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# Data Summary Report for Fission Product Release Test VI-2

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Prepared by M. F. Osborne, J. L. Collins, R. A. Lorenz,  
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Oak Ridge National Laboratory

Prepared for  
U.S. Nuclear Regulatory Commission

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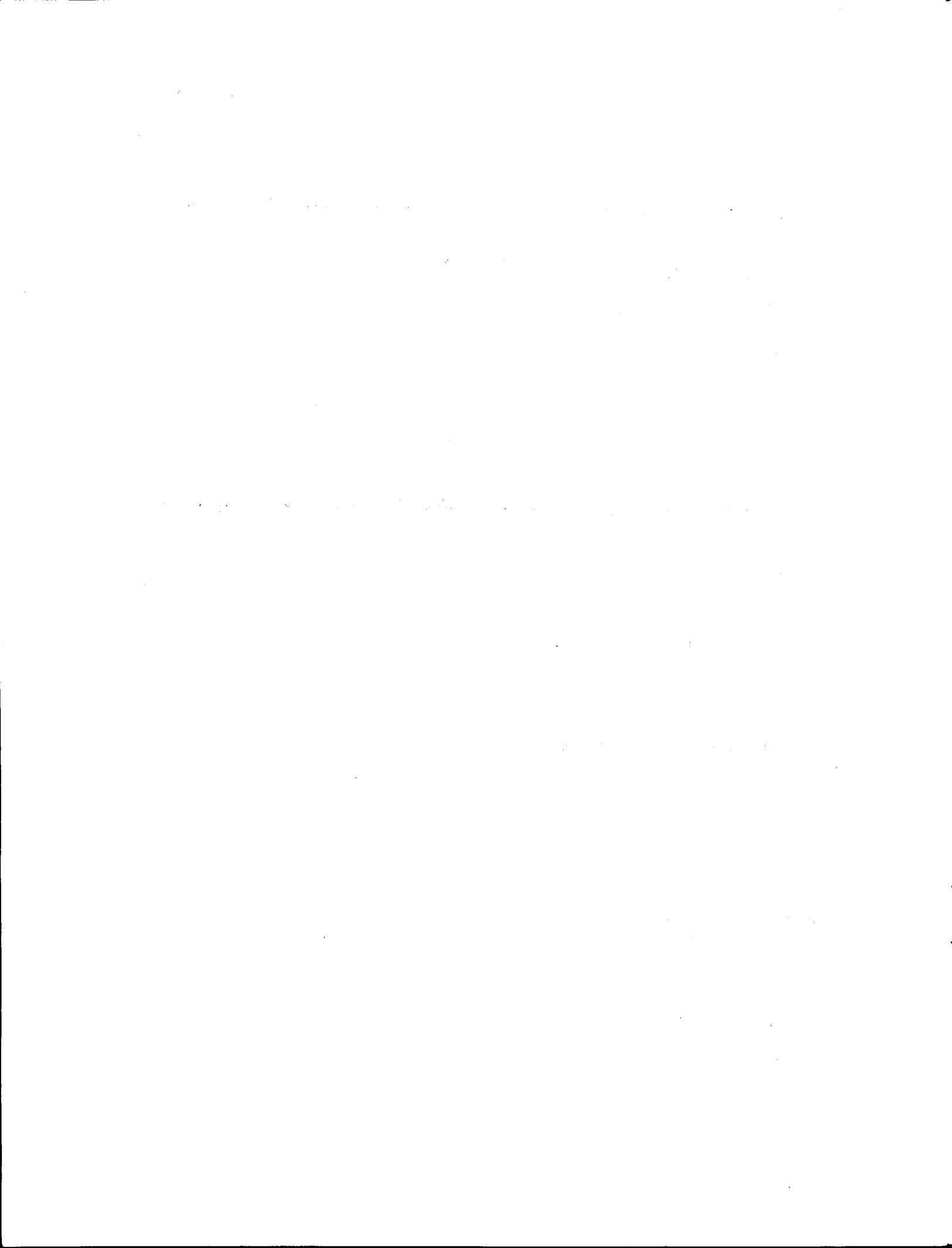
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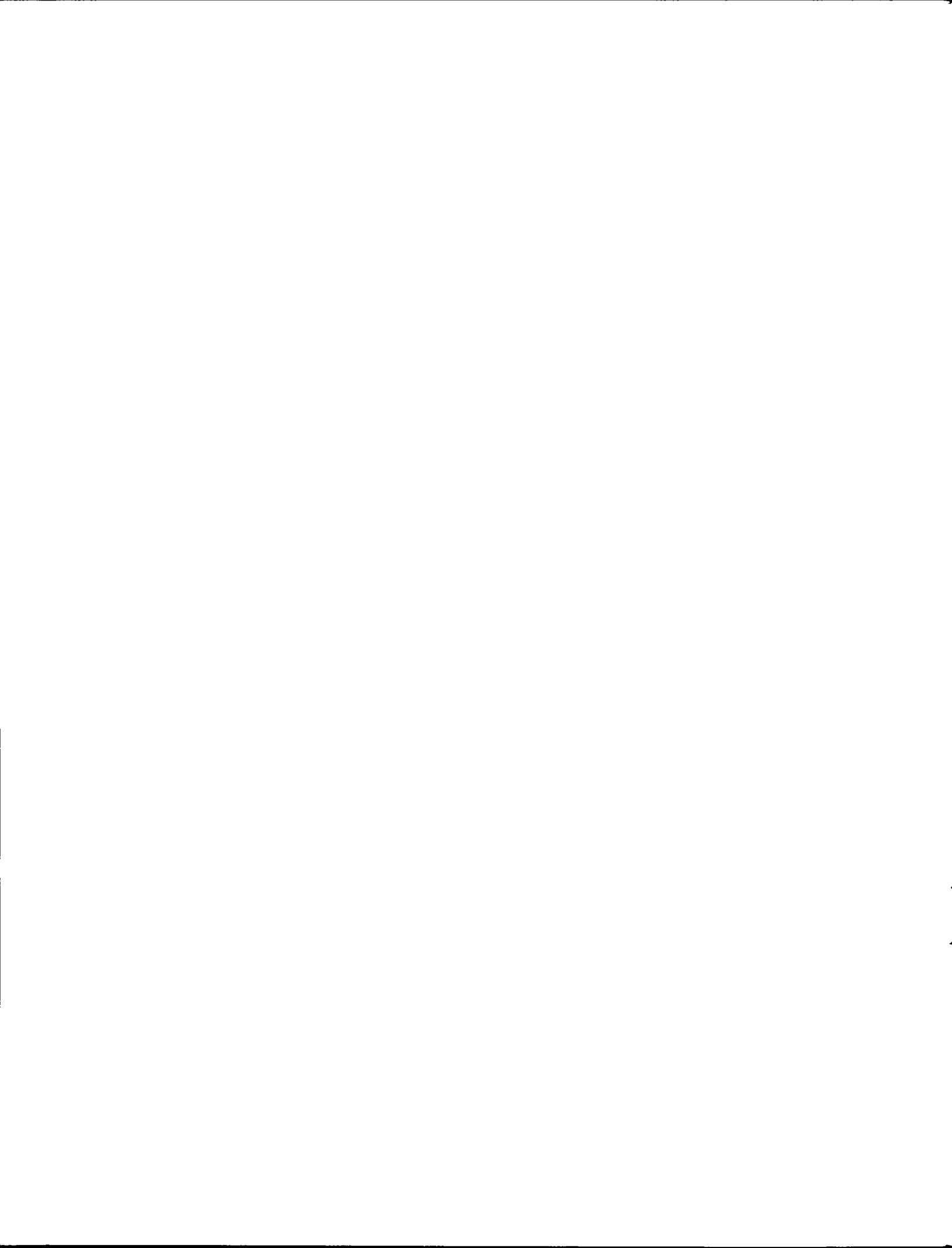


## ABSTRACT

The second in a series of high-temperature fission product release tests in a new vertical test apparatus was conducted in flowing steam. The test specimen, a 15.2-cm-long section of a fuel rod from the BR3 reactor in Belgium, had been irradiated to a burnup of  $\sim 44$  MWd/kg. Using an induction furnace, it was heated under simulated light water reactor (LWR) accident conditions — 60 min at 2300 K — in a hot cell-mounted test apparatus equipped with special components designed to facilitate collection and analysis of the released fission products.

Posttest inspection showed severe oxidation of the cladding but only minimal fragmentation of the fuel specimen; evidence of cladding melting was apparent only near the top end. Based on fission product inventories measured in the fuel or calculated by ORIGEN2, analyses of test components showed total releases from the fuel of 30.7% for  $^{85}\text{Kr}$ , 8.93% for  $^{110\text{m}}\text{Ag}$ , 68.2% for  $^{125}\text{Sb}$ , and 63.4% for  $^{137}\text{Cs}$ . Large fractions (63% and 23%, respectively) of the released  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$  were retained in the furnace, but most of the released cesium (96%) escaped to the collection system. Pretest and posttest analysis of the fuel specimen showed that  $^{134}\text{Cs}$  was released in the same fraction as  $^{137}\text{Cs}$ . Smaller release fractions for many other fission products were measured. In addition, very small amounts of fuel material — uranium and plutonium — were released at relatively constant rates. Total mass release from the furnace to the collection system, which included fission products, fuel material, structural materials, and impurities, was 1.08 g.

The results from the first two phases (2 min and 18 min) of this test were compared with the results from two earlier tests at similar conditions. The total release of cesium was equal in one test (HI-3) and somewhat greater in another (VI-1). Differences in fuel characteristics may contribute significantly to this difference in release behavior. Comparison of the six tests in the HI series and tests VI-1 and VI-2 shows that the release rates for cesium are in reasonable agreement, and that the release rates for all eight experiments fall significantly below a widely accepted standard used in LWR safety analyses.



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## FOREWORD

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

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, 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.
6. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-6, NUREG/CR-4043 (ORNL/TM-9943), September 1985.
7. 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.
8. 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.
9. 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.
10. M. F. Osborne, J. L. Collins, P. A. Haas, R. A. Lorenz, J. R. Travis, and C. S. Webster, Design and Final Safety Analysis Report for Vertical Furnace Fission Product Release Apparatus in Hot Cell B, Building 4501, NUREG/CR-4332 (ORNL/TM-9720), March 1986.

11. M. F. Osborne, J. L. Collins, and R. A. Lorenz, Highlights Report for Fission Product Release Tests of Simulated LWR Fuel, ORNL/NRC/LTR-85/1, February 1985.
12. M. F. Osborne, J. L. Collins, R. A. Lorenz, and T. Yamashita, Highlights Report for Fission Product Release Test VI-1, ORNL/NRC/LTR-86/7, March 1986.
13. M. F. Osborne, J. L. Collins, R. A. Lorenz, and T. Yamashita, Highlights Report for Fission Product Release Test VI-2, ORNL/NRC/LTR-86/18, January 1987.
14. M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, C. S. Webster, and T. Yamashita, Data Summary Report for Fission Product Release Test VI-1, NUREG/CR-5339 (ORNL/TM-11104), April 1989.

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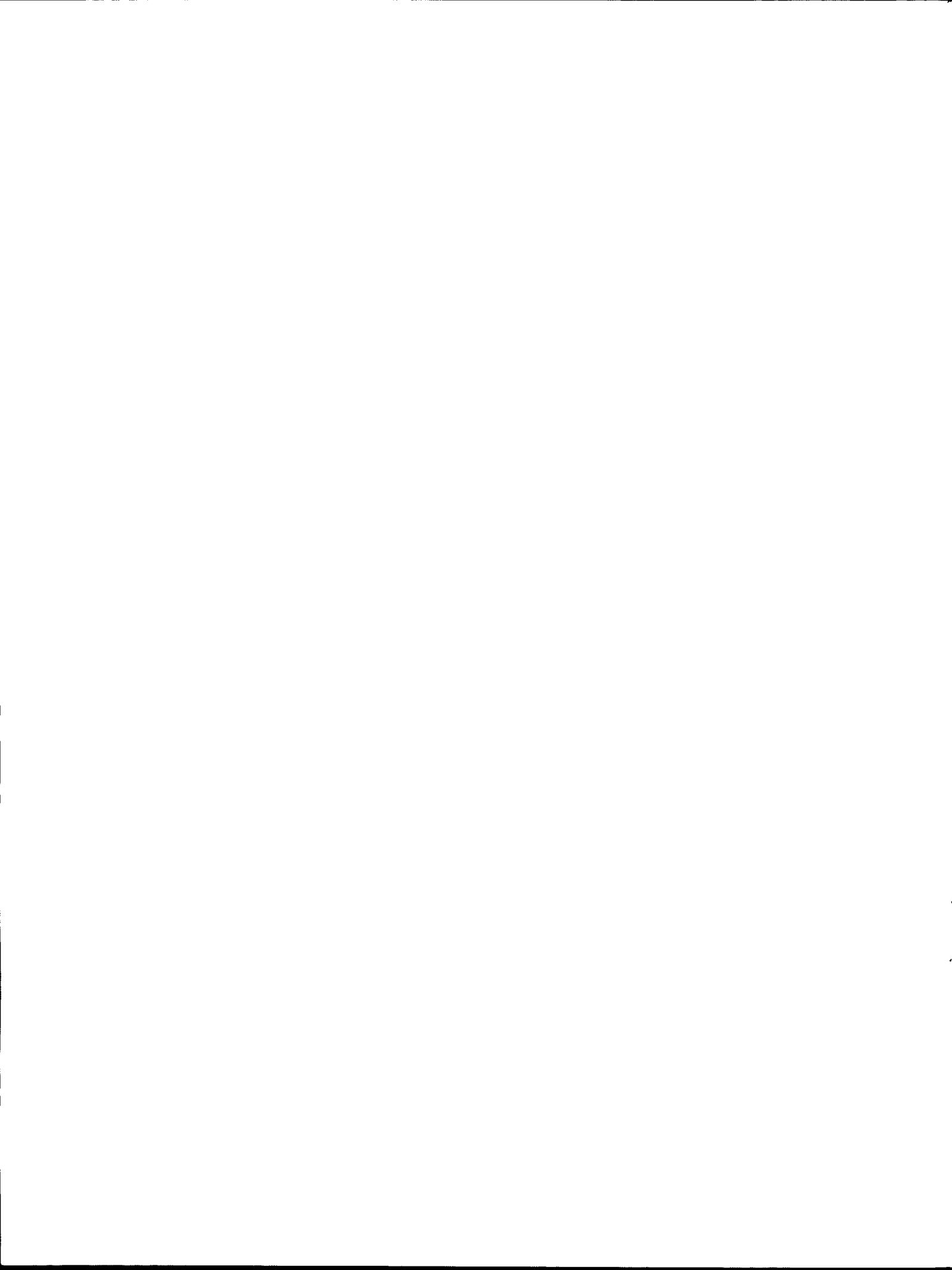
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DATA SUMMARY REPORT FOR FISSION PRODUCT RELEASE TEST VI-2

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

## 1. EXECUTIVE SUMMARY

The objective of this report is to document as completely as possible the observations and results of this fission product release test (VI-2). Although all final data are not currently available, complete interpretation and correlation of these results with related experiments and with theoretical behavior will be included in a subsequent report, which will consider the results of several tests over a range of test conditions. Similar data summary reports for previous tests in this project are listed in the Foreword.

The fuel specimen used in this test was cut from a fuel rod that was irradiated in the BR3 reactor in Belgium from July 15, 1976, to September 26, 1980. The fabrication and irradiation history of this fuel rod was compiled by Adams and Dabell. The fission product inventories, as measured in the fuel and calculated by ORIGEN2, are listed in Sect. 3. The test results and some preliminary interpretations are presented in Sect. 4, and these results are compared with previous data in Sect. 5. The most important results are:

1. This was the second test in a new, vertically oriented induction furnace with three sequentially operated fission product collection trains and equipment for the continuous measurement of hydrogen generated by the reaction of steam with the Zircaloy cladding. With the exception of some problems with the on-line radiation detectors (caused by higher than expected levels of radiation), all of the experimental apparatuses performed quite well.
2. Compared with previous tests, the test conditions were relatively severe in that a high test temperature (2300 K) was maintained for 60 min. (In earlier tests, the highest temperature was held for a maximum of 20 min.) The long test period was used in this case to investigate the change in release behavior with fraction released.
3. Posttest examination indicated that the Zircaloy cladding was completely oxidized, contained numerous minor fractures, and had experienced limited melting and/or fuel-cladding interaction near the top end of the specimen. The 15-cm-long specimen, however, did not collapse but remained in its original vertical orientation.
4. The oxidation behavior of the Zircaloy cladding, as indicated by hydrogen generation, was in good agreement with the Zircaloy oxidation model developed by Yamashita, using the generally accepted oxidation rates as a function of temperature.

5. The values for total release from the fuel specimen for  $^{85}\text{Kr}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{125}\text{Sb}$ , and  $^{137}\text{Cs}$  were 30.7, 8.93, 68.2, and 63.4%, respectively. About two-thirds of the released  $^{110\text{m}}\text{Ag}$  and one-fourth of the released  $^{125}\text{Sb}$ , however, were deposited on the  $\text{ZrO}_2$  ceramics at the outlet end of the furnace, compared to almost complete transport of the released krypton and cesium from the furnace to the collection system. Pretest and posttest gamma ray spectrometry of the fuel was valuable in determining the release fractions as well as the distributions of the fission products along the fuel rod.
6. Small amounts of uranium were found at various locations in the test apparatus. Based on chemical analyses of leach solutions, 17.7 mg of uranium were released from the fuel, with 7.4 mg (42%) of this total deposited in the outlet end of the furnace. The remainder (10.3 mg) was distributed among the three collection trains in proportion to the time of operation, indicating a relatively constant release rate.
7. The fission product distributions in the thermal gradient tubes (TGTs) showed a large cesium peak at  $\sim 600^\circ\text{C}$  and several smaller peaks at lower temperatures in tube A, and narrow peaks of both cesium and antimony at the entrance to tube B. In tube C, small peaks of cesium, antimony, and silver appeared near the entrance (at  $\sim 800^\circ\text{C}$ ), followed by a generally higher level of cesium throughout the length of the tube. After leaching to remove the soluble deposits, cesium, antimony, and silver were measured throughout all three tubes, apparently alloyed or insolubly combined with the platinum deposition tubes, in approximately logarithmically decreasing concentrations.
8. The masses of deposits on the TGTs and filters were determined by direct weighing. A total of 1.082 g of material was found, with almost half (0.55 g) being deposited during the second (18 min at 2300 K) phase of the test.

## 2. INTRODUCTION

This report summarizes data from the second test in a new vertical test apparatus. This series of tests is designed to investigate fission product release from light water reactor (LWR) fuel in steam and/or hydrogen in the temperature range of 2000 to 2700 K. Earlier tests, conducted under similar conditions at temperatures of 773 to 1873 K (500 to  $1600^\circ\text{C}$ ), were reported in detail by Lorenz et al.;<sup>1-4</sup> additional tests at higher temperatures ( $\sim 1700$  to 2300 K) were reported by Osborne et al.<sup>5-11</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 a variety of heatup accidents in LWRs.\* The primary objectives of this program are:

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\*M. F. Osborne, R. A. Lorenz, and R. P. Wichner, "Program Plan for Fission Product Release from LWR Fuel in Steam," memorandum to U.S. Nuclear Regulatory Commission, April 1982.

1. to determine fission product release from commercial LWR fuel at temperatures up to and including fuel melting (~2700 K);
2. to identify, to the extent possible, the chemical and physical forms of the released fission products;
3. to compare the observed fission product release and behavior with the physical and chemical changes in the fuel specimens; and
4. to compare the results with data from related programs and to 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. The applicability of simulated fuel (unirradiated  $UO_2$  containing fission product tracers) has been, and will be, considered where appropriate.<sup>12,13</sup> All tests have been conducted in a flowing mixture of steam and helium or of hydrogen and helium at atmospheric pressure; steam concentrations have been varied to simulate different accident sequences or core locations.

This vertical test apparatus<sup>14</sup> expands test capabilities over the previous horizontal equipment<sup>15</sup> in several ways: (1) the furnace and TGTs are vertical, more nearly simulating the orientation of the fuel rods in an LWR; (2) three collection trains, each including a TGT and filter package, are used in sequence during the test to provide better time/temperature resolution; (3) temperature measurement and control have been improved, limiting the axial thermal gradient over the fuel specimen to <50 K; (4) gas flow rate and distribution have been modified to provide more uniform atmospheric (oxidizing vs reducing) conditions over the length of the specimen; and (5) additional equipment was added to provide continuous measurement of the hydrogen generated by steam oxidation of the cladding.

The procedures and techniques used in posttest examination and analysis were very similar to those used for the six tests in the HI series<sup>5-10</sup> and in test VI-1.<sup>11</sup> The analytical techniques used are listed in Table 1. This report provides a brief description of test VI-2 and a tabulation of all the results obtained to date. Because some of the analyses have been delayed and the results will not be available for several months, they will be published in a later report. Thorough data evaluation and correlation with other test results will be included in subsequent reviews and reports covering this series of fission product release tests at temperatures up to 2700 K.

### 3. DESCRIPTION OF TEST VI-2

The tests planned for this vertical apparatus include temperature (2000 to 2700 K) as the dominant variable; steam flow rate (0.3 to 1.5 L/min) and time (1 to 60 min), both of which affect the extent of oxidation, are secondary variables. A tentative test matrix, which defines the conditions for a series of tests, is shown in the Appendix.

Table 1. Analytical techniques for fission product analysis

Technique	Time	Location	Elements
Gamma spectrometry	Pretest, posttest	Fuel specimen	Long-lived, gamma-emitting fission products — Ru, Ag, Sb, Cs, Ce, Eu
	On-line	Thermal gradient tube, charcoal traps, filters	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, (Te, Mo, Ba) <sup>a</sup>
Chemical analysis	Posttest	Furnace, filters	U, Pu
Spark-source mass spectrometry	Posttest	Samples from furnace, thermal gradient tube, filters	All elements with atomic numbers >10
Scanning electron microscope, with energy dispersive X-ray system	Posttest	TGT, aerosol sampler	All elements with atomic numbers >10

<sup>a</sup>Iodine and bromine have been analyzed by neutron activation throughout HI and VI test series; neutron activation analysis techniques for Te, Mo, and Ba are in various stages of development.

The objective of this particular test was to obtain release data at 2300 K over a period of 60 min in steam flowing at the maximum rate, 1.5 L/min, so that an excess of steam would be present at all times. Summing the data from the first two phases of this test (2 min and 18 min, respectively) provided the opportunity to compare these results with the results of an earlier test for a 20-min period (HI-3)<sup>7</sup> and to the results from phase C (20 min at 2300 K) of test VI-1.<sup>11</sup>

### 3.1 FUEL SPECIMEN DATA

The test specimen was a 15.2-cm-long section of rod I-1002 from the BR3 reactor in Belgium. This fuel was irradiated from July 15, 1976, until September 26, 1980. Details of the irradiation and of the characteristics of this particular specimen were reported by Adams and Dabell\* and are listed in Table 2. Fission product inventories for the specimen were measured by direct gamma spectrometry of the fuel and/or were calculated with the ORIGEN2 computer program,<sup>16</sup> with adjustments based on direct analyses for <sup>137</sup>Cs in the fuel; these data are shown in Tables 3 and 4. The agreement between these two methods, however, was not consistently good. As we have observed in previous work<sup>2</sup> and is shown in Table 5, the agreement was particularly poor for <sup>125</sup>Sb and <sup>154</sup>Eu but surprisingly good for <sup>144</sup>Ce. In calculating the percent release values shown in subsequent tables and discussion, we used the <sup>137</sup>Cs-based ORIGEN2 values for all nuclides other than <sup>125</sup>Sb and <sup>137</sup>Cs; the measured inventory values were used for these two nuclides.

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

Fuel rod identification	Rod I-1002, Series B 300 BR3 reactor
Irradiation data:	
Period	July 15, 1976 to Sept. 26, 1980
Specimen burnup	~44 MWd/kg U
Specimen data:	
Length	15.2 cm
Fuel loading	82.0 g UO <sub>2</sub> (72.3 g U)
Total weight	103.3 g
Weight of Zircaloy	21.3 g
Gas release during irradiation	2% (from specimen)

\*J. P. Adams and B. R. Dabell, "Characteristics of UO<sub>2</sub>-Zr Fuel Rods Irradiated in the BR3 Reactor," private communication, 1986.

Table 3. Inventories of principal fission product elements  
in test VI-2 fuel<sup>a</sup>

Elements	Amounts in fuel (g/MTU)	Amounts in test specimen <sup>b</sup> (g)
Se	7.937E+01	5.738E-03
Br	3.043E+01	2.200E-03
Kr	5.348E+02	3.867E-02
Rb	5.261E+02	3.804E-02
Sr	1.218E+03	8.806E-02
Y	6.937E+02	5.015E-02
Zr	5.175E+03	3.742E-01
Mo	4.528E+03	3.274E-01
Tc	1.043E+03	7.541E-02
Ru	2.690E+03	1.945E-01
Rh	5.539E+02	4.005E-02
Pd	1.337E+03	9.667E-02
Ag	6.648E+01	4.807E-03
Cd	9.568E+01	6.918E-03
In	2.579E+00	1.865E-04
Sn	9.441E+01	6.826E-03
Sb	2.202E+01	1.592E-03
Te	5.877E+02	4.249E-02
I	2.752E+02	1.990E-02
Xe	6.960E+03	5.032E-01
Cs	3.368E+03	2.435E-01
Ba	2.237E+03	1.617E-01
La	1.663E+03	1.202E-01
Ce	3.258E+03	2.356E-01
Eu	1.636E+02	1.183E-02
Total fission products	4.554E+04	3.293E+00
U	9.450E+05	6.832E+01
Pu	8.410E+03	6.080E-01
Total actinides	9.544E+05	6.900E+01

<sup>a</sup>Inventories for this BR3 fuel were calculated by ORIGEN2, assuming a burnup of 44,060 MWd/MTU, and were adjusted based on the results of gamma ray spectrometric analysis of the fuel before testing; corrected for decay to July 1, 1986.

<sup>b</sup>The test VI-2 specimen contained 72.3 g U, which is equal to 7.23E-05 MTU before testing.

Table 4. Inventory of long-lived radionuclides in test VI-2 fuel<sup>a</sup>

Radionuclides	Amounts in fuel		Amounts in test specimen <sup>b</sup>	
	(g/MTU)	(Ci/MTU)	(g)	(Ci)
<sup>85</sup> Kr	2.269E+01	8.905E+03	1.641E-03	6.438E-01
<sup>90</sup> Sr	6.881E+02	9.389E+04	4.975E-02	6.788E+00
<sup>106</sup> Ru	1.953E+00	6.536E+03	1.412E-04	4.726E-01
<sup>110m</sup> Ag	1.543E-03	7.334E+00	1.116E-07	5.303E-04
<sup>125</sup> Sb <sup>c</sup>	1.354E+00	1.398E+03	9.792E-05	1.011E-01
<sup>129</sup> I	2.138E+02	3.776E-02	1.546E-02	2.730E-06
<sup>134</sup> Cs	1.786E+01	2.312E+04	1.291E-03	1.672E+00
<sup>137</sup> Cs	1.355E+03	1.180E+05	9.797E-02	8.535E+00
<sup>144</sup> Ce	1.713E+00	5.468E+03	1.239E-04	3.953E-01
<sup>154</sup> Eu	2.731E+01	7.376E+03	1.975E-03	5.333E-01
Total fission products	4.554E+04	5.226E+05	3.293E+00	3.778E+01
<u>Actinide elements</u>				
U	9.450E+05	4.718E+00	6.832E+01	3.411E-04
Pu	8.410E+03	8.626E+04	6.080E-01	6.237E+00
Total actinides	9.544E+05	8.820E+04	6.900E+01	6.377E+00

<sup>a</sup>Inventories for this BR3 fuel were calculated by ORIGEN2, assuming a burnup of 44,060 MWd/MTU, and were adjusted based on the results of gamma ray spectrometric analysis for <sup>137</sup>Cs in the fuel before testing; corrected for decay to July 1, 1986.

<sup>b</sup>The test VI-2 specimen contained 72.3 g U, which is equal to 7.23E-05 MTU before testing.

<sup>c</sup>Using a measured value of <sup>125</sup>Sb in the fuel specimen (0.1011 Ci) and calculating other values.

Table 5. Fission product inventories in test VI-2:  
comparison of measured and ORIGEN2 values

Nuclide	Curies in specimen before test		Agreement (ORIGEN2/measured)
	Measured <sup>a</sup>	ORIGEN2 <sup>b</sup>	
<sup>106</sup> Ru		0.4726	
<sup>125</sup> Sb	0.1011	0.2054	Poor, 2.032
<sup>134</sup> Cs	1.271	1.672	Poor, 1.315
<sup>137</sup> Cs	8.531	8.535	Normalized, 1.000
<sup>144</sup> Ce	0.3961	0.3953	Good, 0.998
<sup>154</sup> Eu	0.2054	0.5333	Poor, 2.596

<sup>a</sup>By gamma spectrometry, using best gamma peak and gamma-balanced shielding corrections.

<sup>b</sup>Calculated using irradiation history and postirradiation fuel data, then normalized to the measured <sup>137</sup>Cs in fuel.

No axial scan of the gamma radioactivity along the intact fuel rod was made before the rod was sectioned. Scans of nearby rods with similar operating histories, however, indicated that rod I-1002 contained no unusual distributions of fission products, and that the operating power was not high enough to cause migration of such volatile fission products as cesium.\* Although no gas analyses were obtained for rod I-1002, examination of the data from comparable rods suggested that the release of <sup>85</sup>Kr from the fuel in the region of the test specimen was ~2%.

In addition to the test VI-2 fuel specimen, three other 15.2-cm-long specimens were cut and prepared for future testing; three short samples (1- to 2-cm long) were cut for metallographic examination, for dissolution and chemical analysis, and for possible future use.

Tapered end caps of Zircaloy-2 were pressed onto the ends of the test specimen, not as gas seals but to prevent loss of the fractured UO<sub>2</sub> fuel during subsequent handling. The bottom end cap included a pin to facilitate vertical mounting. A small hole, 1.6 mm diam, 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. 1.

\*J. P. Adams and B. R. Dabell, "Characteristics of UO<sub>2</sub>-Zr Fuel Rods Irradiated in the BR3 Reactor," private communication, 1986.

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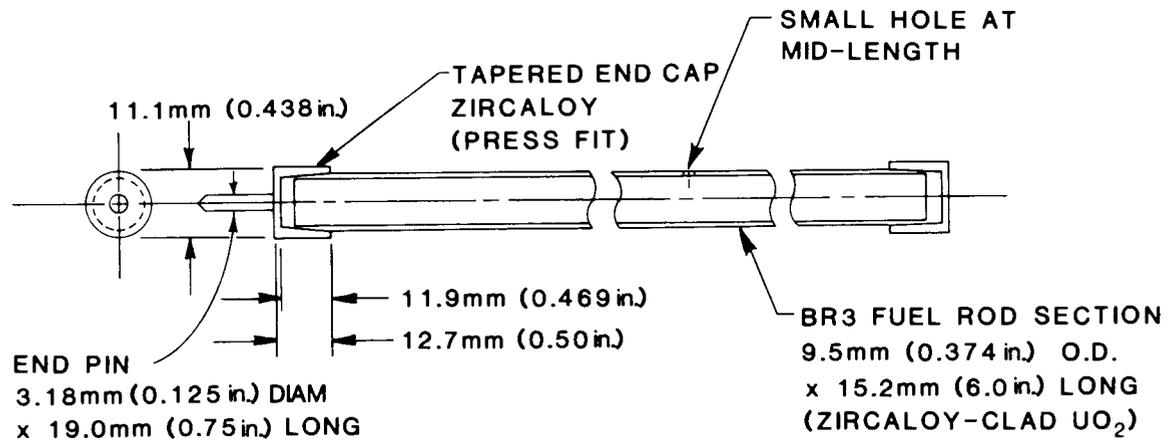


Fig. 1. Details of fuel specimen used in test VI-2.

### 3.2 EXPERIMENTAL APPARATUS

The fuel specimen was heated in an induction furnace, as illustrated in Fig. 2, and described previously.<sup>14</sup> The graphite susceptor was protected from extensive steam oxidation by a blanket of helium gas that leaked through joints in the ZrO<sub>2</sub> ceramics into the flowing helium-steam mixture (see Fig. 2). The furnace was mounted inside a stainless steel containment box in a hot cell, as shown in Fig. 3. The fission product collection system included (1) platinum liners for the thermal gradient tubes (for the collection of condensable vapors according to their deposition temperatures); (2) graduated fiberglass filters (for the collection of aerosol particles); (3) heated charcoal (for the adsorption of iodine species such as elemental iodine, hydrogen iodide, and organic iodides); and (4) cooled charcoal (for the adsorption of inert fission gases). The steam was collected in a condenser and a dryer, as indicated in Fig. 4, prior to reaching the cooled charcoal. Furnace temperature was monitored and controlled by means of a W-5% Re vs W-26% Re thermocouple installed in a ceramic thermowell in the inlet end of the furnace (Fig. 2), and a manually operated optical pyrometer of the disappearing filament type and two automatic two-color pyrometers, all located outside the hot cell, as indicated in Fig. 3. Two NaI(Tl) detectors and one Ge(Li) detector, all connected to a multichannel analyzer, monitored the accumulation of radioactivity (primarily <sup>134</sup>Cs, <sup>137</sup>Cs, and <sup>85</sup>Kr) on the TGTs, filters, and cold charcoal traps. Infrared analyzers measured the concentrations of CO and CO<sub>2</sub> in the exit gas stream, and a heated CuO bed with a following condenser/conductivity cell measured the concentration of hydrogen, as indicated in Fig. 4. Conventional electrical power supplies, Type K thermocouples, and gas pressure and flow instruments also were used. 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.

### 3.3 TEST CONDITIONS AND OPERATION

As in each of the previous experiments, the test apparatus was assembled by direct handling, which is possible because the hot cell and test apparatus are decontaminated after each test. Also, new furnace internals, TGT liners, and filter package components are used in each test. Only the transfer and loading of the highly radioactive fuel specimen and the final closure of the furnace and containment box required the use of master slave manipulators. No in-cell operations were required during the test. Before the apparatus heating and steam flow were begun, the test apparatus was evacuated and purged with helium. All connecting lines to the furnace, TGTs, and filter assemblies were preheated to at least 125°C to prevent steam condensation during the test.

This test was intended to investigate fission product release at a constant temperature level (2300 K) for an extended period under strongly oxidizing conditions. The operating conditions are summarized in Table 6, and the temperature history is shown in Fig. 6. The more important events

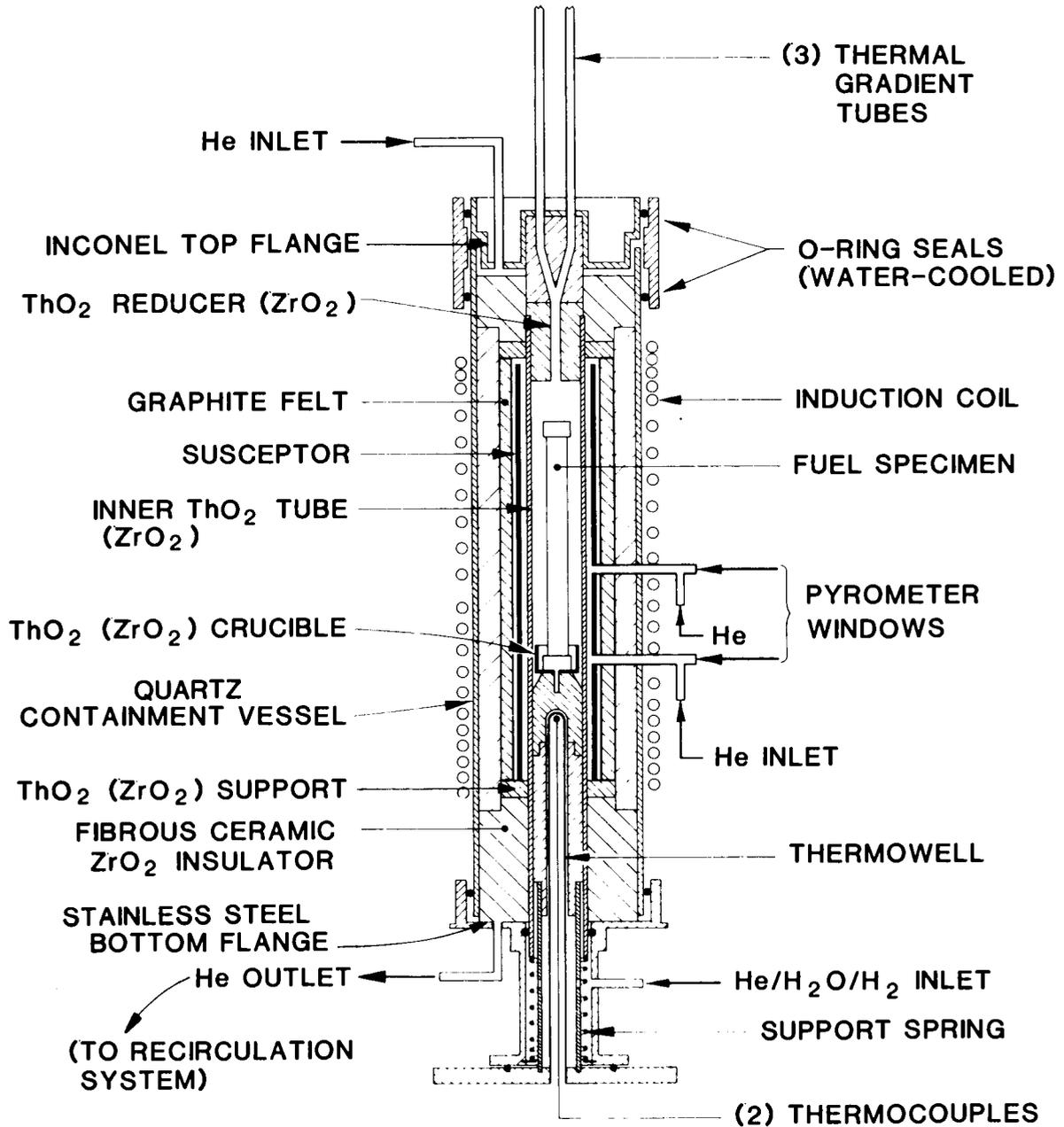


Fig. 2. Vertical induction furnace for fission product release tests.

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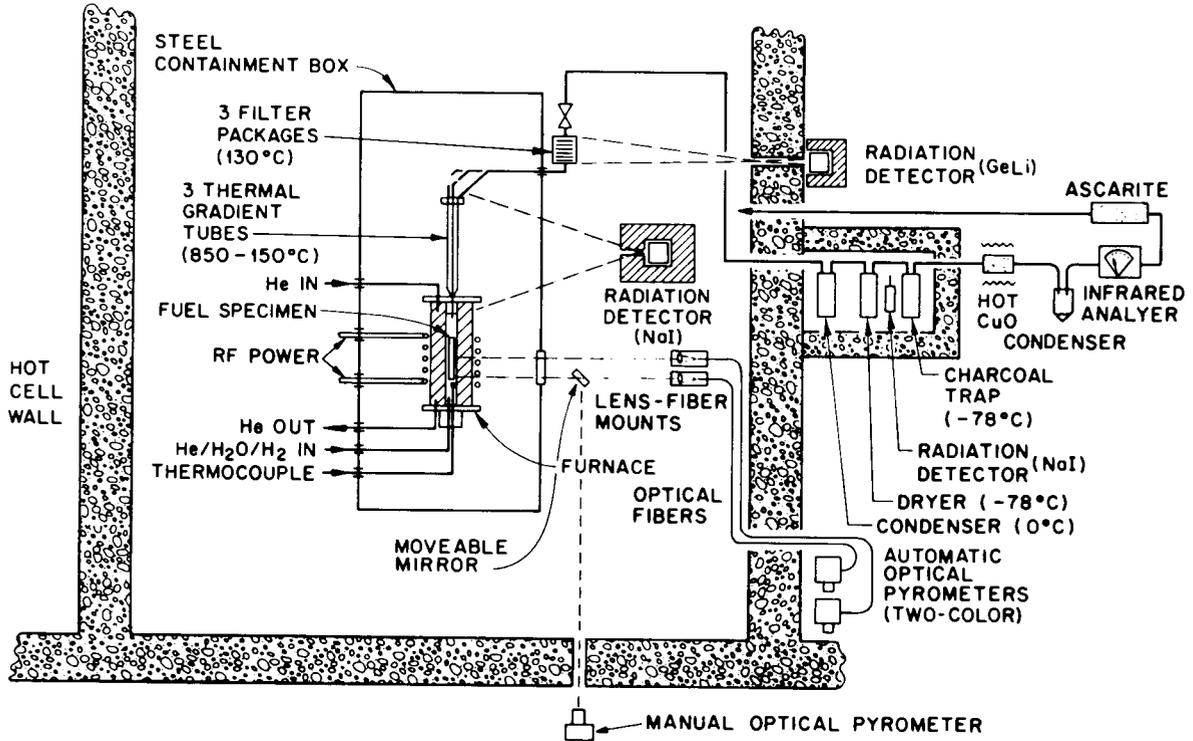


Fig. 3. Vertical fission product release test apparatus.

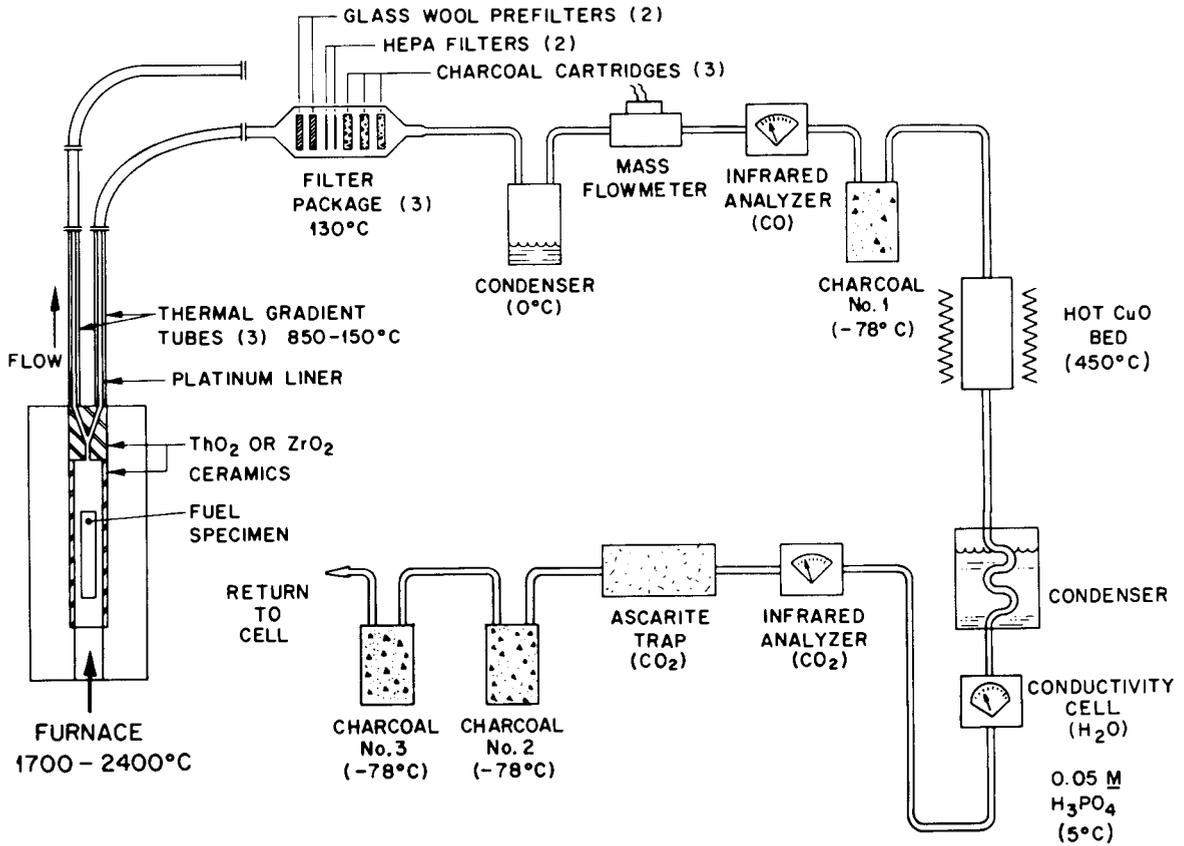


Fig. 4. Components of fission product collection system.

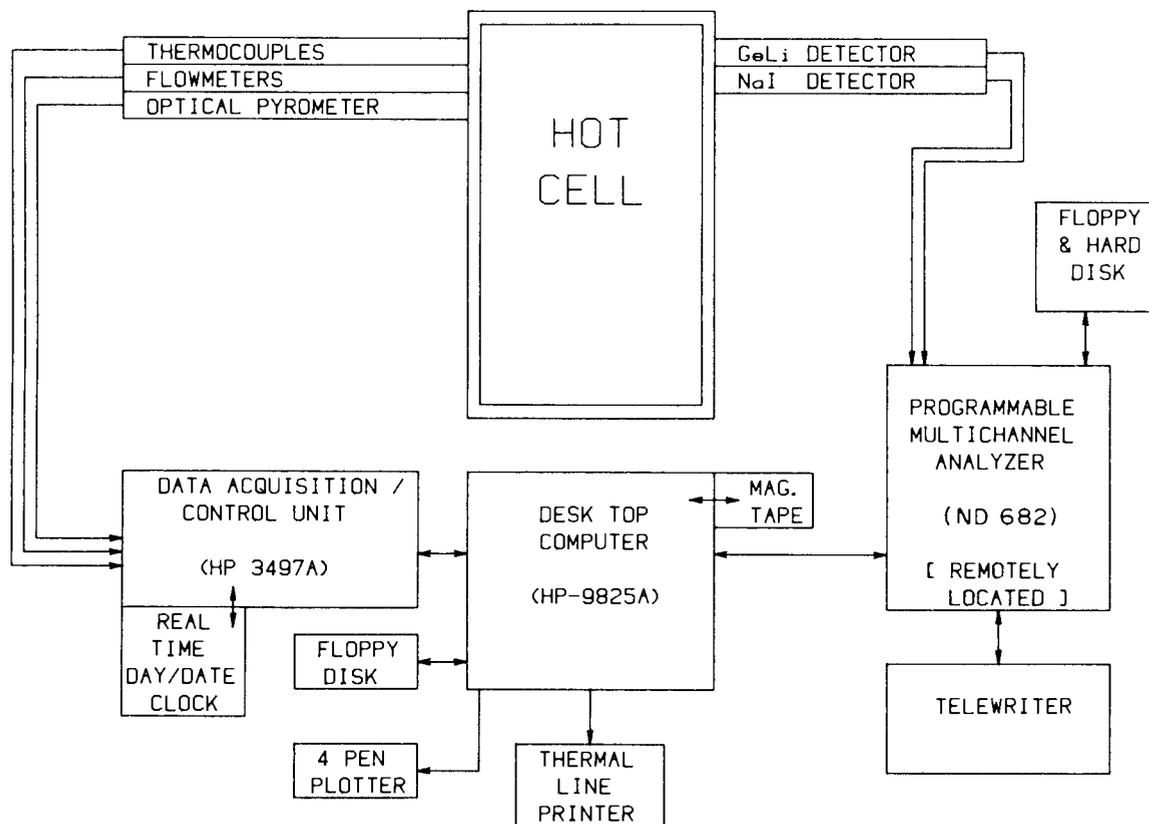


Fig. 5. Data acquisition and processing system for fission product release tests.

Table 6. Operating data for test VI-2

Specimen temperature (K):	
At start of heatup ramp	~495
During 60-min test period	2300
Heatup rate	0.8/s
Cooldown rate	0.5/s
Time above 2000 K	70 min
Nominal gas flow rate data (L/min at 20°C, 1 bar): <sup>a</sup>	
He to recirculation system	0.20
He to steam generator	0.30
Steam into furnace	1.54
Recirculation/purification system	1.4
Integral volume flow data (L): <sup>b</sup>	
He purge to thermocouple	5.64
He to recirculation system	25.92
He to steam generator <sup>c</sup>	36.98
Steam into furnace	179
Recirculation	119
Total H <sub>2</sub> generated <sup>d</sup> (as reacted with CuO)	64.5
H <sub>2</sub> from cladding and end cap oxidation <sup>e</sup>	10.3

<sup>a</sup>Measured by mass flowmeters, except steam measured in condenser.

<sup>b</sup>Measured by totalizers on mass flowmeters during the 126 min of steam flow into apparatus, at a room temperature of 25°C.

<sup>c</sup>Absolute pressure in furnace during test was 1.0116 MPa (759 mm Hg).

<sup>d</sup>At a normal atmospheric pressure of 0.09867 MPa (740.2 mm Hg), measured during test.

<sup>e</sup>Theoretical value assuming 100% oxidation of Zr to ZrO<sub>2</sub>.

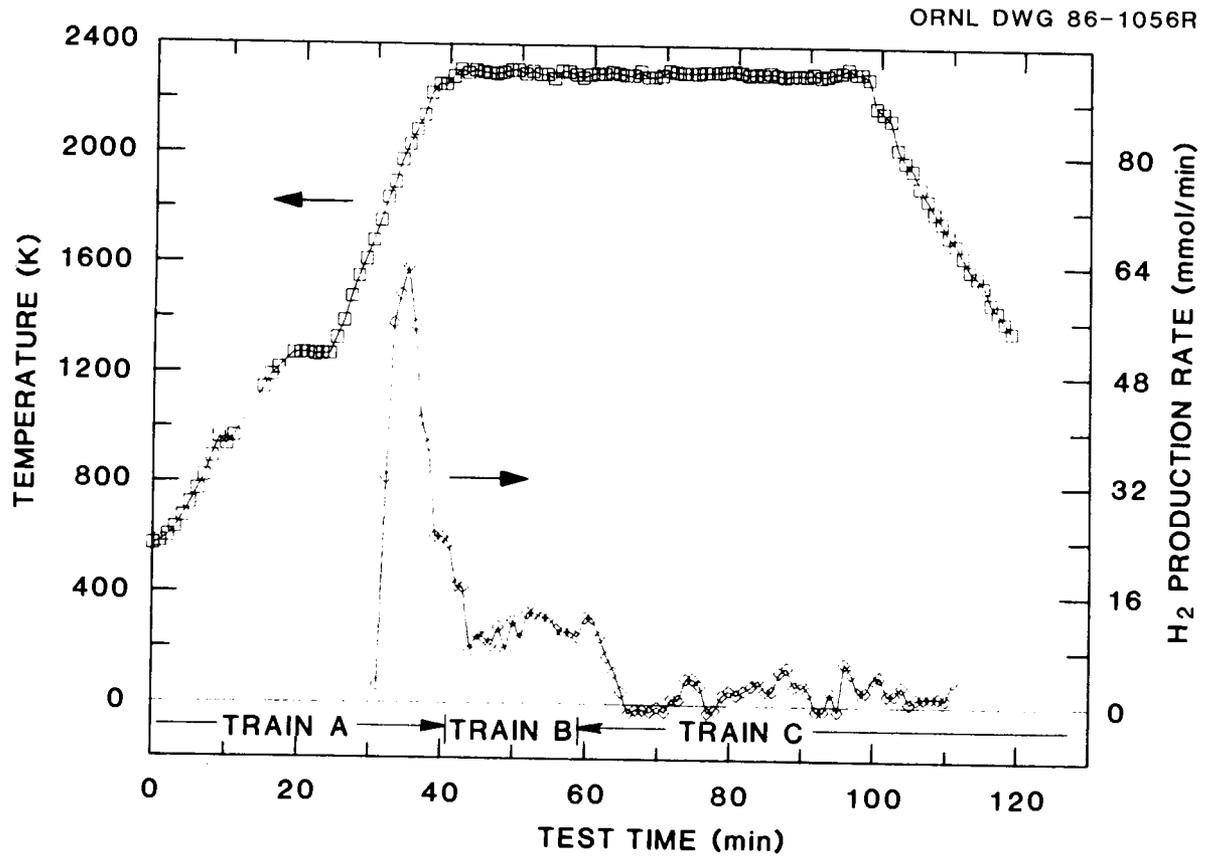


Fig. 6. Temperature and hydrogen production history of test VI-2.

during the test are listed in the test chronology, Table 7. The time periods for operation of the three collection trains (Fig. 6) were: for Train A, 0 to 41 min; for Train B, 41 to 59 min; and for Train C, 59 min to end of test, including cooldown. A preheat period was included to slowly heat the specimen to 550 K before beginning steam flow to the furnace. Time zero was defined as that time when the controlled heating ramp was begun, with stable steam flow through the warm furnace established. Temperature measurement and control were generally good. The 5-min period at 1400 K was included to ensure heatup of ceramics in the outlet end of the furnace and to compare the data from the three optical pyrometers before any significant release of fission products had occurred.

### 3.4 POSTTEST DISASSEMBLY AND EXAMINATION

After the test was completed, the apparatus was monitored for the distribution of radioactivity and then disassembled. Initially, the filter assemblies and the TGT liners were removed and transferred to another hot cell to avoid potential contamination from fuel handling. The top flange was removed from the furnace, and after removal of the ceramic components from the outlet end, the top end of the fuel specimen could be observed. The Zircaloy end cap appeared to be heavily oxidized, and the top end of the specimen was tilted to make contact with the  $ZrO_2$  furnace tube, but the specimen remained mainly intact and upright. To preserve this geometry for sectioning and subsequent microstructural examination, the furnace tube was filled with epoxy resin. After the resin had hardened, the furnace tube/fuel specimen assembly was transferred to the High Radiation Level Examination Laboratory for sectioning and the examination of cross sections.

## 4. TEST RESULTS

### 4.1 TEST DATA

Test temperature and the hydrogen production rate are plotted as a function of test time in Fig. 6, and the change in hydrogen/water ratio with time, both in the furnace and the collection system, is shown in Fig. 7. Hydrogen was measured by conversion of  $H_2$  to  $H_2O$  in the hot  $CuO$  bed, then collection of the  $H_2O$  in a condenser, as indicated in Fig. 4. A modified conductivity cell was used to measure continuously the dilution of an acid solution. In agreement with test VI-1,<sup>11</sup> the hydrogen production rate curve in Fig. 6 indicates a peak at about 2000 K then a decline as most of the Zircaloy cladding became fully oxidized to  $ZrO_2$ . The plateau from about 43 to 60 min probably indicates the period of oxidation of the thicker Zircaloy end caps after completion of cladding oxidation. This cladding oxidation behavior was indicated by measurement of the hydrogen production, with a possible contribution from steam oxidation of the  $UO_2$ . The oxidation rate agreed quite well with a model developed by Yamashita.<sup>17</sup> This model incorporated the oxidation rate coefficients of both Baker and Just<sup>18</sup> and Urbanic and Heidrick,<sup>19</sup> as well as the specific geometry and temperature of our test, to calculate the progressive oxidation of the cladding during the test.

Table 7. Chronology of test VI-2  
July 31, 1986

Event/status	Time		Furnace Temperature	
	Clock	Test (min)	Pyrometer <sup>a</sup> (°C)	True (K)
Load fuel into furnace	1330			RT
Heat steam system	1330			RT
Begin furnace preheat	1545			RT
Begin steam flow to furnace	1632		~225 <sup>b</sup>	~500
Test phase A:				
Start ramp	1640	0	222 <sup>b</sup>	495
First L&N measurement	1650	10	760	1033
Reached first plateau	1659	19	1031	1401
Hold 5 min				
Resumed ramp	1704	24	1029	1399
Reached test temperature	1719	39	1828	2261
Hold 60 min				
Begin test phase B, continue 18 min	1721	41	1852	2287
Begin test phase C, continue 40 min	1739	59	1860	2304
Midpoint test phase C	1759	79	1874	2311
Power off — reduce temperature	1819	99	1856	2291
Cooldown (~2000 K)	1825	105	1585	1996
Steam flow off, continue He only	1838	118	1137	1514
Last L&N measurement	1853	133	818	1175
End test				

<sup>a</sup>Measured with manually operated L&N pyrometer.

<sup>b</sup>Estimated from thermocouple data.

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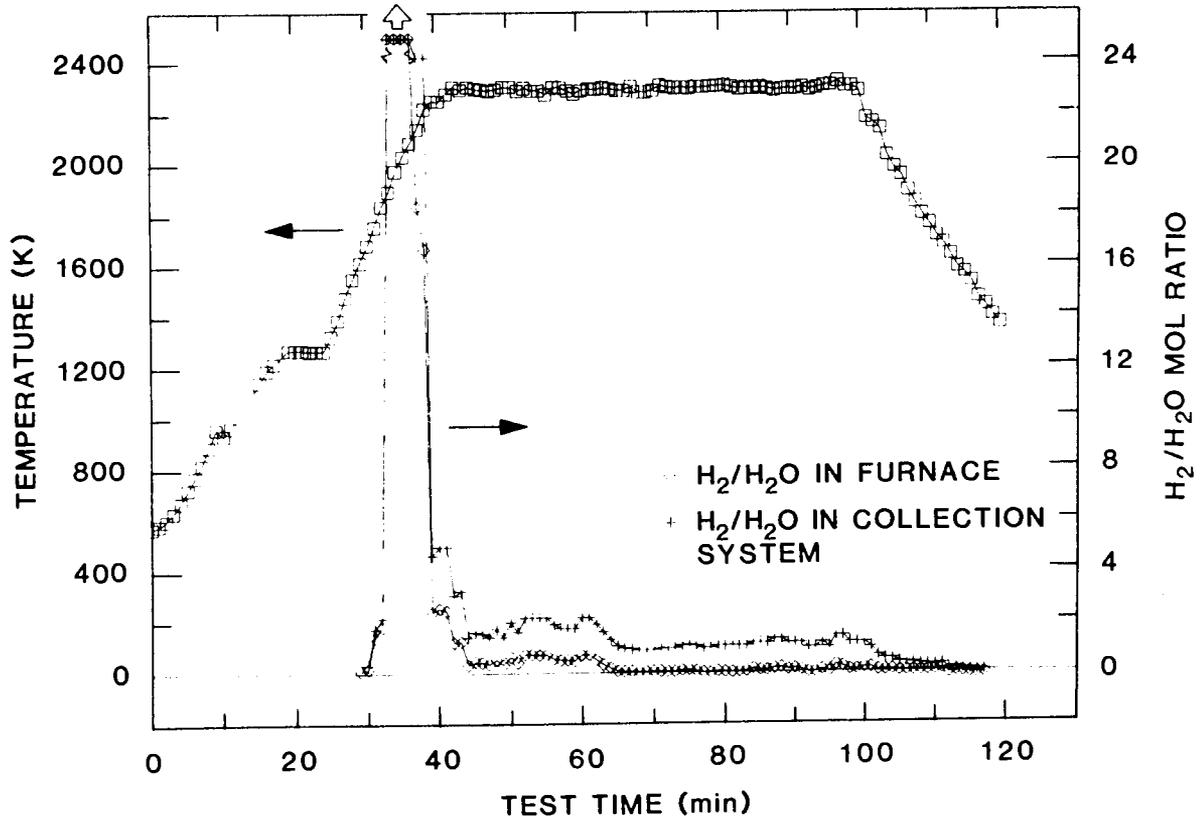


Fig. 7. H<sub>2</sub>/H<sub>2</sub>O ratios in test VI-2 as functions of time and temperature.

## 4.2 POSTTEST DATA

### 4.2.1 Gamma Spectrometry

All experimental components and collectors were analyzed after the test under well-defined geometry to determine the concentration of the gamma-emitting fission products. In addition, the new technique that was developed for processing the release data for test VI-1 was used to provide more precise results. Because all samples required at least double containment (metal, glass, or plastic) and because many samples were actually distributed within other materials (especially the fuel specimen itself), it was difficult to calculate accurately the effective shielding for the various gamma-ray energies of interest. Considering the ubiquitous nature of cesium and the broad range of gamma-ray energies inherent to  $^{134}\text{Cs}$  (475 to 1365 KeV), an empirical method of determining the effective shielding to obtain a mass balance for cesium among several of the  $^{134}\text{Cs}$  gamma-ray energies was developed. This shielding value was then applied to other nuclides, such as  $^{106}\text{Ru}$ ,  $^{110\text{m}}\text{Ag}$ , and  $^{125}\text{Sb}$  to provide more accurate results. This empirical method for energy-dependent shielding correction of gamma analyses will be described in a separate publication.

Pretest gamma spectrometric analysis of the fuel specimen (a 15.2-cm-long section cut from rod I-1002 from the BR3 reactor in Belgium, as described previously\*) was used to determine the fission product inventories in the fuel. Long-lived gamma emitters —  $^{106}\text{Ru}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$ , and  $^{154}\text{Eu}$  — were counted directly; a calculation by the computer program ORIGEN, with corrections based on  $^{137}\text{Cs}$  data, supplied the inventory values for other fission products, activation products and fuel nuclides, as shown in Tables 3 and 4. The distributions of the fission products measured in the fuel before the test are shown in Fig. 8. These pretest data, based on measurements at 3-cm intervals, showed that the distributions of these major fission products was relatively uniform along the rod. Consequently, we conclude that (1) the burnup was similarly uniform, and (2) the operating temperature was low enough to prevent significant migration of volatile species such as cesium.

As has been typical of these tests,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were the dominant activities in almost all samples and interfered with the analysis of less abundant fission products. The release behavior of cesium as functions of time and temperature is illustrated in Fig. 9; these curves show that very little release occurred during the last half of the high-temperature period. A summary of the fractional release results for Kr, Ag, Sb, and Cs, as determined by gamma spectrometry, is presented in Table 8. Unlike all tests in the HI series, the total release values for  $^{85}\text{Kr}$  and  $^{137}\text{Cs}$  were not in agreement; the indicated releases were 30.7%

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

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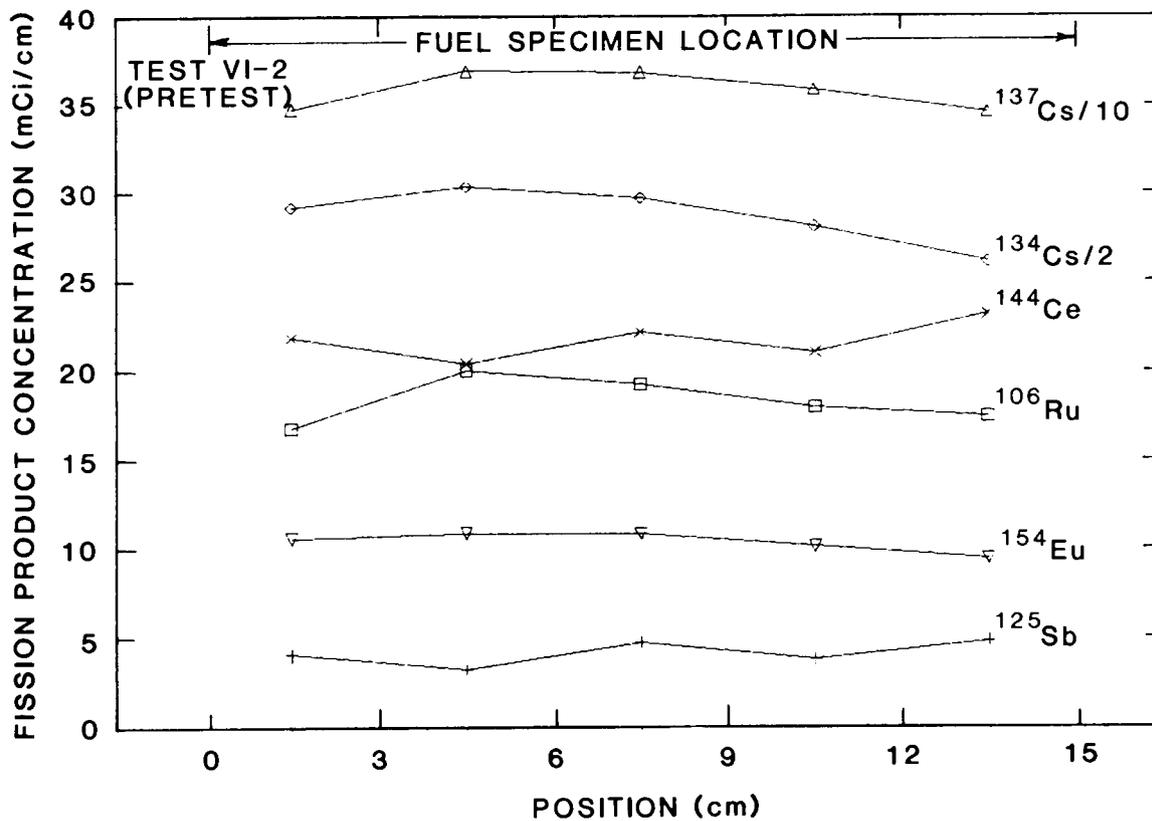


Fig. 8. Distribution of major fission products in fuel specimen before test VI-2.

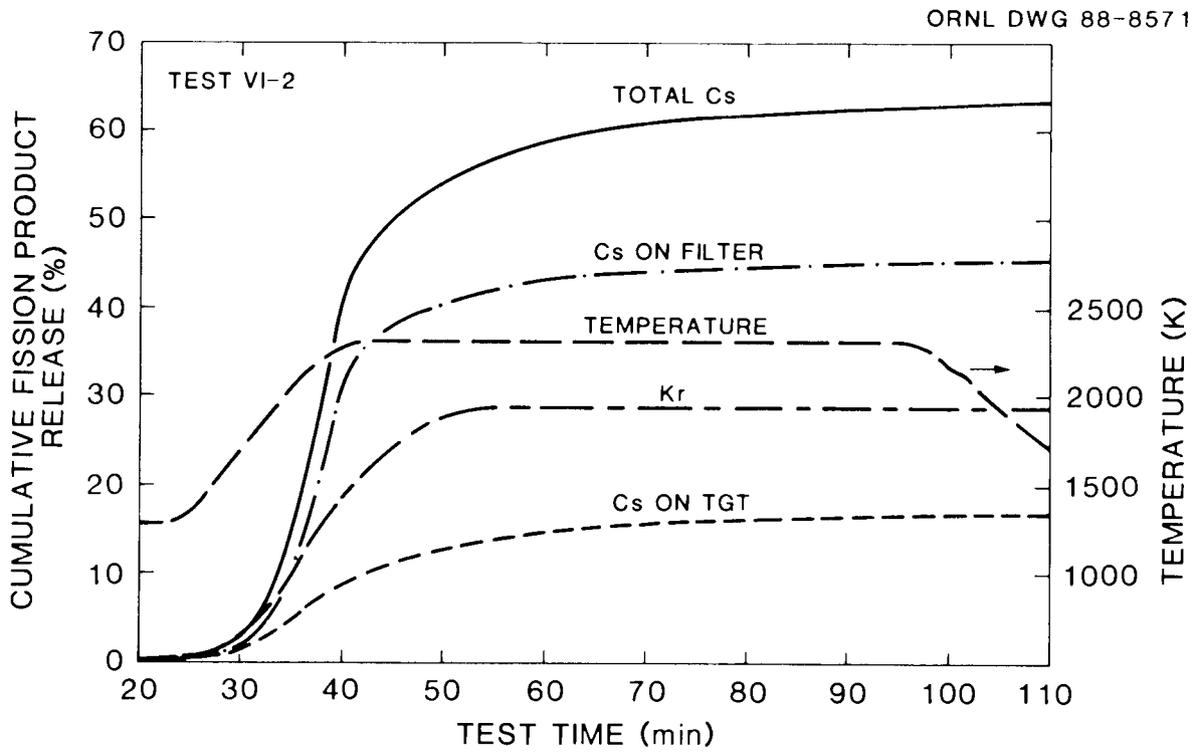


Fig. 9. Release behavior of krypton and cesium in test VI-2.

Table 8. Summary of release data for test VI-2

Component/collector	Operating time T >2000 K (min)	Percent of fission product inventory <sup>a</sup> found			
		<sup>85</sup> Kr	<sup>110m</sup> Ag	<sup>125</sup> Sb	<sup>137</sup> Cs
Furnace	70	0	5.65	16.43	2.71
Train A	7				
TGT			0.144	0.041	9.08
Filters			0.014	0.025	19.35
Total			0.158	0.066	28.43
Train B	18				
TGT			0.598	7.41	4.65
Filters			0.070	29.83	18.80
Total			0.668	37.24	23.45
Train C	45				
TGT			2.45	5.31	2.53
Filters			0	9.15	6.25
Total			2.45	14.46	8.78
Cold charcoal, condenser, etc.	70	28.72	0	0	0.003
Total for test	70	30.7 <sup>b</sup>	8.93	68.15	63.36

<sup>a</sup>Inventories based on pretest and posttest fuel analysis by gamma spectrometry; ORIGEN2 calculations were used to determine inventories of nuclides not gamma-measurable in the fuel (<sup>85</sup>Kr and <sup>129</sup>I).

<sup>b</sup>Includes an estimated 2% released during irradiation.<sup>17</sup>

and 63.4% of fuel inventory, respectively. However, it was discovered during posttest transfer of the <sup>85</sup>Kr that appreciable sorption on the Ascarite in the hydrogen and carbon monoxide collection apparatus had occurred (see Fig. 4). This sorption almost certainly resulted in a significant loss of krypton before it could be quantitatively analyzed. Therefore, we assume that only about half of the released <sup>85</sup>Kr was included in the final analysis, and that the release fractions of krypton and cesium were comparable, ~63%. Moreover, reexamination of the data for test VI-1 indicates that a similar krypton loss (~25%) probably occurred in that test also. On-line analysis indicated that the krypton release value, which includes an estimated 2% of inventory that was released from the UO<sub>2</sub> to the pellet-cladding gap during irradiation (as indicated by data collected from other comparable fuel rods) reached the

maximum value by the end of test phase B. Although no data for  $^{134}\text{Cs}$  are shown, the agreement between  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  was consistently good; the indicated total release values for both nuclides were ~63%. The distribution of cesium within the test apparatus is shown in detail in Table 9; the general distribution is illustrated in Fig. 10. As has been noted previously, the cesium tended to be released early in the test; the largest fraction was found in Train A, which included the heatup and only 2 min at test temperature. A similar amount of cesium was collected during the following 18 min period (Train B), but only approximately one-third as much was collected in Train C during the final 40 min at 2300 K. As usual, the cesium fractions collected on the filters were much larger than the fractions collected in the TGTs, an indication that the majority of the cesium was associated with the aerosol (either as particles or deposited on other particles) rather than being transported as vapor. The release data for iodine are discussed in the following section, 4.2.2.

The indicated release values for  $^{110\text{m}}\text{Ag}$  and  $^{125}\text{Sb}$ , which are fission products of much lower yield and are generally less hazardous than cesium and iodine, are shown in Table 10. The release of antimony (68.2%) appears to have been higher than in any previous test, whereas the release of silver (8.9%) is somewhat lower than in tests VI-1 and HI-5. The data for silver, however, are relatively sparse; this is at least partially a result of the longer decay time for the BR3 fuel in this test (~7 years), compared to the Oconee fuel in test VI-1 (~6 years), but it may also indicate a failure to find a significant fraction of the released silver. It should be noted that large fractions (approximately two-thirds and one-fourth, respectively) of the released silver and antimony were deposited on  $\text{ZrO}_2$  surfaces in the outlet region of the furnace, where temperatures were believed to be 1300 to 1500 K during the test.

The relative posttest distributions of fission product  $^{106}\text{Ru}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{144}\text{Ce}$ , and  $^{154}\text{Eu}$  in the fuel specimen are shown in Fig. 11. Note that these curves have not been corrected for self-shielding in the fuel, and the multipliers have been applied to the  $^{134}\text{Cs}$  and  $^{125}\text{Sb}$  curves to facilitate comparison of the five different elements. (The distribution of  $^{137}\text{Cs}$ , which is not shown, was essentially identical to that of  $^{134}\text{Cs}$ .) The curve for  $^{154}\text{Eu}$ , which is relatively involatile and has a high energy gamma ray useful for accurate analysis, should be the best indicator of  $\text{UO}_2$  position. It is significant that almost no change in the  $^{154}\text{Eu}$  distribution occurred during the test. The two curves, pretest and posttest distributions of europium, shown in Fig. 12, verify that no significant change in  $\text{UO}_2$  fuel distribution occurred. (The previously described empirical method for determining self-shielding was used to correct these data; as may be seen by comparison with Fig. 11, this correction resulted in an increase of ~25% for  $^{154}\text{Eu}$ .) As shown in Fig. 12, however, there was a major decrease in the magnitude and a small change in the shape of the  $^{134}\text{Cs}$  curve. The former (magnitude) reflects the large release of cesium, and the latter (shape) shows that somewhat more cesium was released from the lower half of the specimen, indicating a small axial temperature gradient. We calculate that a temperature

Table 9. Fractional release and distribution of cesium in test VI-2

Location	Approximate temperature (K)	Cesium found in each location			
		Amount		Percent of inventory <sup>b</sup>	Percent of released
		( $\mu$ Ci $^{137}\text{Cs}$ )	(mg total Cs) <sup>a</sup>		
<b>Furnace components</b>					
First ZrO <sub>2</sub> plug	~2000	1.582E+3 <sup>c</sup>	0.0451	0.01	
Second ZrO <sub>2</sub> plug	~1500	1.420E+5	4.051	1.665	
ZrO <sub>2</sub> donut	500-1000	4.630E+4	1.321	0.54	
Exit flange	350-1150	4.090E+4	1.167	0.479	
Total		2.308E+5	6.585	2.706	4.27
<b>Train A</b>					
TGT A	420-1130	7.749E+5	22.11	9.083	
TGT - filter line	430	1.043E+5	2.976	1.224	
First prefilter	410	6.714E+5	19.16	7.870	
Second prefilter	410	6.268E+5	17.88	7.347	
HEPA	410	2.479E+5	7.073	2.906	
Total		2.425E+6	69.19	28.43	44.87
<b>Train B</b>					
TGT B	420-1150	3.970E+5	11.33	4.654	
TGT - filter line	430	1.217E+5	3.472	1.427	
First prefilter	440	3.769E+5	10.75	4.418	
Second prefilter	440	1.027E+6	29.30	12.038	
HEPA	440	7.760E+4	2.214	0.910	
Total		2.000E+6	57.06	23.45	37.01
<b>Train C</b>					
TGT C	420-1140	2.155E+5	6.148	2.526	
TGT - filter line	430	5.440E+4	1.552	0.639	
First prefilter	410	2.040E+5	5.820	2.391	
Second prefilter	410	2.098E+5	5.986	2.459	
HEPA	410	6.560E+4	1.872	0.769	
Total		7.493E+5	21.38	8.783	13.86
Total in condenser, etc.		2.230E+2	0.006	0.003	0.004
Total found		5.406E+6	154.2	63.36	100.

<sup>a</sup>Based on analysis of  $^{137}\text{Cs}$  in the fuel and ORIGEN2 calculations of other cesium nuclides:  $2.853\text{E}-02 \text{ g Cs/Ci } ^{137}\text{Cs} = 28.53 \text{ mg Cs/Ci } ^{137}\text{Cs}$ .

<sup>b</sup>Measured  $^{137}\text{Cs}$  inventory in fuel before test was 8.531 Ci.

<sup>c</sup>Exponential notation:  $1.582\text{E}+3 = 1.582 \times 10^3$ .

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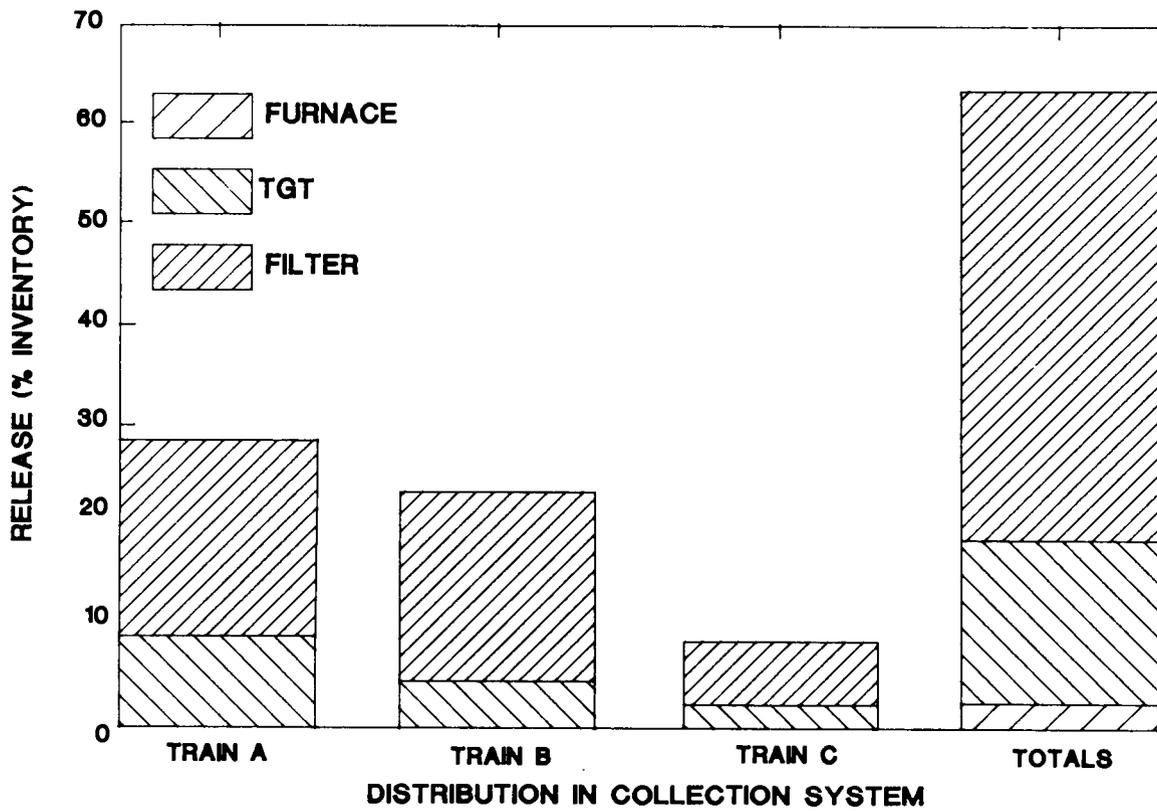


Fig. 10. Cesium release in test VI-2, based on gamma spectrometry.

Table 10. Fractional release and distribution of silver and antimony in test VI-2

Location	Fission products found			
	$^{110m}\text{Ag}$		$^{125}\text{Sb}$	
	( $\mu\text{Ci}$ )	(Percent of inventory) <sup>a</sup>	( $\mu\text{Ci}$ )	(Percent of inventory) <sup>a</sup>
Furnace components	2.996E+1 <sup>b</sup>	5.65	16.56E+3	16.38
Train A				
TGT	7.647E-1	0.144	41.E+0	0.041
Filters <sup>c</sup>	0.750E-1	0.014	25.E+0	0.025
Total	8.397E-1	0.158	66.E+0	0.066
Train B				
TGT	3.169E+0	0.598	7.49E+3	7.41
Filters <sup>c</sup>	0.373E+0	0.070	30.16E+3	29.83
Total	3.542E+0	0.668	37.65E+3	37.24
Train C				
TGT	12.97E+0	2.45	5.37E+3	5.31
Filters <sup>c</sup>	N.D.	0	9.25E+3	9.15
Total	12.97E+0	2.45	14.62E+3	14.46
Total test	4.731E+1	8.93	68.90E+3	68.15
Assumed inventory <sup>a</sup>	5.303E+2		1.011E+5	

<sup>a</sup>Inventories based on pretest analysis for  $^{125}\text{Sb}$ , and, for  $^{110m}\text{Ag}$ , on ORIGEN2 calculation corrected by pretest analysis for  $^{137}\text{Cs}$ .

<sup>b</sup>Exponential notation:  $2.996\text{E}+1 = 2.996 \times 10^1$ .

<sup>c</sup> $^{110m}\text{Ag}$  not generally found on filters, but was found on some lines between TGTs and filters.

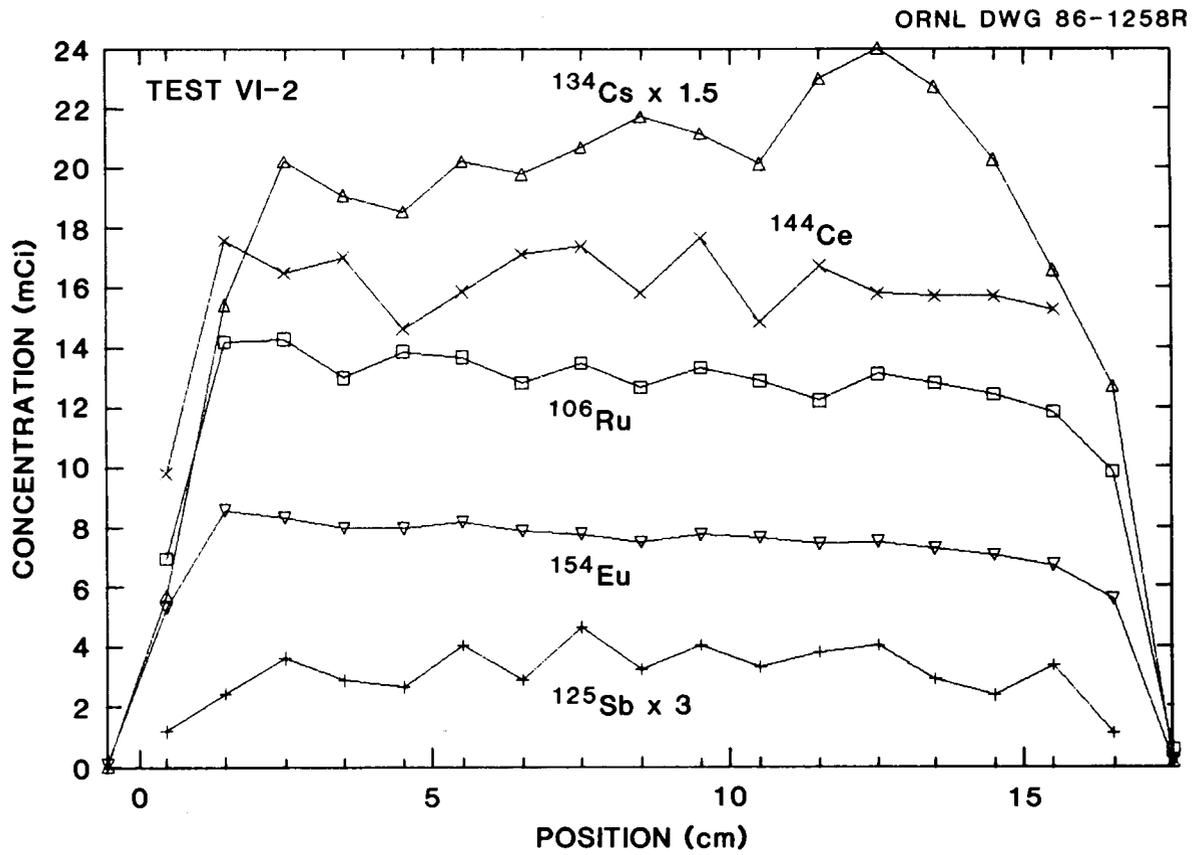


Fig. 11. Concentration of fission products along length of fuel specimen after test VI-2.

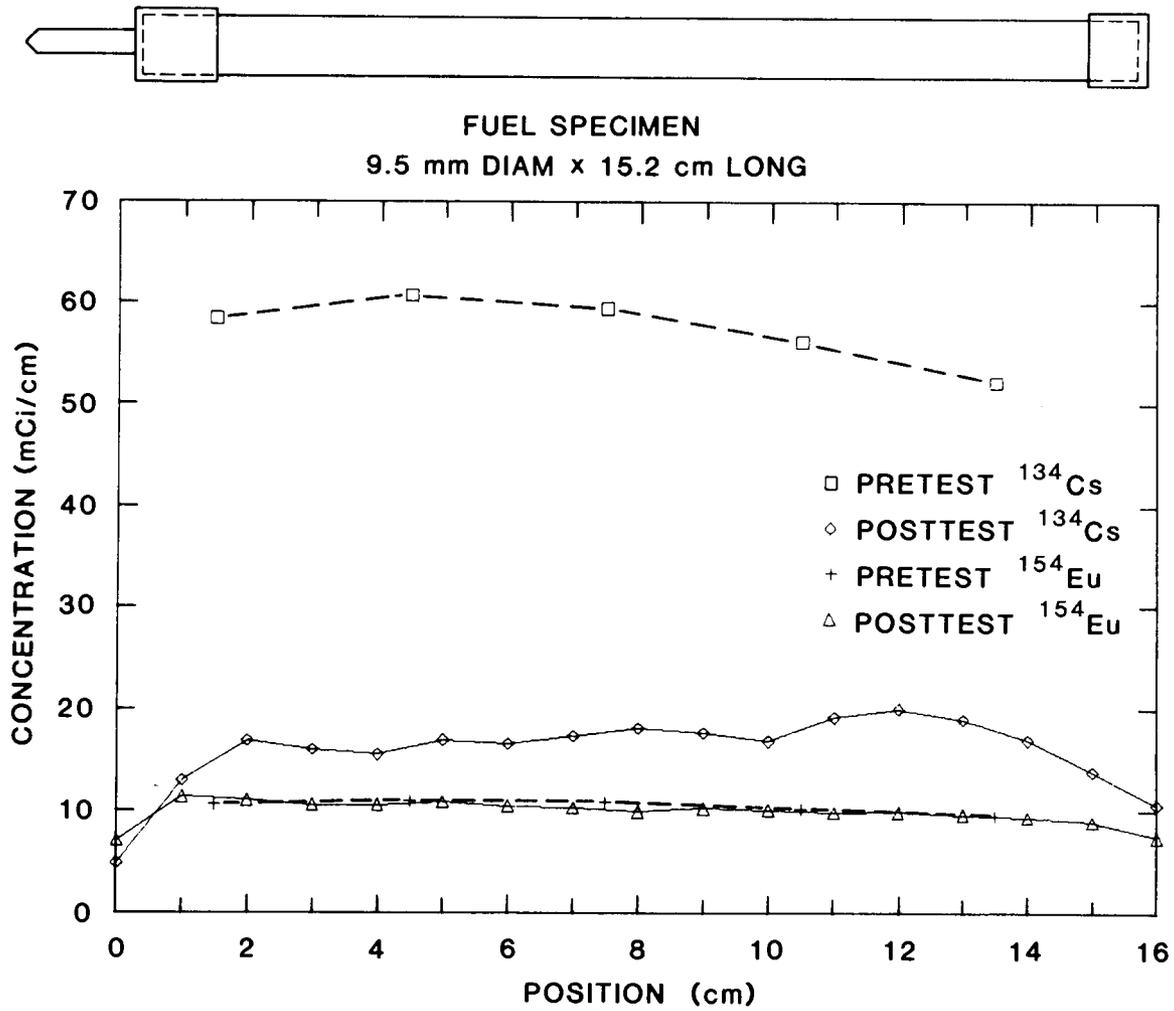


Fig. 12. Changes in distribution of  $^{134}\text{Cs}$  and  $^{154}\text{Eu}$  in fuel in test VI-2.

difference of ~50 K, which is consistent with our temperature calibration of the furnace, would be sufficient to cause this difference in release.

#### 4.2.2 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  $\text{NH}_4\text{OH} + \text{H}_2\text{O}_2$  leach solution. Small aliquots of the solutions were chemically treated to remove cesium, loaded onto ion exchange media, irradiated, and the 12.5-h  $^{130}\text{I}$  was counted. As a result of sample loss in some of the initial analyses and delays in obtaining neutron activation of the second set of samples, the results of iodine analyses for this test are incomplete at this time. Consequently, iodine release for test VI-2 will be discussed in a later report. Final, complete data for the iodine collected on the charcoal cartridges downstream from the filters showed that 63  $\mu\text{g}$   $^{129}\text{I}$  was found on all of these collectors, with most of this, 60.3  $\mu\text{g}$ , located on the first cartridge in Train A. This iodine on the charcoal is that fraction of released iodine that was in a volatile form (i.e.,  $\text{I}_2$ ,  $\text{HI}$ , or organic iodides). As observed and discussed previously, in steam atmosphere tests a small amount of iodine is always found on the charcoal, but the total has never exceeded 100  $\mu\text{g}$ , nor has it ever exceeded 0.5% of the released iodine in tests with I releases  $>100 \mu\text{g}$ .<sup>20</sup> The available results indicate that the final results for this test will be consistent with this established pattern of behavior, and that the iodine found on the charcoal will be  $<0.5\%$  of that released.

#### 4.2.3 Spark Source Mass Spectrometry

Small smear samples of the deposits on the three TGTs were collected on graphite electrodes and were analyzed by spark source mass spectrometry (SSMS). Gamma scans of the TGTs, which showed the distributions of  $^{137}\text{Cs}$  and  $^{125}\text{Sb}$ , were used to determine the sample locations most likely to provide interesting data. Assuming that the smear samples were representative of the deposits at that location, the easily measured  $^{137}\text{Cs}$  (by gamma spectrometry) was used to quantify the amounts of the other elements in the TGT — fission products, structural materials, and impurities — vaporized in the mass spectrometer. Data from the filters were normalized to the total mass determined by weighing. This technique has the advantage of providing data for a wide variety of elements, most of which are not radioactive and are not readily measured otherwise. The major disadvantage of this technique is that it is not highly accurate; precision is generally considered to be about a factor of 2.

Samples of the deposits from several TGT sections and from the filters from each collection train were analyzed. In addition to Cs, the fission product elements Se, Br, Rb, Mo, Pd, Ag, Cd, Sb, Te, I, and Ba were detected in one or more samples. The amounts for silver and

palladium were too great to be fission products only; apparently, some normal silver and palladium were either present in the system or the samples were contaminated during handling or analysis, thereby confusing the results. These data are summarized in Tables 11 and 12, which show the approximate masses and the mass fractions, respectively. The results are divided into three categories: fission products, reactor materials (elements normally included in the construction of some LWRs), and other materials, which would not normally be found in a reactor system.

Unfortunately, these data do not appear to be as consistent as those in some previous tests and are difficult to interpret. Apparently, unavoidable problems with instrument operation and/or changes in analytical techniques were a major factor in these inconsistencies. As shown in Table 11, molybdenum and tellurium were the most abundant fission product elements in these samples; the maximum concentrations of both were found in Train B, whereas the maximum concentrations of the more volatile cesium and rubidium were found in Train A. Because the oxides of molybdenum are much more volatile than the element, this high release (compared to earlier tests) would be expected during the extended time at the highly oxidizing conditions of this test. Section BT4 of TGT B contained unusually high amounts of several fission products (Te, Cd, Se, and Ba) as well as of U and Sn. These and other data will be reevaluated after some additional analyses.

Of the cladding metals, relatively large amounts of Sn were released throughout the test, while the release rates for the more abundant but much less volatile zirconium were generally lower. Small and relatively uniform amounts of cesium and magnesium, which are stabilizers in the  $ZrO_2$  ceramics, were collected also. Significant amounts of uranium were found during all three test phases, with the largest amounts on the filters from phases B and C. These masses on the Train B filters appear to be much greater, however, than the uranium masses indicated by the more accurate fluorimetric analysis of leach solutions, as shown in Sect. 4.2.5. Following further analysis, these data will be discussed in a subsequent report.

Although a number of impurity elements (Al, B, Na, Cu, V, Cl, etc.) were detected, the amounts were low in comparison to the fission product and fuel/furnace materials. As shown in Table 11, the most abundant impurities were steel components — iron, manganese, and chromium — which are likely to be contaminants in any reactor system. These data show that our efforts to reduce the level of impurities, particularly those that would be atypical of an LWR system, have been successful.

#### 4.2.4 Thermal Gradient Tube Deposits

The  $^{137}Cs$  profiles and the temperature profiles for the period of collection in the three TGTs are shown in Figs. 13 to 15. In addition, smaller amounts of  $^{125}Sb$  were found throughout TGTs B and C, and very low levels of  $^{110m}Ag$  were found on the first 8 cm of TGT C. It should be noted (see also Table 8) that the highest concentrations of  $^{137}Cs$  were

Table 11. Test VI-2 SSMS data<sup>a</sup>

Temp. (°C)	Thermal gradient tube sections <sup>b</sup>								Filters <sup>c</sup>			
	AT2 (mg/cm)	AT3 (mg/cm)	BT1 (mg/cm)	BT2 (mg/cm)	BT4 (mg/cm)	CT1 (mg/cm)	CT2 (mg/cm)	CT4 (mg/cm)	AG2c (mg)	BG1c (mg)	BG2c (mg)	CG1c (mg)
	600	530	860	580	190	840	620	310	125	170	170	135
Fission products <sup>d</sup>												
Mo	0.56	0.05	1.4	0.18	4.1	8.7	0.43	0.41	3.6	9.4	38	17
Te	3.0	2.0	0	0.04	6.1	0	0	0	6.6	10.0	22	1.9
Cs	2.0	1.1	0.72	0.27	0.37	0.12	0.19	0.18	13.0	4.3	10	3.1
Rb	2.5	1.96	0.41	0.08	0.32	0.1	0.13	0.07	1.8	0.96	1.5	1.0
Cd	0.13	0.17	0.51	0.12	0.58	0.1	0.13	0.05	2.5	1.1	3.0	0.86
I	0.25	0.17	0	0	0.08	0	0	0	1.4	0	0.32	0.28
Br	0.17	0.1	0	0.39	0.08	0.1	0	0.05	0.08	0.56	0	0
Se	0.17	0.04	0.62	0.12	0.73	0.05	0	0.05	0.72	0.4	0.72	0
Ba	0.04	0	0	0	0.32	0	0.04	0.02	0.24	0.4	1.7	0.68
Ag <sup>e</sup>	0.42	0.1	0.51	0.08	0.32	0.05	0.43	0.23	0.96	0.56	1.7	2.1
Pd <sup>e</sup>	0	0	0	0	0	0	0	0	0	0	14	21
Total	9.2	5.7	4.2	1.3	13.0	9.2	1.4	1.1	31	28	93	48
Reactor materials <sup>d</sup>												
Sn	1.4	0.1	0.1	0.08	15.0	1.6	0	2.4	17	52	85	12
Zr	1.1	0.04	1.0	0.2	0.02	0	0.43	0.26	0.56	0.72	1.8	3.8
U	0.83	0.1	0	0	3.3	1.0	0.21	0.23	0	5.6	17	6.9
Fe	0.83	0.15	0.51	0.39	0.49	0.15	0.04	0.12	0.72	0.96	3.4	1.4
Cr	0.21	0.02	0.31	0.2	0.16	0.05	0.04	0.12	0.72	0.56	0.48	0.69
Ni	0.13	0.1	0.1	0.04	0.32	0.5	0	0.02	0.24	0.16	0.72	0.21
Mn	0.13	0.05	0.31	0.2	0.08	0.15	0.04	0.07	1.2	0.56	0.16	0.69
Al	0.04	0.1	0	0.04	0.02	0.15	0.04	0.05	0	0.16	0.16	0
Si	0.42	0.01	0	0.04	0.16	0.5	0.04	0.07	0.16	0.16	0.08	0
B	0.13	0.02	0	0	0.02	0	0	0	0.16	0	0	0.07
Total	5.3	0.69	2.4	1.2	20.0	4.1	0.84	3.3	21	62	110	26
Other materials <sup>d</sup>												
Na	0.13	0.05	0	0.04	0.16	0.15	0	0.02	0.24	0.4	0.72	0.34
Cu	0.13	0.02	0.21	0.12	0.16	0.05	0.04	0.05	0.48	0.56	0.32	0.69
Mg	0.04	0.01	0	0.04	0	0.05	0.04	0.07	0	0.16	0	0
Ca	0.42	0.1	0.1	0.04	0.16	0.5	0.04	0.12	0.48	0.16	0.32	0.34
V	0.13	0.02	0.21	0.12	0.05	0.05	0.04	0	0.48	0.4	0.16	0
P	0.08	0.05	0	0.04	0.16	0.05	0	0	0.48	0.16	0.32	0.14
Cl	0.42	0.15	0.1	0.04	0.08	0.05	0	0.05	0.24	0.16	0.16	0.14
Zn	0.13	0.05	0.1	0.04	0.02	0	0	0.02	0.16	0.16	0.16	0.07
K	0.21	0.05	0.21	0.12	0.08	0.15	0.04	0.07	0.08	0.16	0.16	0.69
S	0.42	0	0	0.04	0.16	0	0	0.23	0.48	0.16	0.08	0.07
Total	2.1	0.5	0.93	0.64	1.0	1.1	0.2	0.63	3.1	3	2.4	2.5
Total all	17	6.9	7.5	3.1	34	14	2.4	5	55	93	210	77
Filter mass <sup>f</sup>								69	115	255	96	
Train mass <sup>f</sup>								245		573	316	

<sup>a</sup>Precision of all data about a factor of 2.<sup>b</sup>Based on <sup>137</sup>Cs.<sup>c</sup>Normalized to total mass on filters, corrected for oxygen (0.8).<sup>d</sup>In approximate order of abundance.<sup>e</sup>Uncertain mixture of fission product and naturally occurring elements.<sup>f</sup>Determine by weighing, Sect. 4.2.6.

Table 12. Test VI-2 Elemental distribution, from SSMS data<sup>a</sup>

	Thermal gradient tube sections <sup>b</sup>								Filters <sup>c</sup>			
	AT2 (%)	AT3 (%)	BT1 (%)	BT2 (%)	BT4 (%)	CT1 (%)	CT2 (%)	CT4 (%)	AG2c (%)	BG1c (%)	BG2c (%)	CG1c (%)
Temp. (C)	600	530	860	580	190	840	620	310	125	170	170	135
Fission products <sup>d</sup>												
Mo	3.30	0.72	19.00	5.80	12.00	62.00	18.00	8.20	6.55	10.11	18.10	22.08
Te	18.00	29.00	0.00	1.30	18.00	0.00	0.00	0.00	12.00	10.75	10.48	2.47
Cs	12.00	16.00	9.60	8.70	1.10	0.86	8.00	3.60	23.64	4.62	4.76	4.03
Rb	15.00	28.00	5.50	2.60	0.94	0.71	5.40	1.40	3.27	1.03	0.71	1.30
Cd	0.76	2.50	6.80	3.90	1.70	0.71	5.40	1.00	4.55	1.18	1.43	1.12
I	1.50	2.50	0.00	0.00	0.24	0.00	0.00	0.00	2.55	0.00	0.15	0.36
Br	1.00	1.50	0.00	13.00	0.24	0.71	0.00	1.00	0.15	0.60	0.00	0.00
Se	1.00	0.58	8.30	3.90	2.20	0.36	0.00	1.00	1.31	0.43	0.34	0.00
Ba	0.24	0.00	0.00	0.00	0.94	0.00	1.70	0.40	0.44	0.43	0.81	0.88
Ag <sup>e</sup>	2.50	1.50	6.80	2.60	0.94	0.36	18.00	4.60	1.75	0.60	0.81	2.73
Pd <sup>e</sup>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	6.67	27.27
Total	55.00	82.00	56.00	42.00	38.00	66.00	57.00	21.00	56.00	30.00	44.00	62.00
Reactor materials <sup>d</sup>												
Sn	8.20	1.50	1.30	2.60	44.00	11.00	0.00	48.00	31.00	56.00	40.00	16.00
Zr	6.50	0.58	13.00	6.50	0.06	0.00	18.00	5.20	1.00	0.77	0.86	4.90
U	4.90	1.50	0.00	0.00	9.70	7.10	8.80	4.60	0.00	6.00	8.10	9.00
Fe	4.90	2.20	6.80	13.00	1.40	1.10	1.70	2.40	1.30	1.00	1.60	1.80
Cr	1.20	0.29	4.10	6.50	0.47	0.36	1.70	2.40	1.30	0.60	0.23	0.90
Ni	0.76	1.50	1.30	1.30	0.94	3.60	0.00	0.40	0.44	0.17	0.34	0.27
Mn	0.76	0.72	4.10	6.50	0.24	1.10	1.70	1.40	2.20	0.60	0.08	0.90
Al	0.24	1.50	0.00	1.30	0.06	1.10	1.70	1.00	0.00	0.17	0.08	0.00
Si	2.50	0.14	0.00	1.30	0.47	3.60	1.70	1.40	0.29	0.17	0.04	0.00
B	0.76	0.29	0.00	0.00	0.06	0.00	0.00	0.00	0.29	0.00	0.00	0.09
Total	31.00	10.23	31.00	38.00	58.00	29.00	35.30	67.00	38.00	65.00	52.00	33.00
Other materials <sup>d</sup>												
Na	0.76	0.72	0.00	1.30	0.47	1.10	0.00	0.40	0.44	0.43	0.34	0.44
Cu	0.76	0.29	2.80	3.90	0.47	0.36	1.70	1.00	0.87	0.60	0.15	0.90
Mg	0.24	0.14	0.00	1.30	0.00	0.36	1.70	1.40	0.00	0.17	0.00	0.00
Ca	2.50	1.50	1.30	1.30	0.47	3.60	1.70	2.40	0.87	0.17	0.15	0.44
V	0.76	0.29	2.80	3.90	0.15	0.36	1.70	0.00	0.87	0.43	0.08	0.00
P	0.47	0.72	0.00	1.30	0.47	0.36	0.00	0.00	0.87	0.17	0.15	0.18
Cl	2.50	2.20	1.30	1.30	0.24	0.36	0.00	1.00	0.44	0.17	0.08	0.18
Zn	0.76	0.72	1.30	1.30	0.06	0.00	0.00	0.40	0.29	0.17	0.08	0.09
K	1.20	0.72	2.80	3.90	0.24	1.10	1.70	1.40	0.15	0.17	0.08	0.90
S	2.50	0.00	0.00	1.30	0.47	0.00	0.00	4.60	0.87	0.17	0.04	0.09
Total	12.00	7.30	12.00	21.00	3.00	7.59	8.30	13.00	5.70	2.70	1.10	3.20
Total all	98.00	99.53	99.00	101.00	99.00	102.59	100.60	101.00	99.70	97.70	97.10	98.20

<sup>a</sup>Precision of all data about factor of 2.<sup>b</sup>Based on <sup>137</sup>Cs.<sup>c</sup>Normalized to total mass on filters, corrected for oxygen (\*0.8).<sup>d</sup>In approximate order of abundance.<sup>e</sup>Uncertain mixture of fission product and naturally occurring elements.

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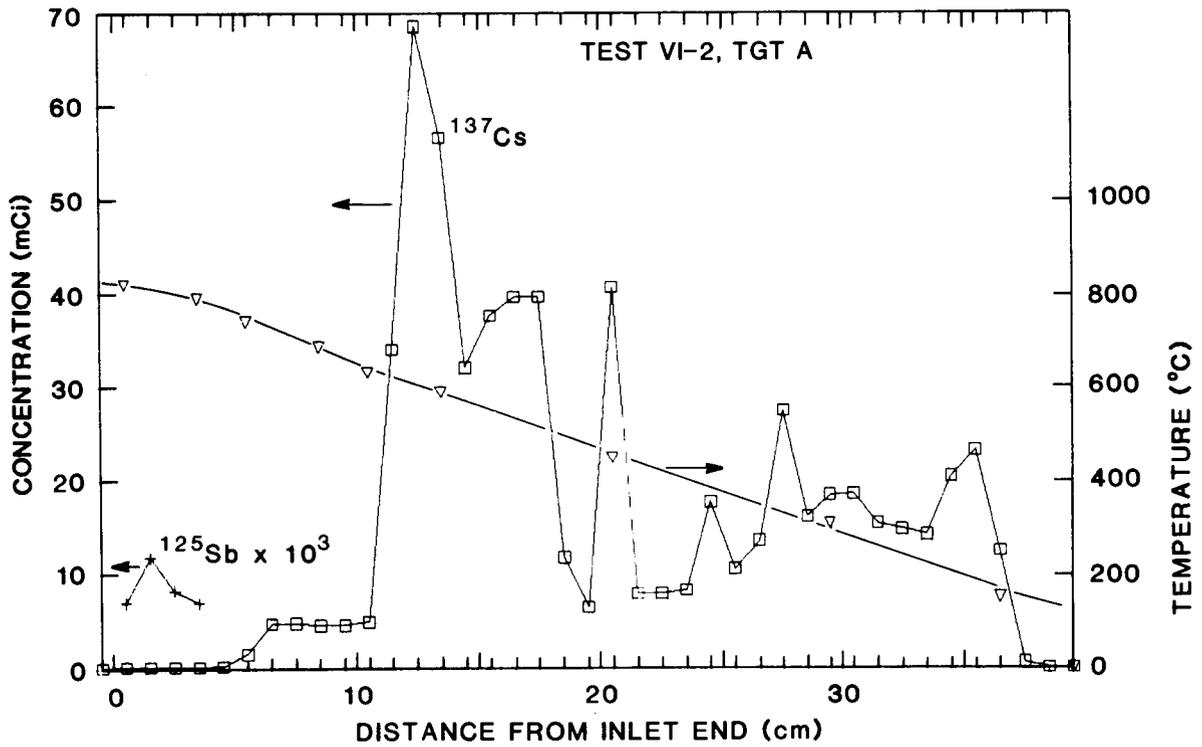


Fig. 13. Distribution of cesium and antimony in thermal gradient tube A.

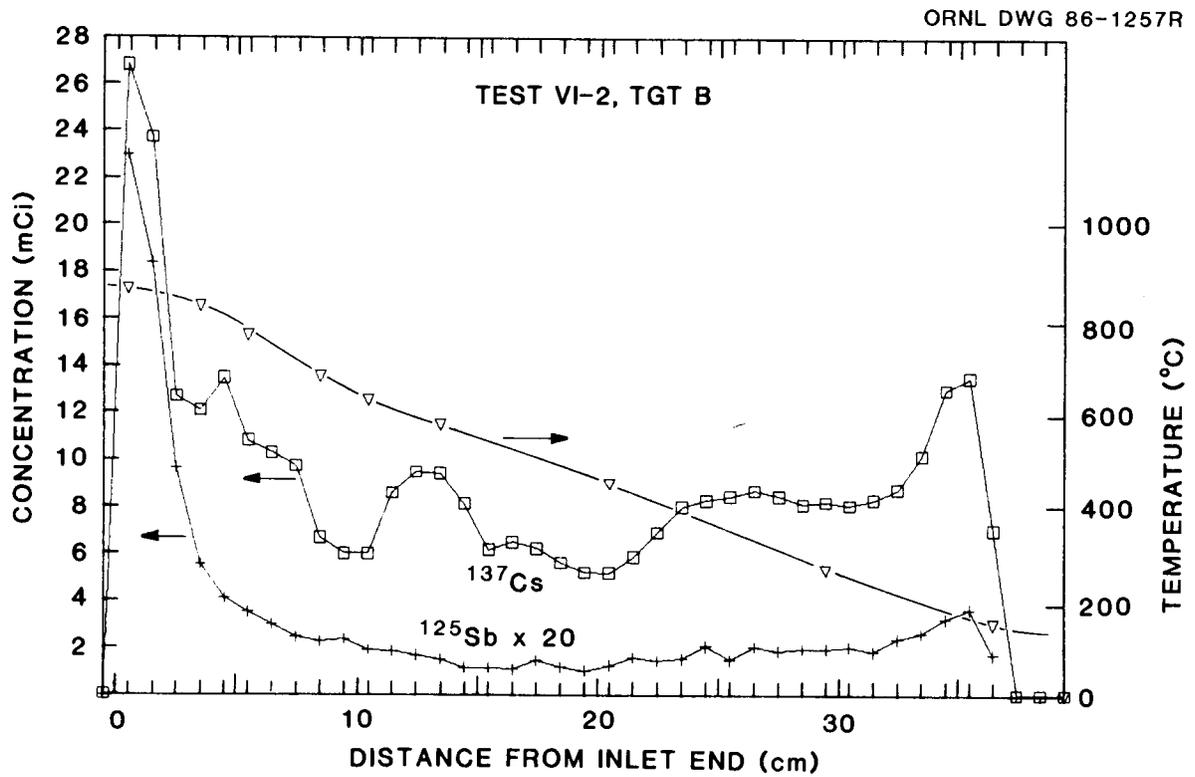


Fig. 14. Distribution of cesium and antimony in thermal gradient tube B.

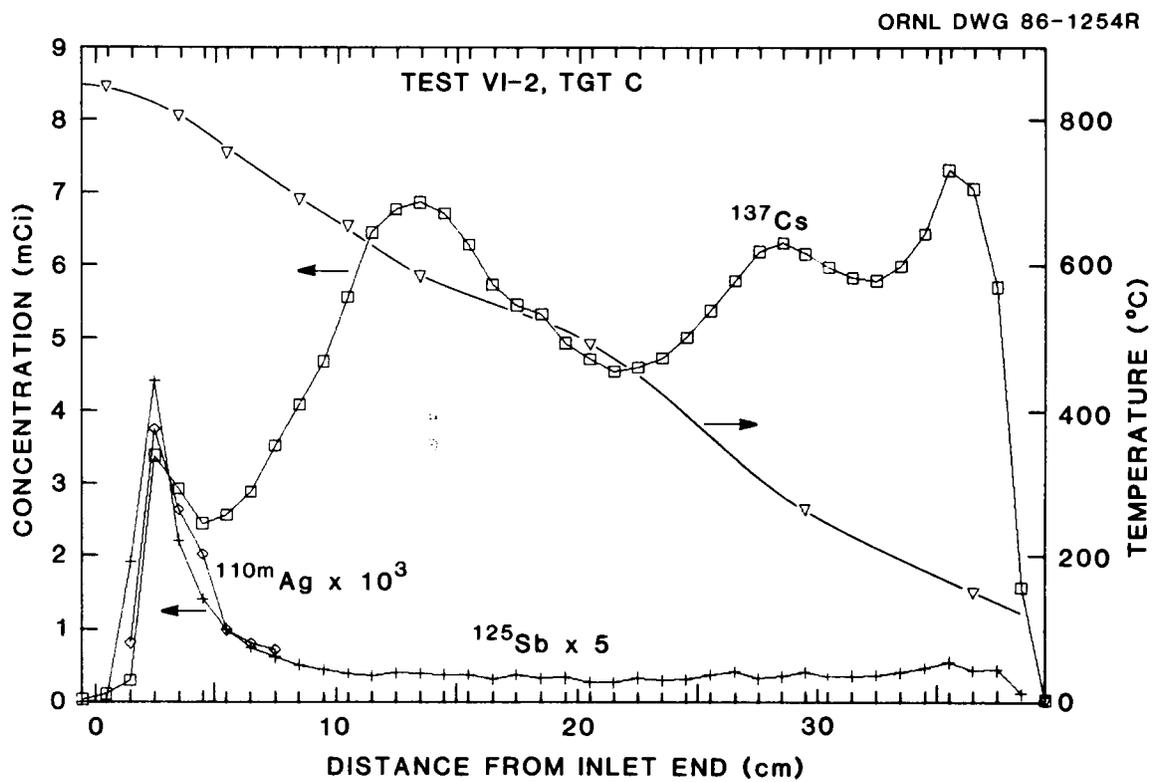


Fig. 15. Distribution of cesium, antimony, and silver in thermal gradient tube C.

found in TGT A (Fig. 13), and that this concentration was located in the region of the "characteristic" cesium peaks observed in previous tests at ~500 to 600°C (773 to 873 K).<sup>20</sup> Small <sup>137</sup>Cs peaks were observed near the outlet end of all three TGTs, possibly resulting from flow disturbances at the transition connection. Narrow <sup>137</sup>Cs and <sup>125</sup>Sb peaks occurring in the highest temperature region near the inlet of TGT B (Fig. 15) have not been seen in previous tests and cannot be explained at this time. It may be speculated that the extended period at very high temperature under highly oxidizing conditions resulted in some different chemical forms compared with previous tests. In order to investigate the fine structure of these fission product depositions in the TGTs, slow continuous scans with a very narrow collimator window (0.25 to 0.50 mm) were made. The very narrow structure of some of the peaks (probably mostly cesium) in TGT A is shown in Fig. 16. The other TGTs (B and C) did not exhibit such narrow peaks.

Earlier work indicated that, under these test conditions, most of the fission product iodine would combine with cesium, forming CsI, and that most of the remaining cesium would be in the form CsOH. In our TGTs, such compounds should deposit in the temperature range 500 to 600°C, as observed in Fig. 13. The high temperature peak in TGT B (Fig. 14) indicates the presence of a form of cesium having a deposition temperature of ~850°C (1123 K), but no identification has been possible. In earlier work, Norwood suggested that such compounds as Cs<sub>2</sub>SO<sub>4</sub> and Cs<sub>2</sub>S, (sulfur occurs as an impurity in the structural ceramics), Cs<sub>2</sub>MoO<sub>4</sub> (from fission product molybdenum), and Cs<sub>2</sub>ZrO<sub>4</sub> (from either cladding or fission product zirconium) might deposit in this temperature range.<sup>21</sup> The limited data for <sup>125</sup>Sb indicate that the deposits near the TGT inlets were of the elemental form, as suggested by Norwood, whereas the small amounts of <sup>125</sup>Sb found throughout the TGT (Fig. 15) were associated with and deposited with aerosol particles.<sup>21</sup> Unfortunately, the occasional deposits of antimony (a very low yield fission product) and iodine found in the SSMS analyses of TGT samples (see Table 10) were insufficient for correlation of these data with the SSMS data (Sect. 4.2.3).

The platinum TGT liners were cut into sections of 7 to 11 cm in length and opened to permit visual examination and the collection of smear samples for SSMS analysis. The sections were leached with basic solution (NH<sub>4</sub>OH and H<sub>2</sub>O<sub>2</sub>) to remove iodine from the surface and to retain it in solution. Subsequent leaches with acidic solution (HNO<sub>3</sub> and HF) removed other surface deposits. These chemical treatments removed ~99% of the highly radioactive <sup>134</sup>Cs and <sup>137</sup>Cs, allowing more accurate gamma spectrometric analysis of radionuclides that had alloyed with the platinum, presumably from the elemental vapor phase. The distributions of <sup>137</sup>Cs, <sup>125</sup>Sb, and <sup>110m</sup>Ag, along the three TGT liners after leaching, are shown in Figs. 17-19. The log of the average linear concentration (mg/cm) of each fission product on each TGT section is plotted vs distance from the TGT inlet in these figures. Although there is appreciable scatter of the data, it is interesting to note that the results for all three elements show a similar rate of decrease in concentration along the TGT; the diagonal dashed line on each figure indicates

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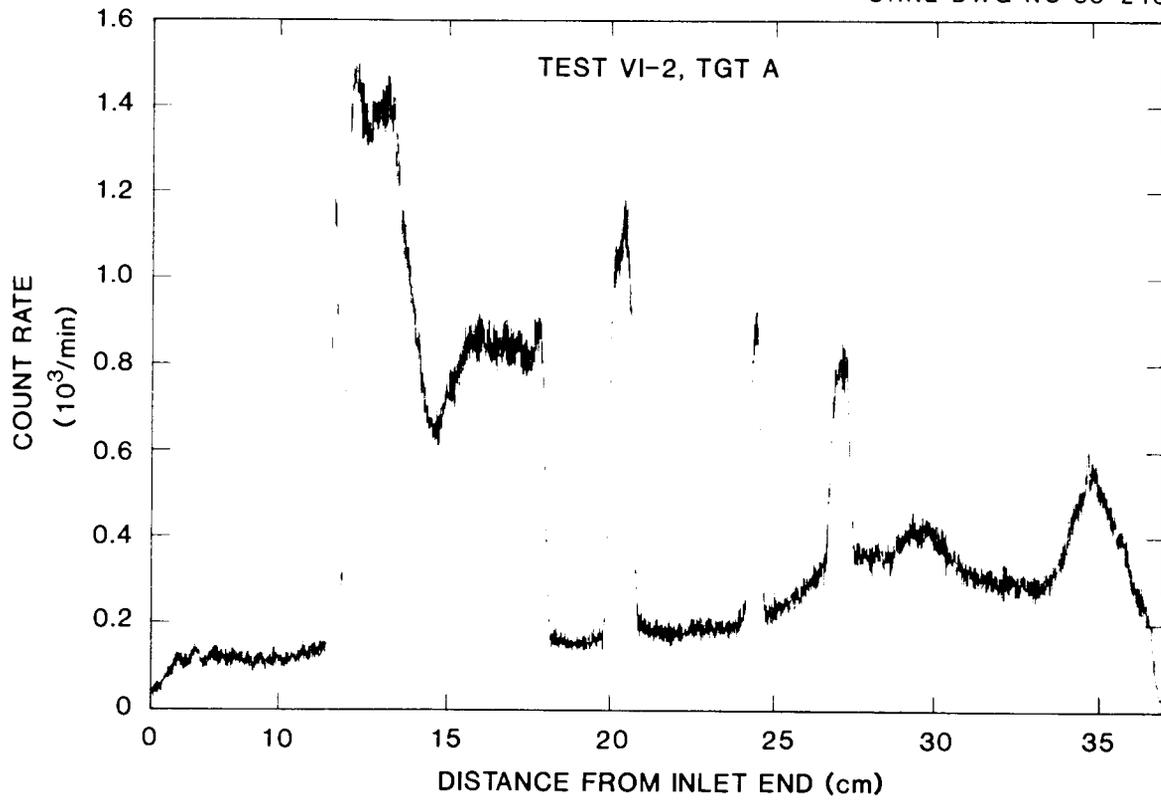


Fig. 16. Detailed distribution of total radioactivity (mostly cesium) along TGT A, in test VI-2. Compare with 1-cm measurements shown in Fig. 13.

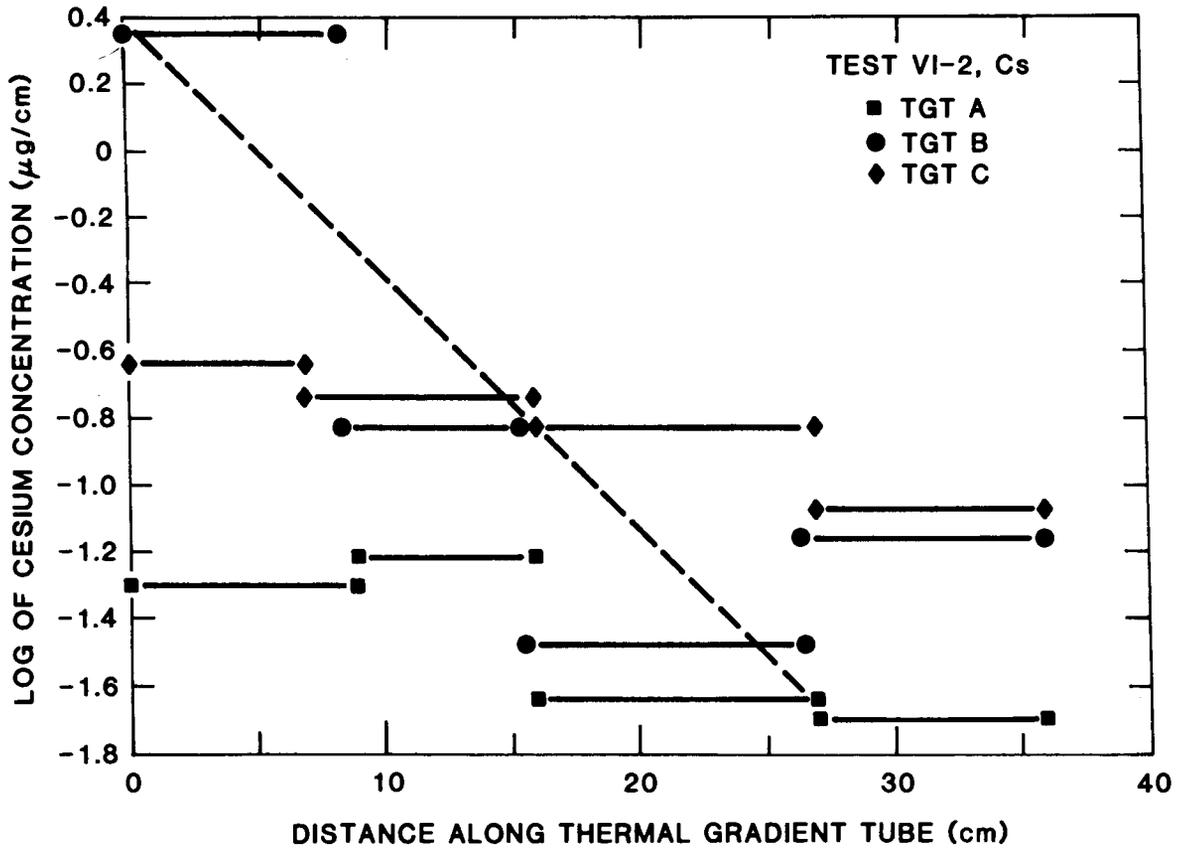


Fig. 17. Distributions of cesium in platinum TGT liners after leaching to remove surface deposits.

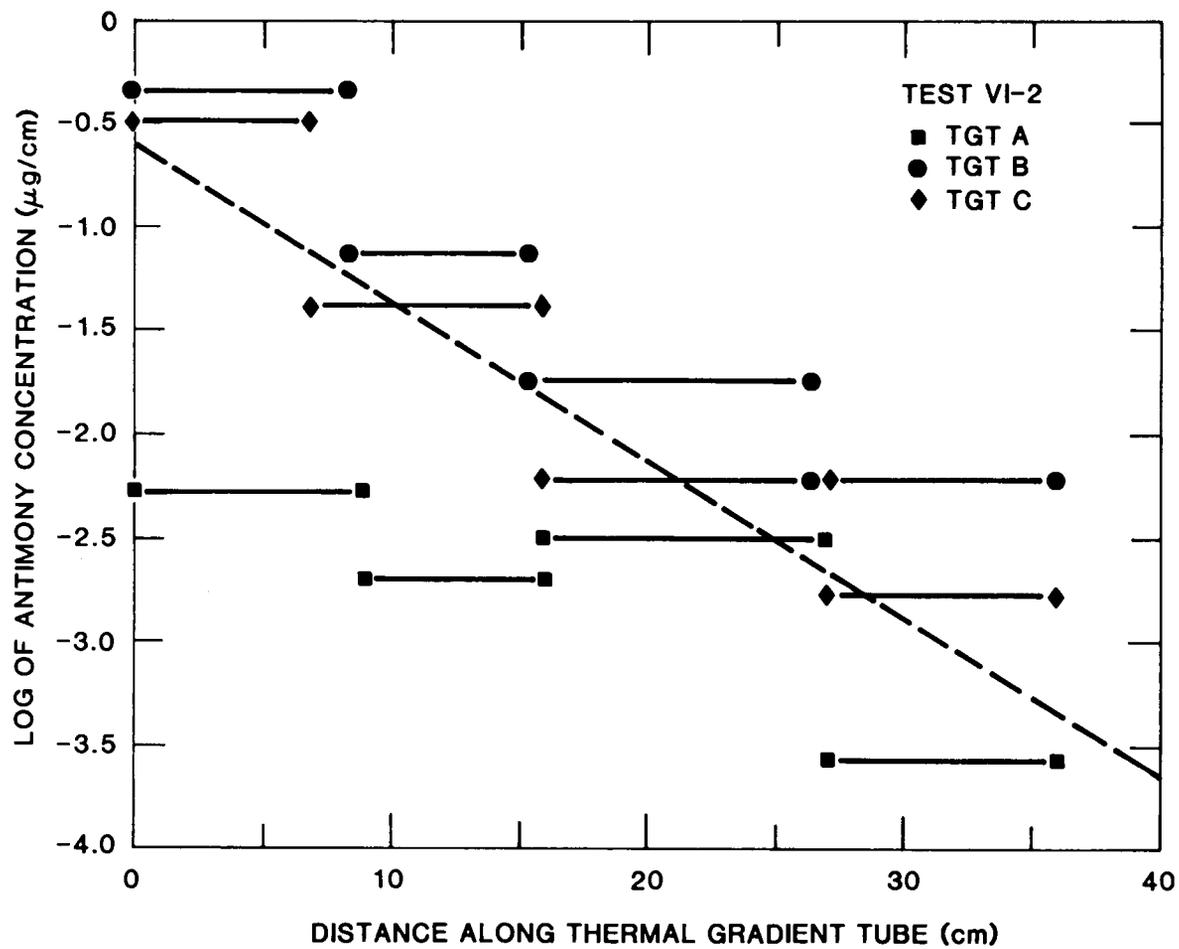


Fig. 18. Distributions of antimony in platinum TGT liners after leaching to remove surface deposits.

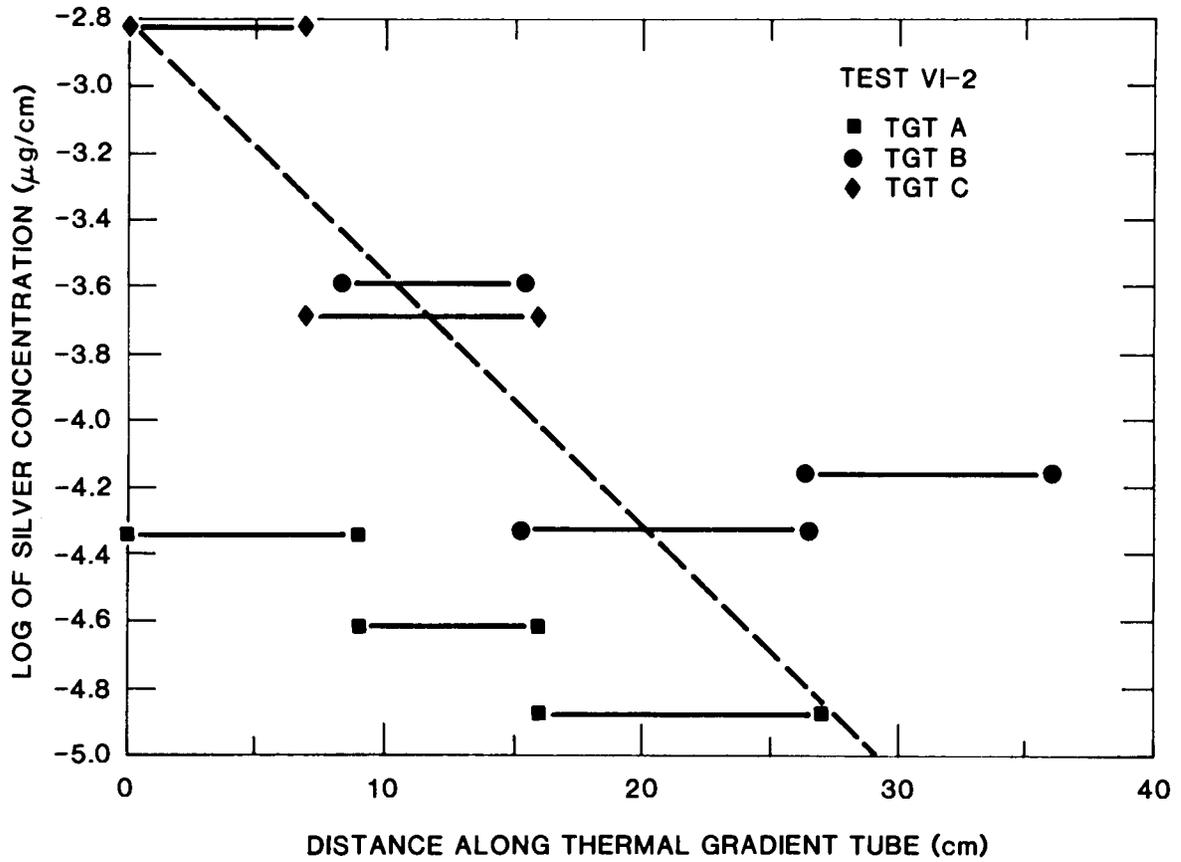


Fig. 19. Distributions of silver in platinum TGT liners after leaching to remove surface deposits.

a factor of 1000 decrease in concentration over 40 cm, which is somewhat greater than the 36 cm length of the tube. This type of behavior (logarithmic decrease with length along the tube) suggests that this fraction of the material entered the TGT as the elemental vapor, condensed and was dissolved into the platinum surface, and was retained in the matrix, rendering it inaccessible to the acidic leach solution.

#### 4.2.5 Uranium Release

Since the release and transport of fuel material (uranium and plutonium) provides a potential mechanism for the release of low-volatility fission products in addition to the direct hazard of the heavy metals, various samples were collected and analyzed for uranium and plutonium. These data, obtained by fluorimetric analysis for uranium and by alpha particle analysis for plutonium, are summarized in Table 13 and illustrated in Fig. 20. Based on the deposits in the collection trains, very little of the uranium and plutonium released from the fuel occurred during the short period (2 min) of test phase A. In fact, the mass of fuel material released during each test phase was almost directly proportional to the time of that phase, as would be expected if the release were controlled entirely by the rate of evaporation. A significant fraction of the uranium released from the fuel (~42%) was deposited on the ceramics in the outlet end of the furnace. The total mass of uranium released was 17.7 mg, or 0.026% of the uranium inventory in the fuel specimen. The corresponding values for plutonium were 15.0  $\mu\text{g}$ , or 0.0025% of the inventory. Larger release fractions would be expected at the higher temperatures planned for future tests. Some additional release of uranium to the TGTs was indicated by SSMS data; the total amounts, however, are uncertain at this time and additional analyses are being obtained.

#### 4.2.6 Masses of Deposits in TGT and on Filters

Each filter and TGT liner was weighed before and after the test to determine the mass of material collected during the test. Immediately after disassembly of the filter packages, the filters were inspected and photographed, then packaged for weighing. Both prefilters and the first HEPA filter exhibited deposits, but no deposits were visible on the last HEPA filter, indicating efficient collection of the aerosols by the previous filters. The masses of material collected at the various locations are listed in Table 14 and illustrated in Fig. 21. The greatest mass was released during the intermediate 18-min period at high temperature (i.e., it was collected on Train B).

Most of the material deposited in the TGTs is believed to be a result of vapor condensation, and the material deposited on the filters probably was transported primarily as aerosol particles. The average mass concentration of vapor and aerosol during each test phase was calculated from the masses collected and the total gas flow during that period, as shown in Table 14. (To avoid the distortion of the heatup and cooldown periods, it was assumed that no aerosol was formed at temperatures below 2000 K.) These data show that the average mass concentration

Table 13. Distribution of uranium and plutonium in test VI-2 apparatus

Location/component	Mass ( $\mu\text{g}$ )		Mass released per test phase ( $\mu\text{g}$ ) <sup>a</sup>		Release from fuel (ppm) <sup>b</sup>	
	U	Pu	U	Pu	U	Pu
<b>Furnace:</b>						
ZrO <sub>2</sub> plugs	7363	3.36				
Exit flange	19.3	0.27				
<b>Train A:</b>						
Filter supply line	58.5	0.56				
Filters	267.9	0.60				
Total			559.6	1.53	8.2	2.6
<b>Train B:</b>						
Filter supply line	69.2	1.06				
Filters	2295	3.14				
Total			4053	5.54	59	9.4
<b>Train C:</b>						
Filter supply line	390.6	2.69				
Filters	7249	3.33				
Total			13,098	7.94	191	13.5
Total in collection trains	10,330	11.38				
Total released from fuel	17,710	15.01	17,710	15.01	258	25.5

<sup>a</sup>Assuming proportional distribution of furnace fraction among collection train components.

<sup>b</sup>Based on initial fuel loading of 72.3 g U at 5.76% <sup>235</sup>U and calculated postirradiation fuel contents of 68.5 g U and 0.590 g Pu.

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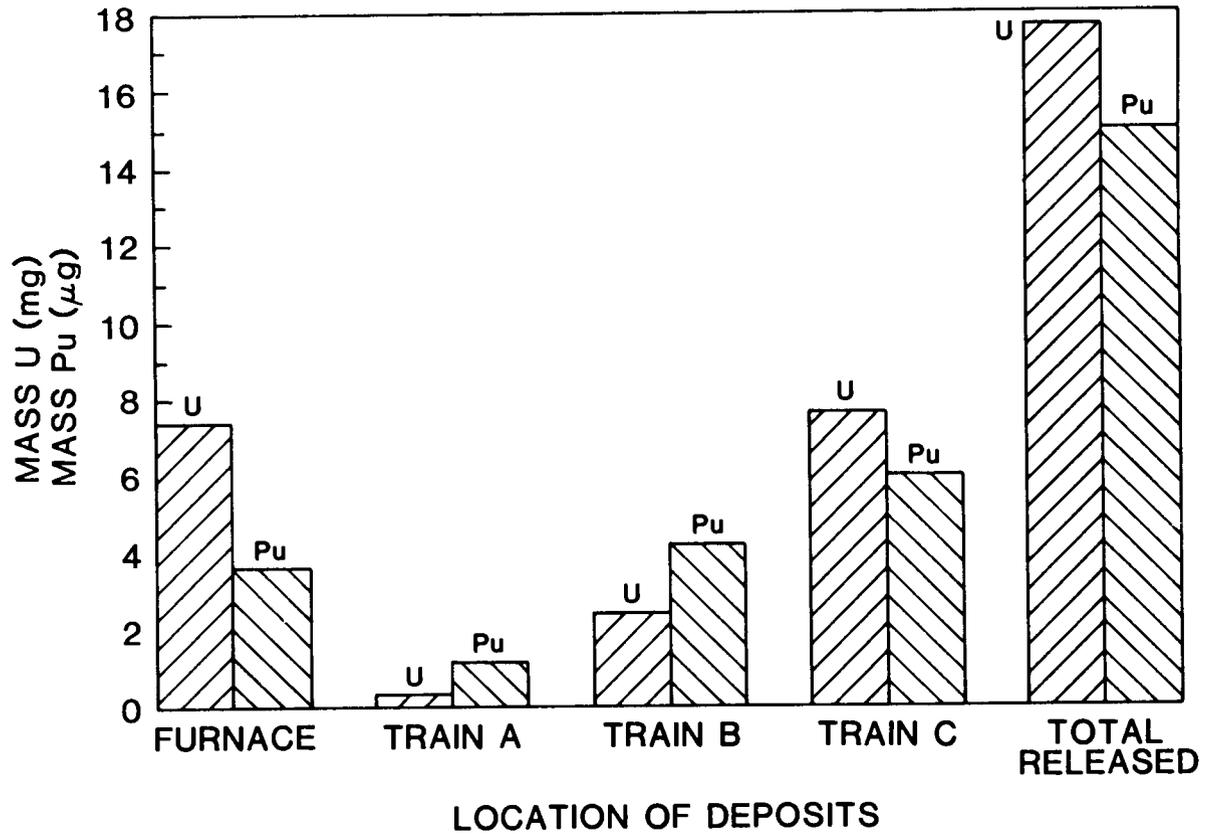


Fig. 20. Distributions of uranium and plutonium in test VI-2 apparatus. Note that uranium is mg and plutonium is  $\mu$ g.

Table 14. Vapor and aerosol deposits in test VI-2

	Weight of deposits (g) <sup>a</sup>			
	Train A	Train B	Train C	Total
Thermal gradient tube	0.057	0.139	0.085	0.281
Filters:				
Prefilter 1 <sup>b</sup>	0.087	0.140	0.112	0.339
Prefilter 2	0.069	0.255	0.085	0.409
HEPAs	0.032	0.039	0.034	0.105
Total filters	0.188	0.434	0.231	0.853
Total: TGT and filters	0.245	0.573	0.316	1.134
Total from experiment			1.134	
Aerosol concentration <sup>c</sup> (g/m <sup>3</sup> )	12.6	11.4	2.6	6.0

<sup>a</sup>Precision =  $\pm 0.003$  g.

<sup>b</sup>Includes estimated mass of deposits in connecting tubes, based on <sup>137</sup>Cs data.

<sup>c</sup>Assumes all aerosol was formed at temperatures  $> 1950$  K, and that the average temperature in the TGTs and filters was 423 K.

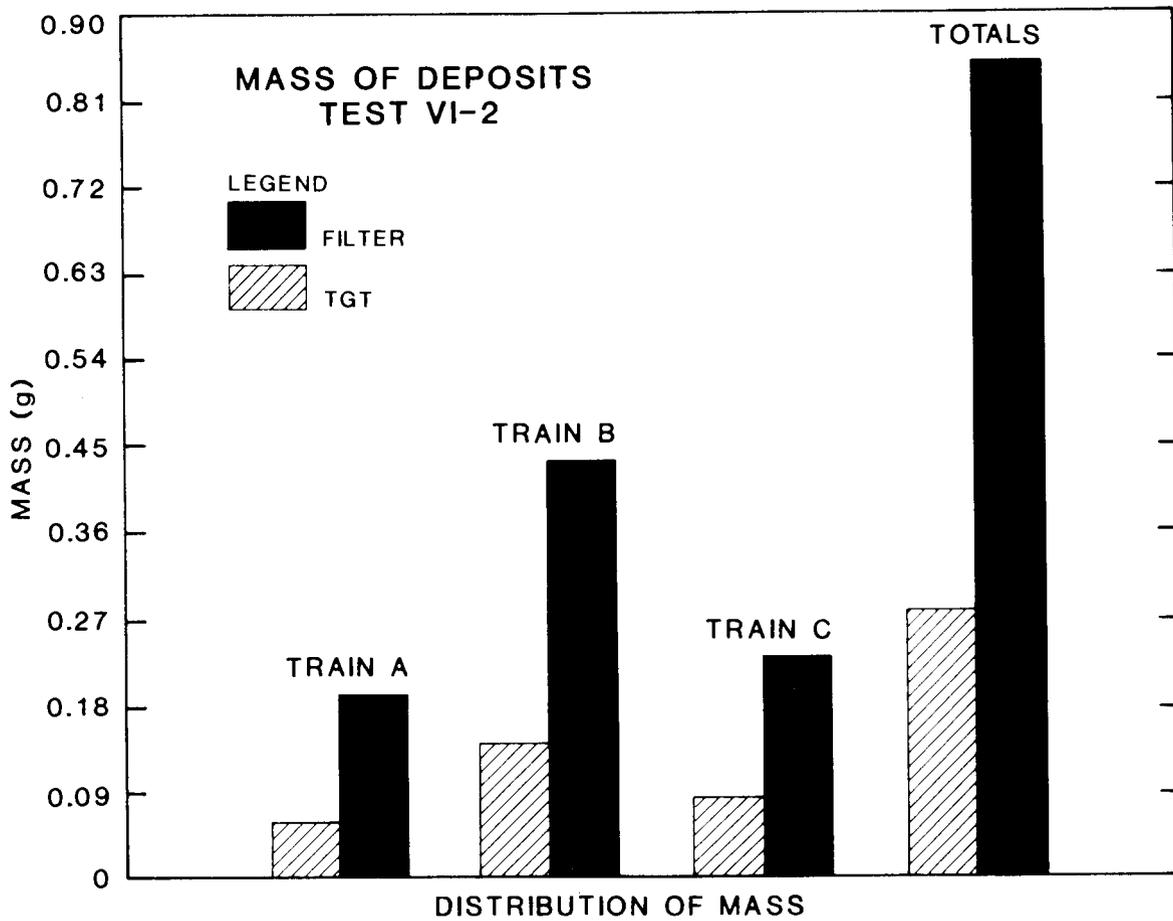


Fig. 21. Distribution of mass deposits in test VI-2 collection trains.

was highest during test phase A, and that this average mass concentration declined during the test as the supply of the more volatile material was depleted.

#### 4.2.7 Fuel Examination

As noted previously, the fuel specimen was cast in epoxy resin to preserve the test geometry, then cut with an abrasive saw for examination of the fuel cross sections. The general appearance of these sections (after only moderate polishing) is shown in Fig. 22. As may be seen, evidence of clad melting is apparent at only one location, at 2 cm from the top end. Otherwise, the cladding appears heavily oxidized with occasional fractures. Two of the above sections were selected for metallographic examination. These two sections (Nos. 4 and 6 from Fig. 22) were sent to Argonne National Laboratory for detailed examination of the microstructures and fission product distributions. The cladding in both sections appeared to be completely oxidized, as would be expected under the severe conditions of this test.

In considering the possibility of  $UO_2$  oxidation during the test, the diameters of the fuel in the sections shown in Fig. 22 were estimated. Since no standard was included in the photos, the inside diameter of the  $ZrO_2$  furnace tube, which was about 2.62 cm (1.03 in.) and should have been stable under the test conditions, was used for comparison. In many cases, the  $UO_2$  was badly broken, making the estimated diameters even more uncertain. In spite of these uncertainties in the data, which are summarized in Table 15, some increase in fuel pellet diameters was indicated in all cases, with the apparent increases greater where the fuel exposure to steam had been greater. Where the cladding was relatively intact, pellet measurements were more precise and the apparent diametral increases were 10 to 15%. In sections where the fuel cladding had suffered more fracturing, however, the indicated expansions were greater, ~15 to 20%, but were based on less precise measurements.

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

The fission product release data from this test (VI-2) have been compared with the results from earlier related experiments and also with a comprehensive NRC review of all relevant fission product release data.<sup>22</sup> This was the second test in a new vertically oriented furnace, and we were particularly interested in comparing the results from this test and from similar tests in a horizontal furnace. One of the tests in the horizontal furnace (HI-3) was conducted with a temperature/time regime similar to the combined phases A and B in test VI-2. The release data from test HI-3 and from test VI-1 (which was conducted in two 20-min phases at 2020 and 2300 K) are compared with the VI-2 data in Table 16. (For the VI tests, both the data for that part of the test most similar in time and temperature and also the data for the entire test are shown.) As these data indicate, the fractional releases of krypton and cesium varied considerably. First, as was seen in all six tests in the HI series, there was good agreement between krypton and cesium release in

## TEST VI-2 FUEL

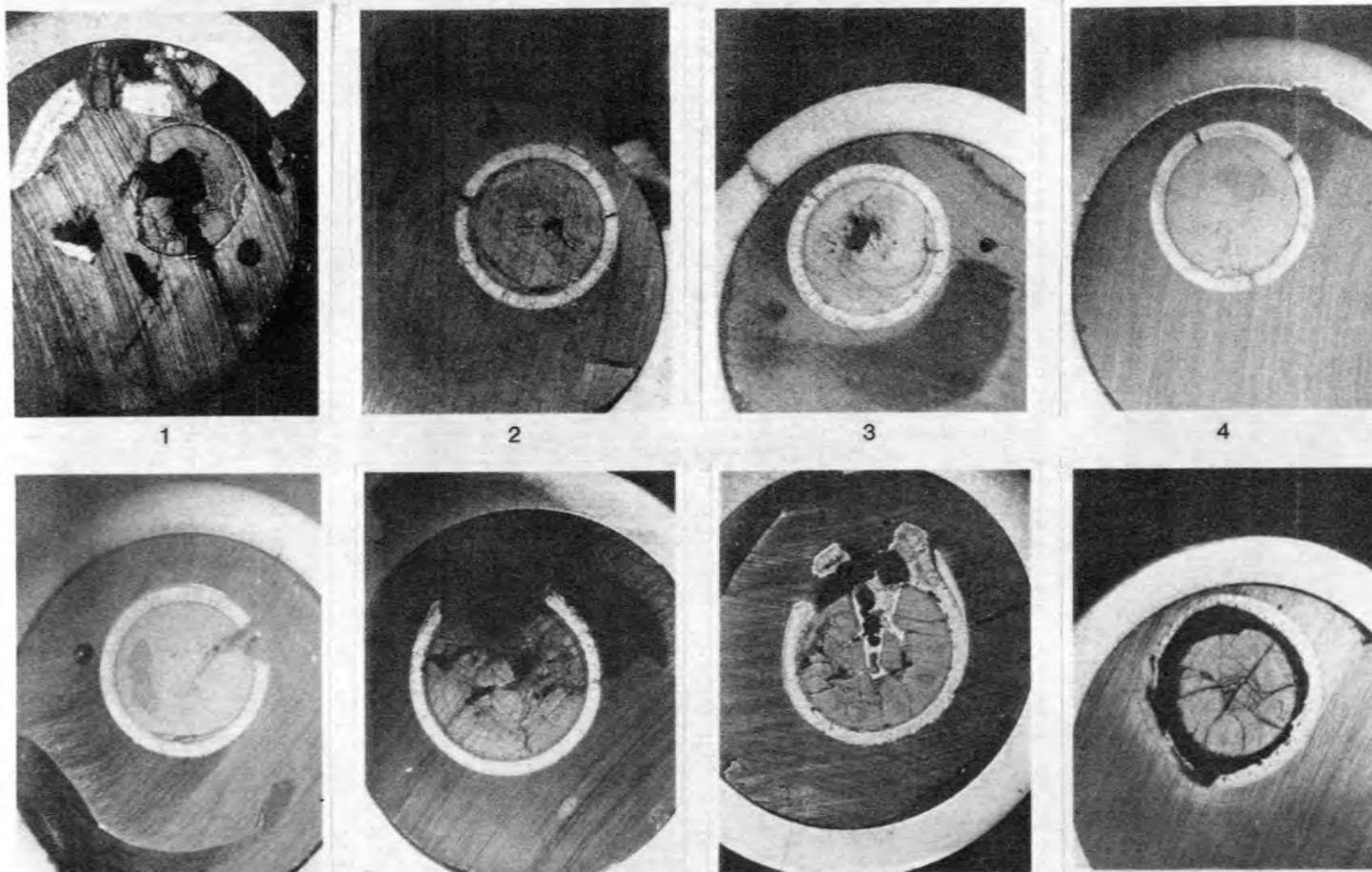


Fig. 22. Successive radials sections from fuel specimen in test VI-2; No. 1 is near bottom end (inlet) and No. 8 is near top end (outlet).

Table 15. Dimensions of fuel sections in tests VI-1 and VI-2

Test, section No.	Initial pellet OD (mm)	Apparent increase in OD (%)
VI-1, 1	9.34	14.7
VI-1, 2	9.34	12.7
VI-1, 3	9.34	~10.3 <sup>a</sup>
VI-1, 4	9.34	~17.0 <sup>a</sup>
VI-1, 6	9.34	~19.0 <sup>a</sup>
Average of 5	9.34	14.7
VI-2, 2	8.04	10.7
VI-2, 3	8.04	11-15 <sup>b</sup>
VI-2, 4	8.04	15-20 <sup>b</sup>
VI-2, 5	8.04	23.0
VI-2, 7	8.04	20-40 <sup>a,b</sup>

<sup>a</sup>Pellet diameter uncertain because of fracturing.

<sup>b</sup>Pellet eccentric where two values given.

Table 16. Comparison of release data from tests at similar conditions

Test phases	Test No.				
	HI-3	VI-1		VI-2	
		B+C <sup>a</sup>	A+B+C	A+B	A+B+C
Temperature (K)	2275	2300	2300	2300	2300
Effective time (min)	21.3	33	56	25	72
Steam flow rate <sup>b</sup>	Low	High	High	High	High
Release (%)					
<sup>85</sup> Kr	59.3	<sup>c</sup>	54.4 <sup>d,e</sup>	30.7 <sup>e</sup>	30.7 <sup>e</sup>
<sup>137</sup> Cs	58.8	35.2	74.3 <sup>d</sup>	54.1	63.4

<sup>a</sup>Test phases: A = heatup + 21.5 min at 2020 K, B = heatup + 5 min at 2300 K, C = 15 min at 2300 K + cooldown.

<sup>b</sup>At high steam flow rate (~1 L/min) cladding was completely oxidized; at low steam flow rate (~0.3 L/min) cladding was only partly oxidized.

<sup>c</sup>No intermediate time values for <sup>85</sup>Kr available.

<sup>d</sup>Total release values; assumes that, in absence of test phase A at lower temperature, the release during phase A would have occurred at 2300 K, phase C.

<sup>e</sup><sup>85</sup>Kr values in tests VI-1 and VI-2 are low because of loss during posttest transfer.

test HI-3. In tests VI-1 and VI-2, however, the indicated releases of cesium were higher than those of krypton. This difference was traced to a holdup of krypton within the Ascarite traps during posttest transfer of the krypton from the first to the second cold charcoal trap (see Sect. 4.2.1.). The delay in sweeping the krypton through the Ascarite traps was not discovered until after test VI-2, and a large fraction of the released krypton was lost. In addition, the data indicate that ~25% of the krypton in test VI-1 was lost, probably in the same manner. These uncertainties prevent more meaningful comparison of the krypton results.

Considering the differences in fuel history (H. B. Robinson in HI-3, Ocone in VI-1, and BR3 in VI-2) and in steam availability (low vs high), the agreement in cesium release values from tests HI-3 and VI-2 (A+B) (58.8 vs 54.1%) is quite good. This agreement is an indication that little if any fuel oxidation had occurred by the end of phase B in test VI-2. We conclude that the previously discussed apparent fuel expansion, as indicated by dimensional measurement of cross section photos, was caused by steam oxidation of the fuel, after fracturing of the oxidized cladding, during the 40-min period of test phase C. It is not surprising that phases A+B in test VI-2 (25 min at 2300 K) showed a much higher total cesium release (54% vs 35%) than did phases B+C in test VI-1 (33 min at 2300 K). In test phase VI-1A, the fuel had already been heated at 2020 K for 20 min, thereby releasing a big fraction (39%) of the total cesium, which we conclude was located in more readily releasable sites. Consequently, much less cesium remained in the fuel, and the release rate for this remaining cesium was much lower in test VI-1 (B+C) than in test VI-2 (A+B). The higher value for total cesium release in test VI-1 (74.3%) compared with test VI-2 (63.4%) may have been related to differences in fuel morphology resulting from different irradiation histories of the fuel specimens. The data show that in both tests the cesium tended to be released early in the high-temperature period.

The release rate of cesium in the two vertical tests (VI-1 and VI-2) is compared with similar data from the six HI tests, and with the NRC review curve, in Fig. 23. Because the VI tests were conducted in three phases, compared to only one phase in the HI tests, each VI test provided three release rate values. In test VI-1, these three data points were plotted for three different effective temperatures (see Fig. 23) and are comparable to but indicate a somewhat weaker temperature dependence (shallower slope) than indicated by the data from the HI tests. Like most of the HI test data, the cesium release rate in test VI-1 falls significantly (by factors of 3 to 10) below the NUREG-0772 curve.<sup>22</sup> For test VI-2, which experienced only one extended test temperature, the early release rate values were comparable to the HI test data, and then show a steady decline, by more than a factor of 10, during the test. This behavior is illustrated more clearly in Fig. 24, in which smoothed on-line release rate data are plotted vs test time. The peak release rate occurred at the beginning of the 2300 K period and declined approximately exponentially thereafter. This result suggests that, for this fuel specimen, the transient temperature (heatup) period was more effective in releasing the contained fission products than the constant temperature period.

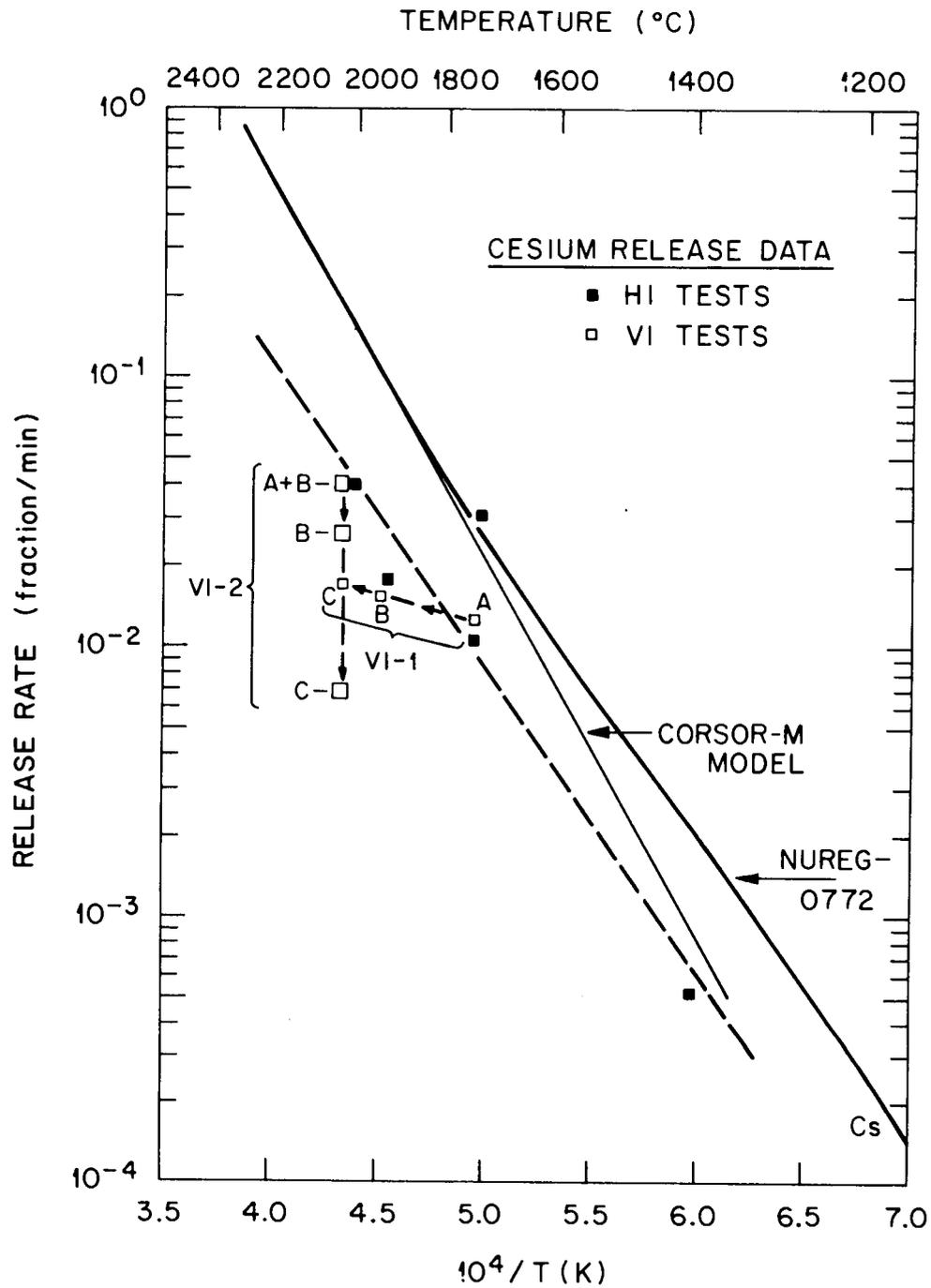


Fig. 23. Comparison of release rate coefficients from tests VI-2 and VI-1 with earlier work.

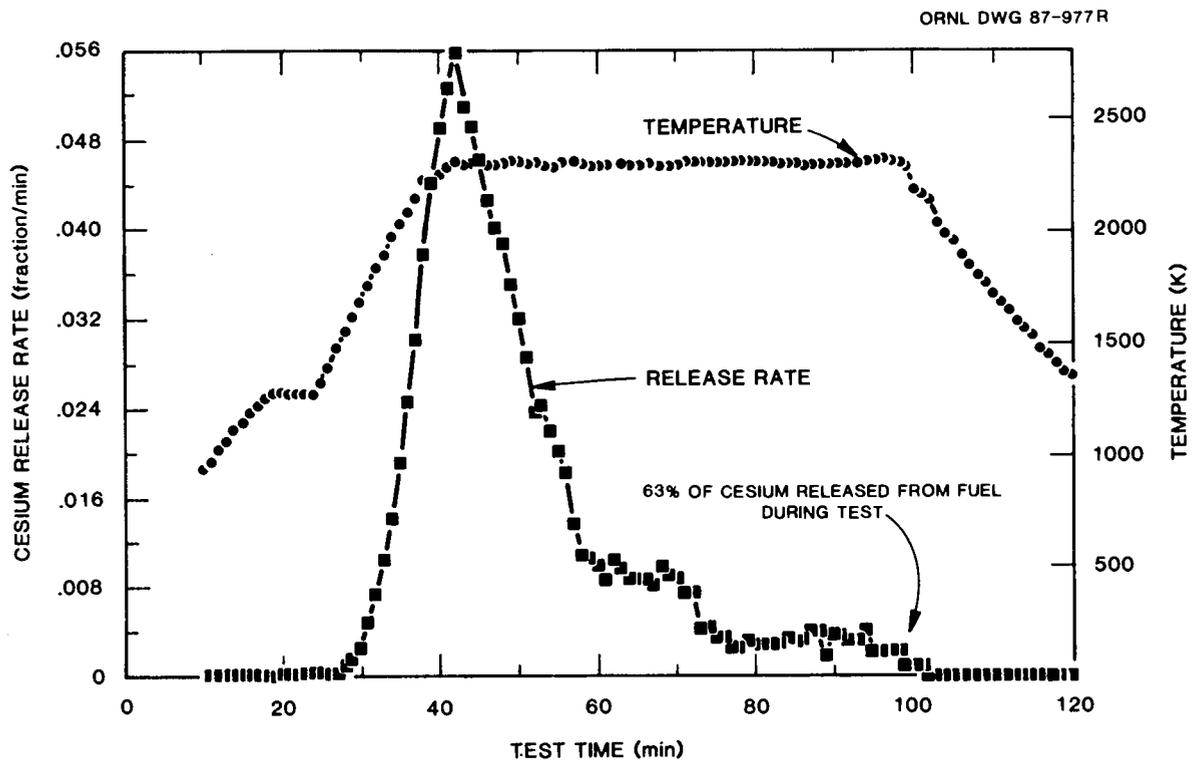


Fig. 24. On-line measurements of cesium release rates during test VI-2.

## 6. CONCLUSIONS

In view of the preliminary nature of this report and the fact that some important analyses/results have not yet been obtained, a thorough interpretation of the results of test VI-2 cannot be done at this time. Upon completion of the currently delayed work, such interpretation will be published. However, several significant observations are appropriate at this time.

1. The vertical furnace operated well and the planned test conditions were accomplished. All components of the expanded fission product collection system provided useful data. The apparatus for the continuous measurement of hydrogen operated very well, proving its usefulness as an aid in the interpretation of test results. Consequently, the test was considered successful.
2. The expansion and perfection of gamma scanning techniques to determine the fission product content of irradiated  $\text{UO}_2$  fuel improved the precision of data on the inventories in the test specimen, both before and after the test, and thereby improved our ability to measure fission product release.
3. The total release values for the more volatile fission products — 31% of Kr, 8.9% of Ag, 68% of Sb, and 63% of Cs — were determined for the specific conditions of this test. Comparison of these data with the results from similar previous tests shows significant differences, but experimental differences (temperature, time, and atmosphere) appear to explain some of the discrepancies. Perhaps of equal or greater importance are differences in the specific fuel materials. As noted in previous tests, irradiation history, especially fuel temperature and burnup, appear to influence significantly the fuel-fission product morphology and thus the fission product migration and release processes.
4. The data obtained by SSMS indicated significant release of several less volatile fission products, most notably molybdenum and barium, as well as small amounts of uranium, and in addition, release of several impurities from the structural ceramics. These data, however, were difficult to interpret. High release values for molybdenum would be expected in the strongly oxidizing atmosphere of this test. The 17.7 mg release of uranium indicated by chemical analysis — 0.026% of the fuel mass — occurred at a relatively constant rate. On the other hand, the total mass of material collected on the TGTs and filters showed that the total release peaked early in the test then declined as the more volatile species were depleted. The indicated lower release rate for plutonium compared to uranium is significant to safety considerations.
5. The oxidation behavior of the Zircaloy cladding, as indicated by continuous measurement of hydrogen generation, was in good agreement with a previously developed model, indicating an adequate understanding of this reaction which significantly influences the behavior of some fission products.

6. Posttest examination of the fuel specimen indicated that the cladding was completely oxidized and that minimal fuel-cladding interactions had occurred, verifying that the planned test conditions had been achieved.

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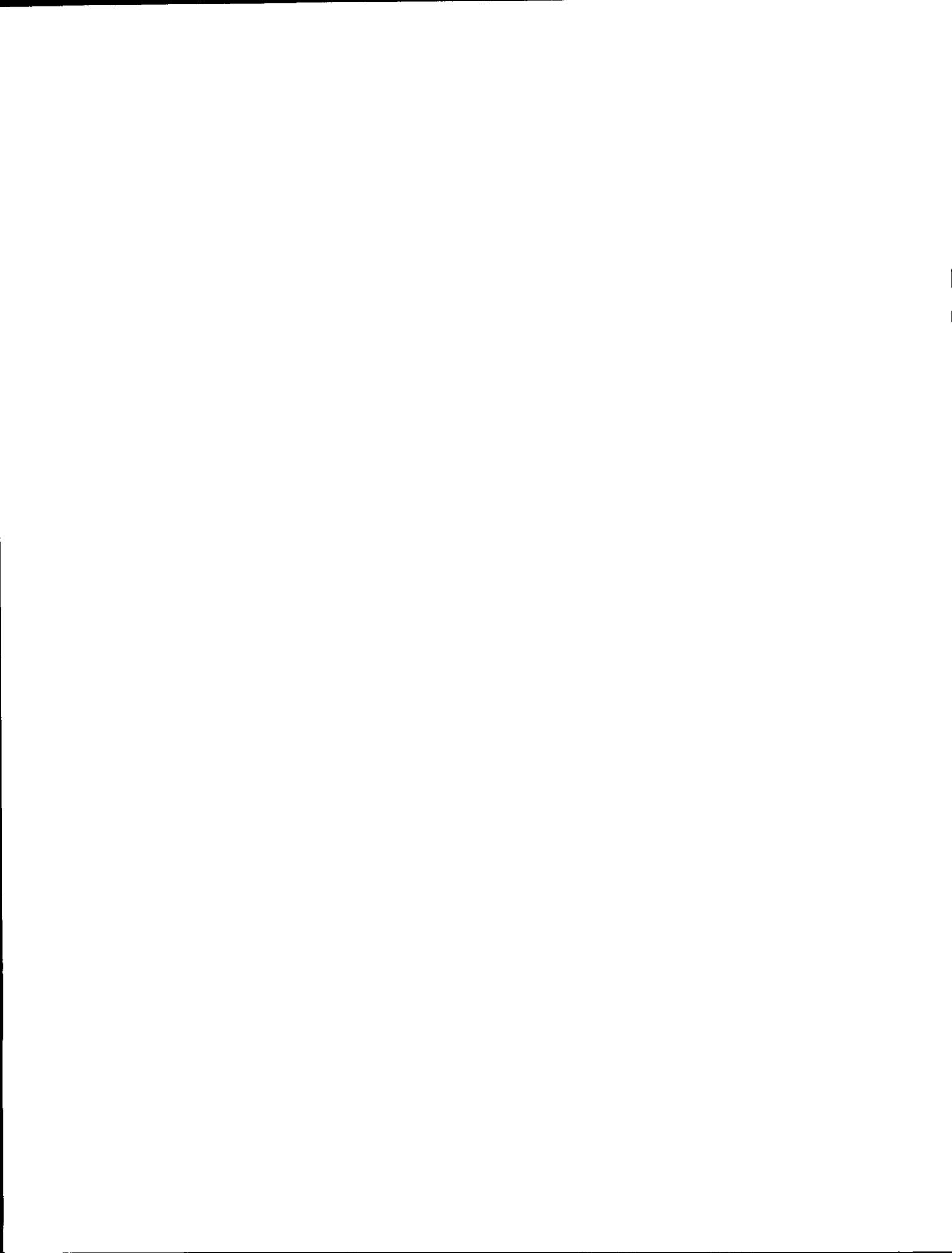


APPENDIX: PLANNED VERTICAL FISSION PRODUCT RELEASE TESTS

Test No.	Temperature (K)	Time (min)	Oxidation rate <sup>a</sup>	Fuel specimen			Objective, comparison, and justification of test
				Reactor	Burnup (MWd/kg)	Gas release (%)	
VI-1A	2020	H <sup>b</sup> + 20	High	OCONEE (PWR)	40	4.1	(a) Compare behavior in vertical vs horizontal furnace (HI-5). (b) Data from multiple collection train.
VI-1B	Heatup	H					
VI-1C	2300	20 + C <sup>b</sup>					
VI-2A	2300	H + 2	High	BR3 (Belgium)	40	~2	(a) Study release rate at constant temperature. (b) Compare with HI-3, HI-6, and VI-1.
VI-2B	2300	18					
VI-2C	2300	40 + C					
----- Above tests completed -----							
VI-3A	2000	H + 20	High	BR3 (Belgium)	42	~2	(a) Study CsI decomposition on SST TGT. (b) Fission product release at 2700 K. (c) Melt(?) behavior with high oxidation.
VI-3B	Heatup	H					
VI-3C	2700	20 + C					
VI-4A	2400	H	0	BR3 (Belgium)	47	~2	Duplicate ACRR test conditions for comparison with in-pile tests.
VI-4B	2400	20					
VI-4C	2400	C					
VI-5A	2000	H + 20	0	BR3 (Belgium)	42	~2	(a) Release rate at 2700 K in reducing atmosphere. (b) Meltdown behavior with low oxidation. (c) Compare with test VI-3 (oxidizing).
VI-5B	Heatup	H					
VI-5C	2700	20 + C					
VI-6A	Heatup	H	0	BR3 (Belgium)	42	~2	(a) Compare with VI-2A except in hydrogen. (b) Compare with VI-2. (c) Assure oxidation of UO <sub>2</sub> , measure ruthenium release.
VI-6B	2300	30	High				
VI-6C	2300	30 + C	High				

<sup>a</sup>Oxidation rate controlled by steam flow, all units L/min: high = 0.8 steam + 0.2 He (80% steam); low = 0.2 steam + 3.0 H<sub>2</sub> (20% steam and 30% hydrogen); and 0 = 0.5 H<sub>2</sub> (no steam).

<sup>b</sup>H = heatup; C = cooldown; heatup rate = 1 K/s.



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12 SUPPLEMENTARY NOTES					
13 ABSTRACT (200 words or less)					
<p>The second in a series of high-temperature fission product release tests in a new vertical test apparatus was conducted. The 15.2-cm-long test specimen had been irradiated to a burnup of ~42 MWd/kg. Using an induction furnace and collection apparatus mounted in a hot cell, it was heated under simulated LWR accident conditions for 60 min at 2300 K in flowing steam.</p> <p>Posttest inspection showed severe oxidation of the cladding but only minimal fragmentation of the Zircaloy cladding. Based on fission product inventories in the fuel, total releases of 30.7% for <sup>85</sup>Kr, 8.93% for <sup>110m</sup>Ag, 68.2% for <sup>125</sup>Sb, and 63.4% for <sup>137</sup>Cs were measured. Smaller release fractions for many other fission products and fuel material — uranium and plutonium — were determined also. Total mass release from the furnace to the collection system was 1.08 g. Comparison of the results from this test showed reasonable agreement with the results from two earlier tests at similar conditions. The release rate coefficients from the six tests in the HI series and tests VI-1 and VI-2 fall significantly below a widely accepted standard used in LWR safety analyses.</p>					
14 DOCUMENT ANALYSIS -- KEYWORDS, DESCRIPTORS				15 AVAILABILITY STATEMENT	
Fission product		Fuel damage		Unlimited	
Fission product release		Induction furnace			
16 IDENTIFIERS OPEN ENDED TERMS				16 SECURITY CLASSIFICATION	
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