimen during the test ( $\sim 75^\circ\text{C}$  cooler at the outlet end), the percentage release gradient indicated by Fig. 22 is reasonable.

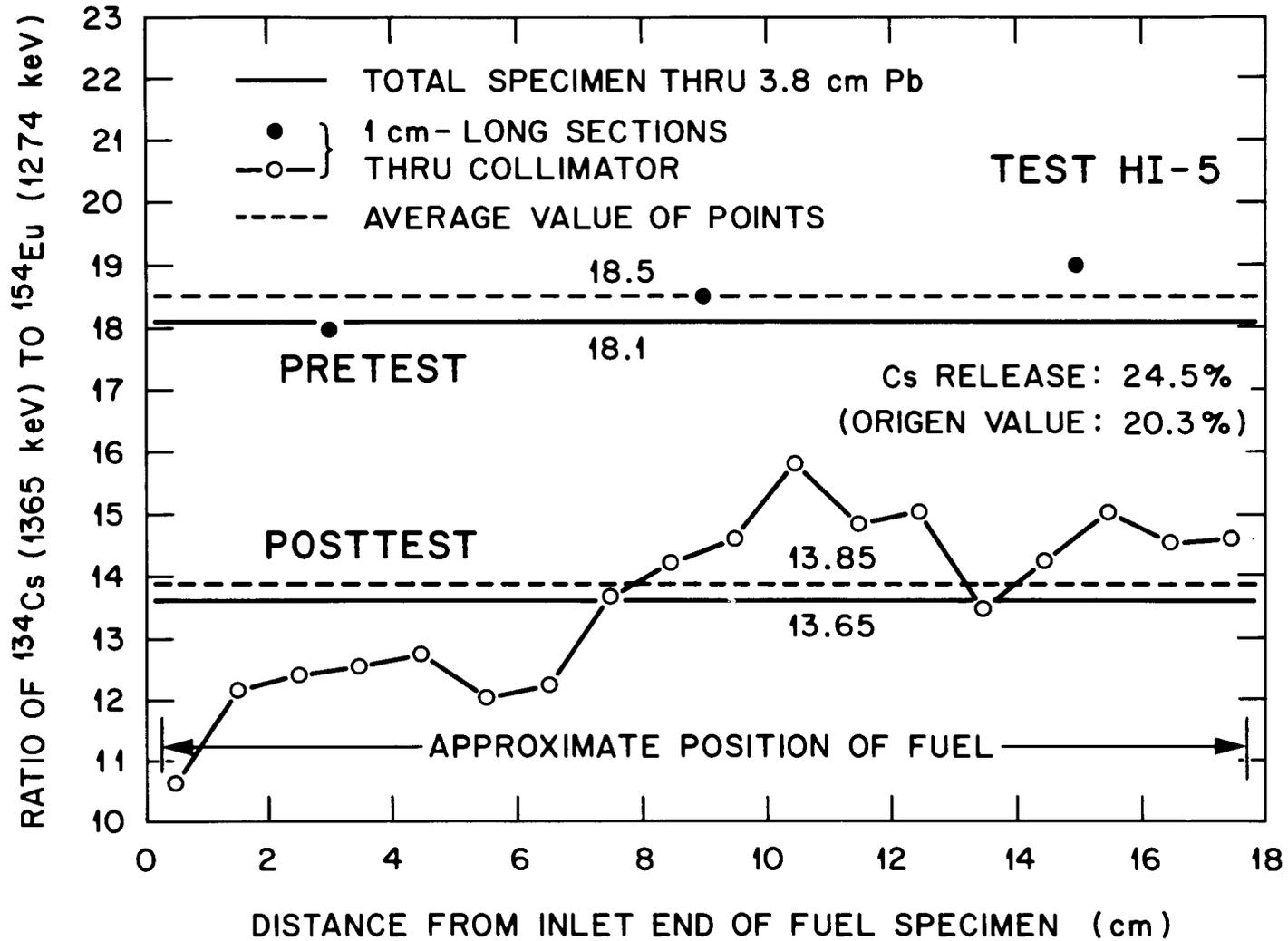


Fig. 22. Comparison of pretest and posttest distribution of  $^{134}\text{Cs}$  in test HI-5 fuel specimen.

#### 4.2.8 Fuel Examination

Following the test and removal of the filter package and TGT from the hot cell, the fuel specimen was removed from the furnace. As illustrated in Fig. 7, a visual examination showed that the inlet end was heavily oxidized, the midlength region had collapsed into rubble, and the outlet end remained relatively intact. In spite of very careful handling, some additional fracturing of the oxidized cladding occurred, indicating its very weak and brittle condition. The specimen was cast in epoxy resin and then cut to 2.5-cm intervals to permit more detailed inspection of the cross sections. These sections, after only moderate grinding for surface preparation, are shown in Fig. 23. Several of these sections were transferred to Argonne National Laboratory, where detailed microstructural and microchemical analyses will be conducted.

#### 5. COMPARISON OF RELEASED DATA WITH PREVIOUS RESULTS

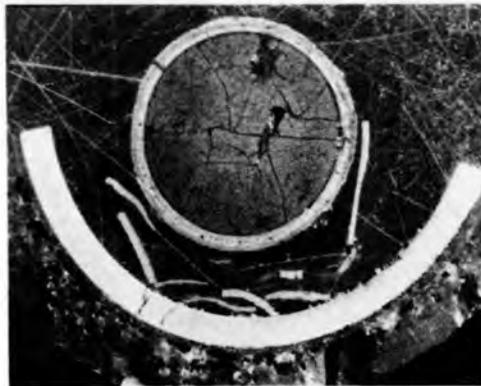
The release data from test HI-5 were compared with the data from the other HI tests and also with the results of a comprehensive review of all relevant fission product release data,<sup>20</sup> which was conducted by the USNRC in 1981. With the exception of lower steam flow rate and incomplete cladding oxidation in test HI-5, experimental conditions were similar to those in test HI-2:  $\sim 1700^{\circ}\text{C}$  for 20 min.<sup>7</sup> The total releases of Kr, I, and Cs, however, were significantly higher in the earlier test,  $\sim 50\%$  in HI-2 vs  $\sim 20\%$  in HI-5. A large longitudinal cladding fracture developed during the HI-2 test (probably during the high-temperature phase of the test period), exposing the  $\text{UO}_2$  to the flowing steam, and perhaps resulting in sufficient oxidation of the  $\text{UO}_2$  to enhance fission product release.<sup>21</sup> Although the fuel specimen in test HI-5 was extensively fractured after the test (Fig. 7), we suspect most of this damage occurred during cooldown, and that the  $\text{UO}_2$  was not extensively exposed to steam during the maximum temperature period.

The release rate data for the five HI tests are compared in with the curves from NUREG-0772<sup>20</sup> in Fig. 24. In four tests, all except HI-2, the Kr, I, and Cs data fall below the corresponding curve. With the exception of test HI-2, these data indicate that the curves (Fig. 24) for Kr, I, and Cs are conservative. In addition, the same data, except for test HI-2, correlate well with the expected straight-line dependency when plotted vs inverse temperature in Arrhenius fashion.<sup>22</sup> Although some data for Ag, Sb, and Te have been collected during these tests, they were omitted from Fig. 24 because of uncertainties and incompleteness. However, the available data for Ag, Sb, and Te (except for Te in HI-2) appear to fall below the corresponding curves, suggesting conservatism for these elements also. As reported by Lorenz et al.<sup>23</sup> and by Albrecht and Wild,<sup>24</sup> tellurium appears to be retained by metallic Zircaloy cladding but may be released in very large fraction when the cladding is nearly completely oxidized.

HI5



1



2



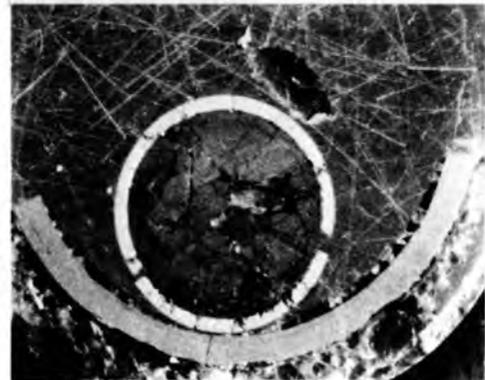
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6

Fig. 23. Successive radial sections from test HI-5 fuel specimen, with 1 being 1 cm from inlet end and 6 near outlet end.

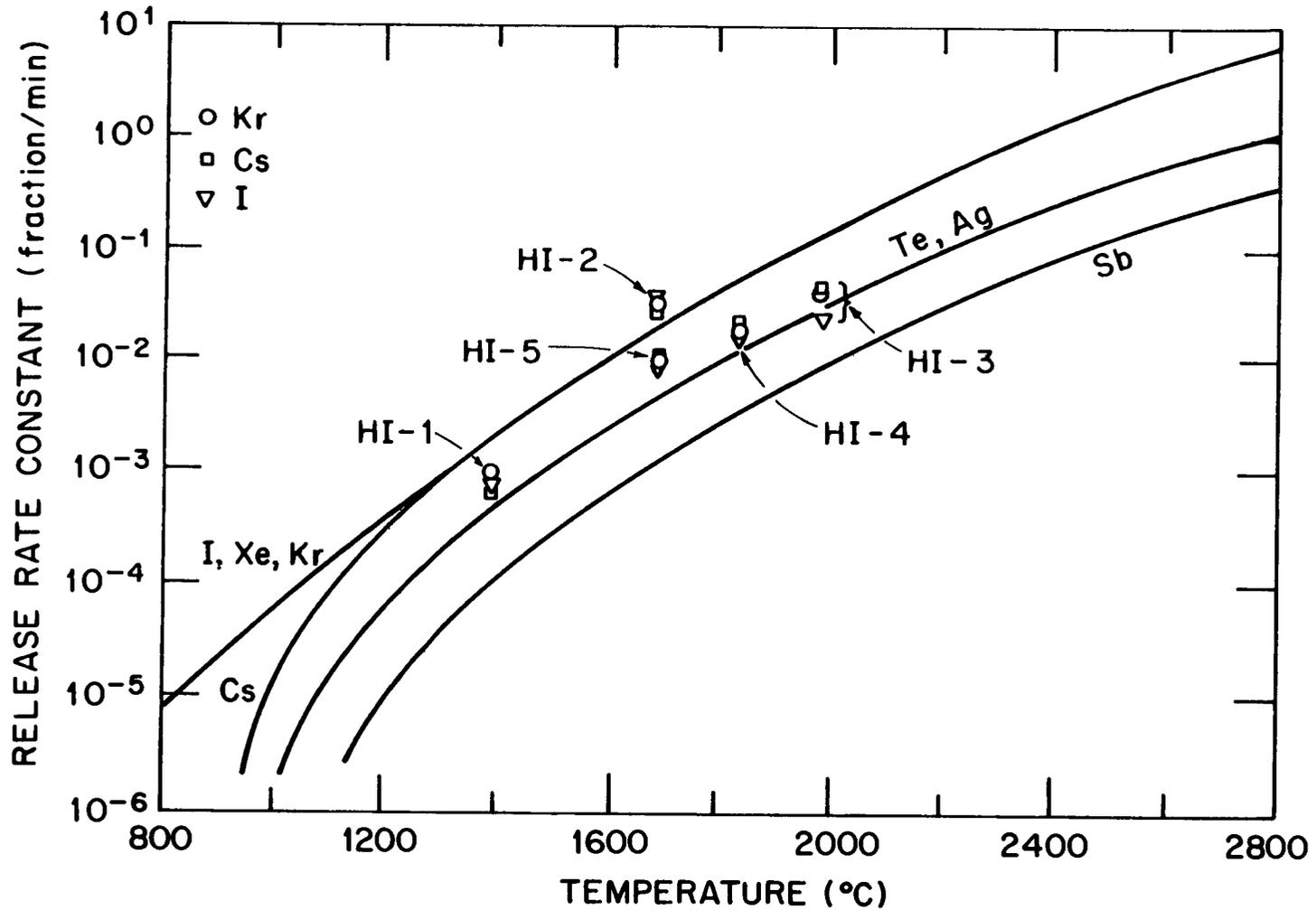


Fig. 24. Comparison of release rate data from HI tests with curves from NUREG-0772. Source: Technical Bases for Estimating Fission Product Behavior During LWR Accidents, NUREG-0772, U.S. Nuclear Regulatory Commission, June 1981.

## 6. CONCLUSIONS

Although a thorough interpretation of the results of test HI-5 are beyond the scope of this document, several significant observations are appropriate:

1. The planned test conditions were accomplished, and all components of the fission product collection system provided useful data; therefore, the test was considered to be successful.
2. The release rates for  $^{85}\text{Kr}$  and  $^{137}\text{Cs}$  (Fig. 8) are similar to those observed in previous tests under similar conditions.<sup>7-9</sup>
3. Based on the analyses of released material and on the calculated inventories in the fuel, the total release fractions for  $^{85}\text{Kr}$ ,  $^{129}\text{I}$ , and  $^{137}\text{Cs}$  (19.9, 22.4, and 20.3%, respectively) were similar. Since the release fractions for these three nuclides have been found to be consistently similar, not only in the current series of tests,<sup>6-9</sup> but also in previous tests,<sup>2-4</sup> the possibility of similar release mechanisms from the  $\text{UO}_2$  is suggested. These data for Kr, I, and Cs were converted to a release rate (fraction/min) and compared with the corresponding curves from an NRC-sponsored review of fission product release data.<sup>20</sup> As shown in Fig. 24, all points except those for test HI-2 fall below the Kr, I, and Cs curve.
4. A comparable fraction of the  $^{110\text{m}}\text{Ag}$  (18%) was released from the fuel; however, >99% of this released silver was deposited on  $\text{ZrO}_2$  ceramics in the outlet end of the furnace. The absence of any  $^{154}\text{Eu}$ , which appears to be a good fuel indicator, verified that this silver had escaped from the fuel. Although only 0.3% of the  $^{125}\text{Sb}$  was released from the fuel, 99% of it also was deposited on the furnace ceramics.
5. Pre- and posttest gamma spectrometric analysis of the fuel specimen indicated that 24.5% of the  $^{134}\text{Cs}$  was released, which is in good agreement with the above value obtained by comparing the "summation of components" value with the ORIGEN-calculated inventory.
6. Broad deposition peaks for cesium and iodine in the TGT at temperatures of 500 to 600°C showed that the behavior of these elements was similar to that in previous tests in this series.<sup>6-9</sup>
7. Some of the particles collected on the aerosol sampler contained almost as much iodine as cesium and were therefore concluded to be primarily CsI with a general background of cesium.
8. The cladding was heavily oxidized and extensively fractured during the test, but no melting or fuel/cladding interaction was observed in the preliminary examination.

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<p>The fifth in a series of high-temperature fission product release tests was conducted for 20 min at 1700°C in flowing steam. The test specimen, a 15.2-cm-long section of a fuel rod which had been irradiated to a burnup of ~38.3 MWd/kg, was heated in an induction furnace under simulated LWR accident conditions in a hot cell.</p> <p>Posttest inspection showed severe oxidation and fragmentation of the fuel specimen, but no cladding melting was apparent. Analyses of test components showed total releases from the fuel of 19.9% for <sup>85</sup>Kr, 22.4% for <sup>129</sup>I, 18.0% for <sup>110m</sup>Ag, and 20.3% for <sup>137</sup>Cs. A smaller fraction of the <sup>125</sup>Sb (0.326%) was released from the fuel, and ~99% of the <sup>110m</sup>Ag and <sup>125</sup>Sb was retained in the furnace. Posttest analysis of the fuel specimen indicated a <sup>134</sup>Cs release of 24.5%, which is reasonably good agreement with the <sup>137</sup>Cs data. These releases were less than half those in test HI-2, where more oxidation and a large axial crack probably were significant factors in the release of fission products.</p>					
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