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Data Summary Report for Fission Product Release Test VI-4

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Prepared by
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Oak Ridge National Laboratory

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Data Summary Report for Fission Product Release Test VI-4

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Prepared by
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ABSTRACT

This was the fourth in a series of high-temperature fission product release tests in a vertical test apparatus. 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 47 MWd/kg. In simulation of a severe accident in a light-water reactor, it was heated in hydrogen in a hot cell-mounted test apparatus to a maximum test temperature of 2400 K for a period of 20 min. The released fission products were collected on components designed to facilitate sampling and analysis.

On-line radioactivity measurements and posttest inspection revealed that the fuel had partially collapsed at about the time the cladding melted. Based on fission product inventories measured in the fuel or calculated by ORIGEN2, analyses of test components showed total releases from the fuel of 85% for ^{85}Kr, <1% for ^{106}Ru, 3.9% for ^{125}Sb, 96% for both ^{134}Cs and ^{137}Cs, and 13% for ^{154}Eu.

Large fractions of the released fission products (up to 96% of the ^{154}Eu) were retained in the furnace. Small release fractions for several other fission products – Rb, Br, Sr, Te, I, and Ba – were detected also. In addition, very small amounts of fuel material – uranium and plutonium – were released. Total mass release from the furnace to the collection system, which included fission products, fuel material, and structural materials, was 0.40 g, with 40% of this material being deposited as vapor and 60% of it being collected as aerosols. The results from this test were compared with previous tests in this series and with an in-pile test at similar conditions at Sandia National Laboratories. There was no indication that the mode of heating (fission heat vs radiant heat) significantly affected fission product release.

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FOREWORD

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

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 1984.

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.

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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, December 1986.

14. Toshiyuki Yamashita, *Steam Oxidation of Zircaloy Cladding in the ORNL Fission Product Release Tests*, NUREG/CR-4777 (ORNL/TM-10272), March 1988.

15. S. K. Wisbey, *Preliminary Studies of the Morphology of Thermal Gradient Tube Deposits for Fission Product Release Experiments*, NUREG/CR-4778 (ORNL/TM-10273), March 1988.

16. C. S. Webster and M. F. Osborne, *The Use of Fiber Optics for Remote Temperature Measurement in Fission Product Release Tests*, NUREG/CR-4721 (ORNL/TM-10366), April 1989.

17. 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), June 1989.

18. M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, and C. S. Webster, *Data Summary Report for Fission Product Release Test VI-2*, NUREG/CR-5340 (ORNL/TM-11105), September 1989.

19. M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, C. S. Webster, H. K. Lee, T. Nakamura, and Y.-C. Tong, *Data Summary Report for Fission Product Release Test VI-3*, NUREG/CR-5480 (ORNL/TM-11399), April 1990.

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DATA SUMMARY REPORT FOR FISSION PRODUCT RELEASE TEST VI-4

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

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-4). Although all final data are not currently available, this report presents most of the results for potential use by other reactor safety researchers. Complete interpretation and correlation of these results with related experiments and with theoretical behavior will be included in a subsequent report. Similar data summary reports for previous tests in this project, and also other reports of related project activities, are listed in the Foreword.

The fuel specimen used in this test was cut from a fuel rod that had been 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. A summary of these data and the fission product inventories, as measured in the fuel by gamma ray spectrometry and calculated by ORIGEN2, are included in Sect. 3. The test results and some preliminary interpretations are presented in Sect. 4; these results are compared with data from previous tests in Sect. 5. The most important results are:

1. This was the first test in this series to be conducted in hydrogen. The experimental apparatus performed quite well, and the desired axial temperature gradient – 2400 K over the lower half of the specimen with the temperature declining to 2000 K at the top end – was achieved. This unusual temperature profile was desired to duplicate the conditions of an in-pile test (ST-1) at Sandia National Laboratories (SNL). Although the fuel specimen collapsed after cladding meltdown, with the result that most of the fuel was at about 2400 K for most of the high-temperature period, the test was quite successful.

2. Posttest examination confirmed that the 15-cm-long fuel specimen had partially collapsed. (Radioactivity measurements during the test had indicated a downward shift of the fuel shortly after it had reached the melting temperature of the Zircaloy cladding.) All of the cladding and about half of the fuel was found in a rubble bed in the lower part of the furnace.

3. The values for total release from the fuel specimen, based on gamma ray spectrometry measurements, were ~85% for ^{85}Kr, <1% for ^{106}Ru, 3.9% for ^{125}Sb, 96% for both ^{134}Cs and ^{137}Cs, and 13.4% for ^{154}Eu. Most of the released ^{125}Sb and ^{154}Eu and 48% of the released ^{137}Cs were deposited on the ZrO_2 ceramics at the outlet (upper) end

of the furnace. Pretest and posttest gamma ray spectrometry of the fuel was valuable in determining the release fractions as well as the axial distributions of the fission products along the fuel rod. Small amounts of other fission products – Br, Rb, Sr, Te, I, and Ba – were detected in the collection system by other analytical techniques.

4. The fission product distributions in the thermal gradient tubes (TGTs) showed the highest concentration of cesium near midlength of TGT A, at a deposition temperature of ~800 K (~525°C). The most probable composition of this material was concluded to be CsI mixed with other cesium compounds. A smaller peak of cesium was found at a similar location in TGT B.

5. The total masses of deposits on the TGTs and filters were determined by direct weighing to be 0.401 g, with 40% of this (0.161 g) being deposited as vapor on the TGTs and 60% (0.240 g) being deposited on the filters as aerosols. The mass release rate was highest during test Phase A, which included the heatup period to 2400 K only. The maximum aerosol concentration, averaged over the Phase A collection period, was 30 g/m^3; for the entire test, however, the average aerosol concentration was only 11 g/m^3.

6. The fission product release results indicated that conditions were more reducing during Phase A than later in the test. We believe that the presence of molten zirconium during this period provided a lower oxygen potential at the surface of the fuel than was present during Phases B and C. This lower oxygen potential would explain the enhanced release of Sr, Ba, and U during Phase A and also the higher fraction of Cs released as vapor than was observed later in the test.

7. Comparison of the release data from this test in hydrogen with the results of previous tests in steam showed quite different release behavior for several fission product elements: much less Ru and Sb and much more Eu were released than would have been expected at the same temperature in steam. In addition, more strontium and barium were released in this test at 2400 K in hydrogen than at 2700 K in steam in test VI-3.

8. Comparison of the fission product release results from this test with those from Test ST-1 at Sandia National Laboratories showed no influence of the experimental method (fission heating in a reactor at SNL vs radiant heating in a hot cell at ORNL) on the results. Hence, there is no indication that these and previous results from ORNL hot cell tests are not applicable for use in LWR accident analyses.

2. INTRODUCTION

This report summarizes data from the fourth fission product release test in a high-temperature vertical 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 series of tests, conducted under similar conditions at temperatures of 773 to 1873 K (500 to 1600°C), were reported in detail by Lorenz et al.;[1-4] additional tests at higher temperatures (~1700 to 2300 K) were reported by Osborne et al.[5-13] The purpose of this recent 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.[14] The specific objectives of this program are:

1. to obtain fission product release and behavior data applicable to the analysis of severe accidents in LWRs, and

2. to apply these data to the development of VICTORIA and other fission product release and transport models.

Tests of high-burnup fuel from commercial reactors are emphasized in this program. The applicability of simulated fuel (unirradiated UO_2 containing radioactive fission product tracers) has been considered, and several simulant tests have been conducted to provide valuable data about the behavior of specific fission product species.[15,16] All previous tests in the current series (prior to this test in helium-hydrogen) were conducted at atmospheric pressure in a flowing mixture of steam and helium in a hot cell. Steam concentrations have been varied to obtain data applicable to different accident sequences or fuel locations within the core.

The experimental procedures and techniques used in preparing and conducting this test, as well as in posttest examination and analysis, were very similar to those used for the six tests in the HI series[5-10] and the three previous tests in the VI series.[11-13] The analytical techniques employed are listed in Table 1. This report provides a description of test VI-4 and detailed tabulations 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 together with comparable results from other tests in a later report. Thorough data evaluation and correlation of all results from the VI test series will be included in subsequent reviews and reports covering this series of fission product release tests at temperatures up to 2700 K.

3. TEST DESCRIPTION

This remotely-operated vertical test apparatus[17] is mounted in a hot cell and is capable of heating fuels at temperatures up to 2700 K for time periods up to ~60 min in reactive atmospheres, such as steam and/or hydrogen mixed with helium. Details of the furnace are shown in

Table 1. Analytical techniques for fission product analysis

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

[a]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.

Fig. 1 and the entire test apparatus is illustrated in Fig. 2. Good measurement and control of temperature and gas flow rates have been demonstrated in previous work.[11-13] Both manual and automatic optical pyrometers are used for temperature measurement; thermocouples located in the inlet end of the furnace supply data during the low-temperature heatup phase (up to 1200 K), where the pyrometers are inoperable.

The released fission products are collected in three sequentially operated, parallel collection trains. Each train is composed of (1) a platinum-lined (or in some cases, a stainless steel-lined) TGT, which is designed to collect vapors according to their condensation temperature; (2) a filter package containing graduated fiberglass filters for collecting aerosols and heated charcoal for iodine sorption; and (3) cold charcoal traps for rare gas collection. The on-line releases of ^{85}Kr and ^{137}Cs are monitored by detectors observing these components. Details of the fission product collection system are shown in Fig. 3. For this test in hydrogen, the components for steam collection and for the collection and measurement of hydrogen and CO were not used.

The tests planned for this vertical apparatus assumed that test temperatures (2000 to 2700 K) were the dominant variables. The flow rate of reactive gas (steam and/or hydrogen flowing at 0.3 to 1.5 L/min) and time (up to ~100 min), both of which affect the extent of reactions, were secondary variables. The objectives of this particular test were twofold: (1) to obtain release data from BR3 fuel heated in flowing hydrogen for a period of 20 min in a specified axial temperature gradient (the lower half of the specimen was to be at 2400 K with the upper half declining linearly to 2000 K at the top end); and (2) to compare the results with data from in-pile test ST-1 at Sandia National Laboratories, which experienced a similar temperature gradient.*

3.1 FUEL SPECIMEN DATA

The test specimen was cut from rod I-1059 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 fuel rod were reported by Adams and Dabell† and are listed in Table 2. The fuel in this rod had an initial enrichment of 8.26% ^{235}U and had attained a maximum burnup of ~47 MWd/kg at a maximum heat rating of 270 W/cm during irradiation.

The test specimen was a 15.2-cm-long section from the high-burnup region of rod I-1059. End caps of Zircaloy-2 were machined and welded

---

*K. O. Reil, "ACRR Source Term Experiments: ST-1 and ST-2 Results," presented at Severe Accident Research Program Partners' Review Meeting, April 25–30, 1988, Albuquerque, NM.

†J. P. Adams and B. R. Dabell, Idaho National Engineering Laboratory, Idaho Falls, ID 83415, "Characteristics of UO_2-Zr Fuel Rods Irradiated in the BR3 Reactor," draft preliminary report, November 1986.

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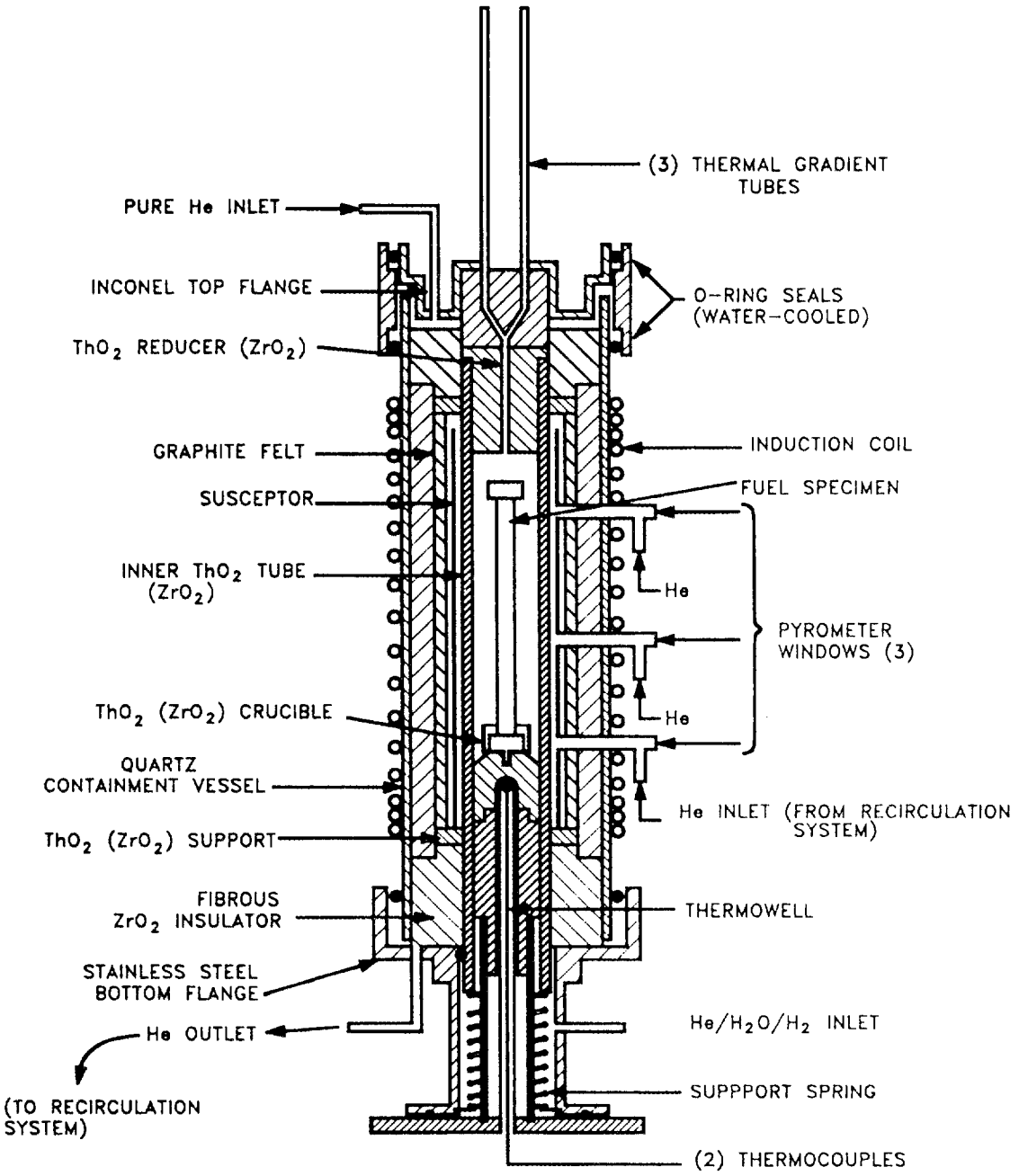

Fig. 1. Vertical fission product release furnace, as used in test VI-4.

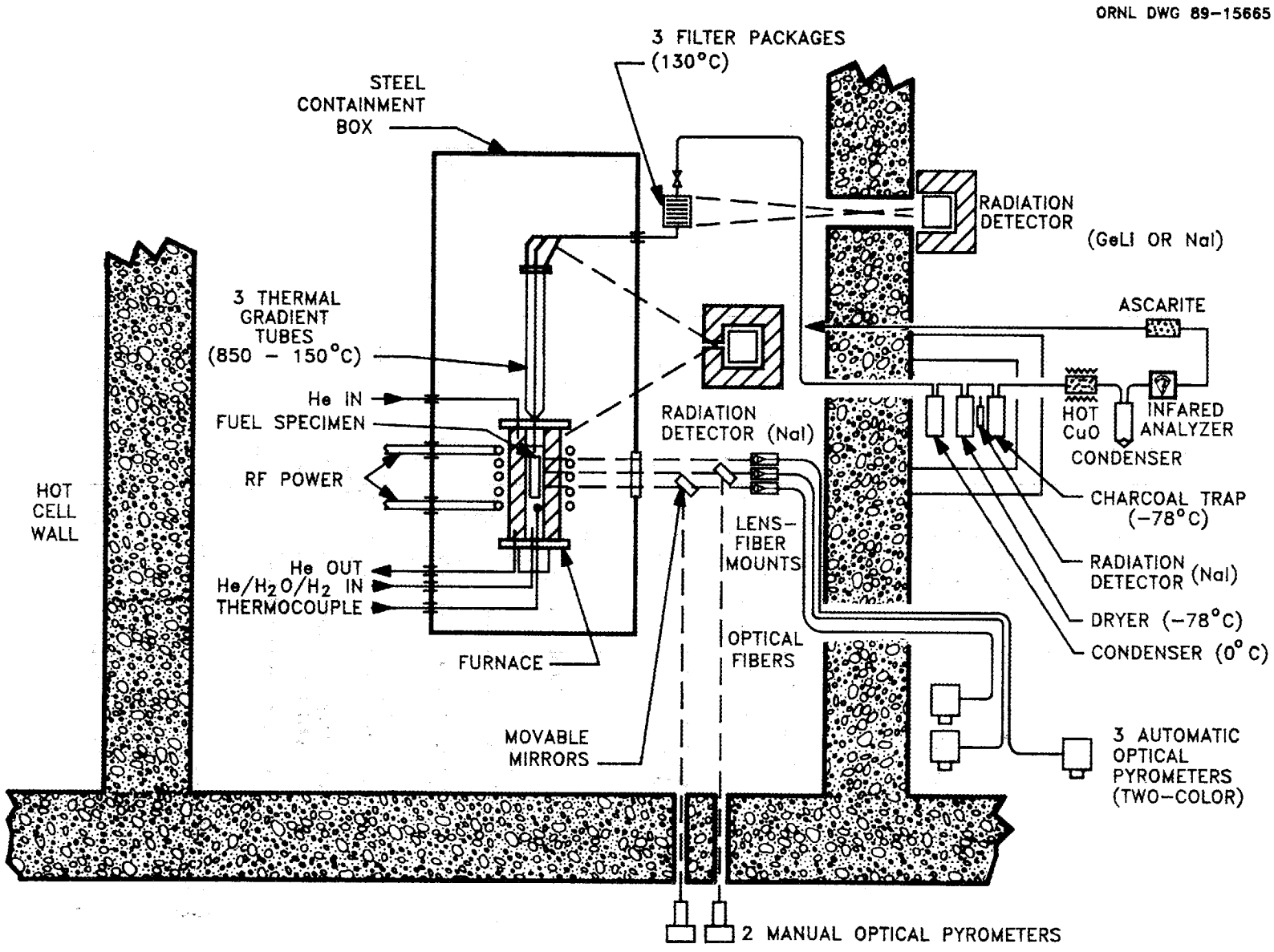

Fig. 2. Vertical fission product release appartus.

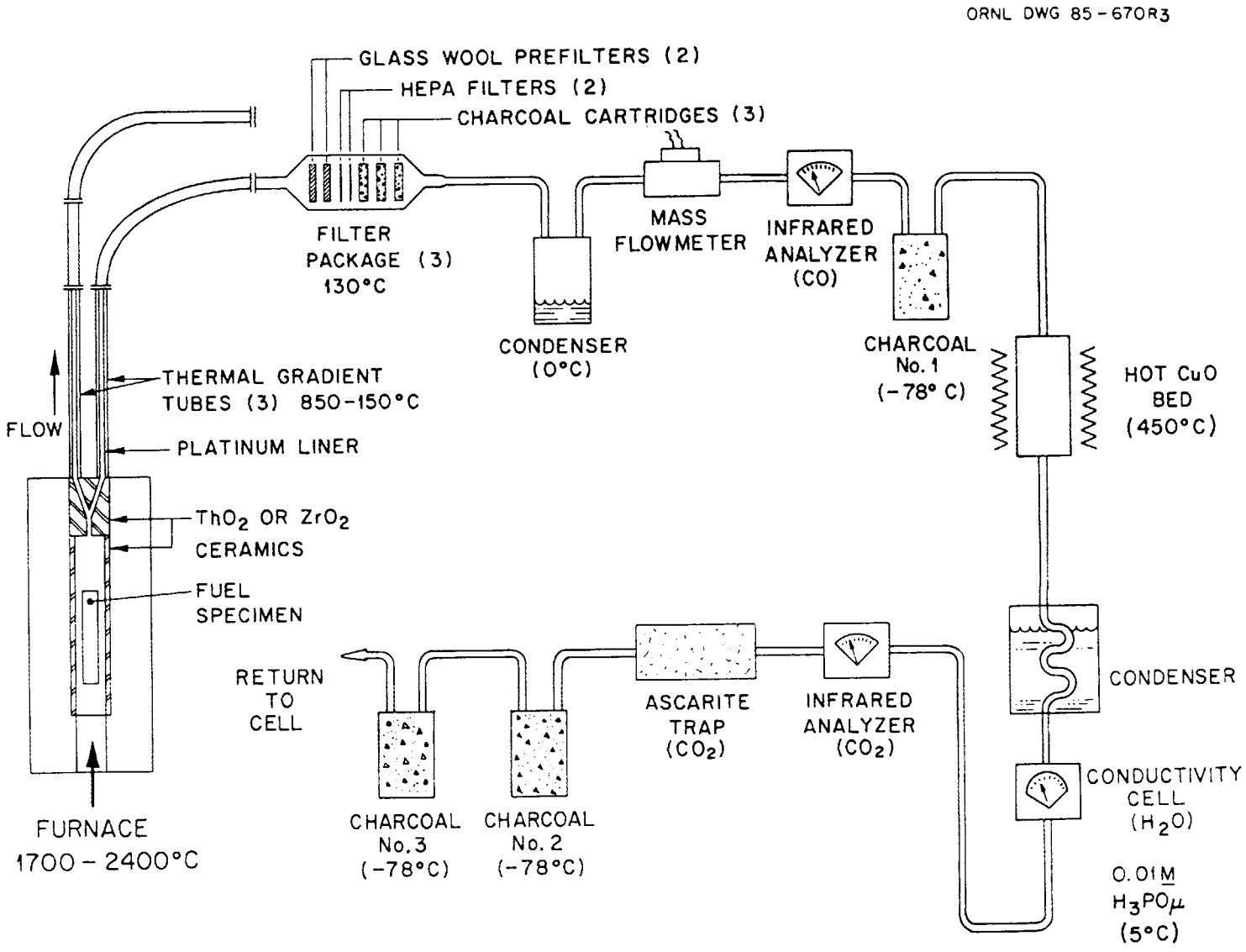

Fig. 3. Components of fission product collection system.

Table 2. Data for fuel specimen used in test VI-4

Fuel specimen identification	Rod I-1059, Section B BR3 Reactor (Belgium)
Irradiation data	
Period	July 15, 1976, to Sept. 26, 1980
Fuel specien burnup	~47 MWd/kg U
Maximum heat rating	270 W/cm
Specimen data	
Length	15.2 cm
Fuel loading	78.22 g UO_2 (68.94 g U)
Initial enrichment	8.26% ^{235}U
Total weight	99.29 g
Weight of Zircaloy	21.07 g
Gas release during irradiation	~10% (from specimen)

into the ends of the test specimen during its preparation at SNL, as shown in Fig. 4. The bottom end cap included a pin to facilitate vertical mounting in the furnace. In addition to the test VI-4 fuel specimen, an adjacent 15.2-cm-long specimen was cut from rod I-1059 and prepared for future testing. Also, a short section (0.5-cm-long) was cut as an archive sample for possible future characterization.

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-1059 contained no unusual distributions of fission products, and that the operating power (270 W/cm) was not high enough to cause significant migration of the volatile fission products, especially cesium. Although no analyses of the gas from this rod were obtained, examination of the data from comparable rods and our thin-slit gamma scan data for this specimen indicated that the release of ^{85}Kr from the fuel during irradiation in the region of the test specimen was ~10%. Fission product inventories for the specimen were measured by direct gamma spectrometry of the fuel and/or were calculated with the ORIGEN2 computer program.[18] These data are shown in Tables 3 and 4.

3.2 TEST CONDITIONS AND OPERATION

As in each of the previous HI and VI 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 were done remotely. No in-cell operations were required during the test. Following closure of the furnace and containment box,

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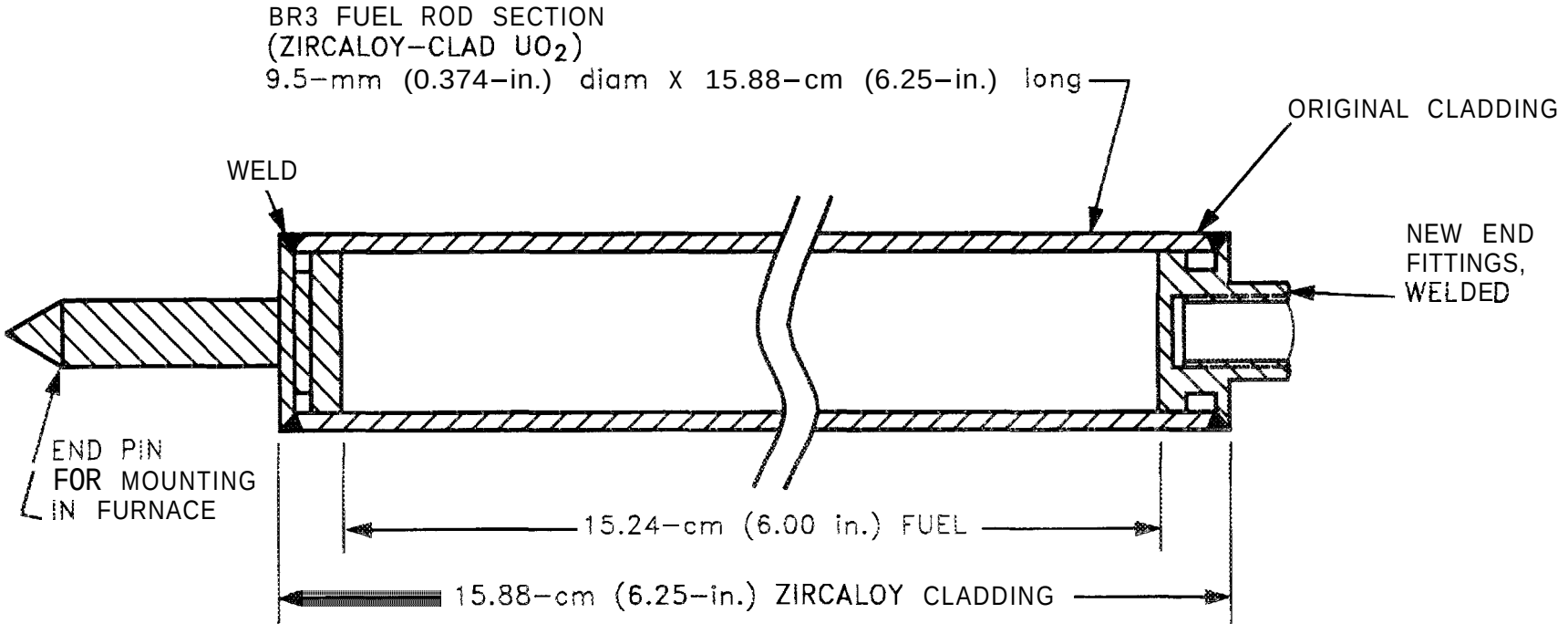

Fig. 4. Fabrication details and dimensions of fuel specimen heated in test VI-4.

Table 3. Inventories of principal elements in test VI-4 fuel

Elements	Amounts in fuel[a] (g/MTU)	Amounts in fuel[a] (Ci/MTU)	Amounts in VI-4 Specimen[a] (g)	Amounts in VI-4 Specimen[a] (Ci)
Fission products:				
Se	8.771E+01	6.023E-01	6.047E-03	4.152E-05
Br	3.381E+01	0.000E+00	2.331E-03	0.000E+00
Kr	6.084E+02	1.008E+04	4.194E-02	6.949E-01
Rb	6.035E+02	3.701E-05	4.161E-02	2.551E-09
Sr	1.402E+03	1.081E+04	9.665E-02	7.452E-01
Y	8.001E+02	1.081E+04	5.516E-02	7.452E-01
Zr	5.796E+03	2.924E+00	3.996E-01	2.016E-04
Mo	4.889E+03	0.000E+00	3.370E-01	0.000E+00
Tc	1.149E+03	1.949E+01	7.921E-02	1.344E-03
Ru	2.696E+03	4.325E+03	1.859E-01	2.982E-01
Rh	5.918E+02	4.325E+03	4.080E-02	2.982E-01
Pd	1.039E+03	7.124E-02	7.163E-02	4.911E-06
Ag	4.739E+01	3.510E+00	3.267E-03	2.420E-04
Cd	6.854E+01	2.989E+01	4.725E-03	2.061E-03
In	2.700E+00	0.000E+00	1.861E-04	0.000E+00
Sn	8.353E+01	1.243E+00	5.759E-03	8.569E-05
Sb	1.961E+01	2.390E+03	1.352E-03	1.648E-01
Te	5.895E+02	5.831E+02	4.064E-02	4.020E-02
I	2.711E+02	3.780E-02	1.869E-02	2.606E-06
Xe	7.303E+03	0.000E+00	5.035E-01	0.000E+00
Cs	3.720E+03	1.438E+05	2.565E-01	9.914E+00
Ba	2.308E+03	1.187E+05	1.591E-01	8.183E+00
La	1.809E+03	0.000E+00	1.247E-01	0.000E+00
Ce	3.522E+03	5.400E+03	2.428E-01	3.723E-01
Eu	1.469E+02	8.079E+03	1.013E-02	5.570E-01
Total fission products	4.867E+04	5.620E+05	3.355E+00	3.874E+01
Actinides:				
U	9.433E+05	7.544E+00	6.503E+01	5.201E-04
Pu	7.128E+03	5.698E+04	4.914E-01	3.928E+00
Total actinides	9.513E+05	5.795E+04	6.558E+01	3.995E+00

[a]VI-4 specimen contained 68.94 g U = 6.894E-5 MTU at 8.26% ^{235}U (= 5.69 g ^{235}U) before testing; all data at July 1, 1986.

Table 4. Inventories of principal nuclides in test VI-4 fuel

Nuclides	Amounts in fuel[a] (g/MTU)	Amounts in fuel[a] (Ci/MTU)	Amounts in VI-4 specimen[a] (g)	Amounts in VI-4 specimen[a] (Ci)
Fission products:				
^{85}Kr	2.569E+01	1.008E+04	1.771E-03	6.949E-01
^{90}Sr	7.923E+02	1.081E+05	5.462E-02	7.452E+00
^{106}Ru	1.292E+00	4.325E+03	8.907E-05	2.982E-01
^{110m}Ag	7.289E-04	3.464E+00	5.025E-08	2.388E-04
^{125}Sb	2.313E+00	2.389E+03	1.595E-04	1.647E-01
^{129}I	2.140E+02	3.780E-02	1.475E-02	2.606E-06
^{134}Cs	1.414E+01	1.831E+04	9.748E-04	1.262E+00
^{137}Cs	1.442E+03	1.254E+05	9.941E-02	8.645E+00
^{144}Ce	1.692E+00	5.400E+03	1.166E-04	3.723E-01
^{154}Eu	2.125E+01	5.737E+03	1.465E-03	3.955E-01
Total fission products	4.867E+04	5.620E+05	3.355E+00	3.874E+01
Actinides:				
^{234}U	8.396E+02	5.248E+00	5.788E-02	3.618E-04
^{235}U	3.253E+04	7.034E-02	2.243E+00	4.849E-06
^{236}U	8.505E+03	5.596E-01	5.863E-01	3.858E-05
^{237}U	1.630E-05	1.333E+00	1.124E-09	9.190E-05
^{238}U	9.013E+05	3.031E-01	6.214E+01	2.090E-05
Total uranium	9.433E+05	7.544E+00	6.503E+01	5.201E-04
^{238}Pu	1.138E+02	1.949E+03	7.845E-03	1.344E-01
^{239}Pu	4.582E+03	2.850E+02	3.159E-01	1.965E-02
^{240}Pu	1.753E+03	3.995E+02	1.209E-01	2.754E-02
^{241}Pu	5.274E+02	5.435E+04	3.636E-02	3.747E+00
^{242}Pu	1.519E+02	5.800E-01	1.047E-02	3.999E-05
Total plutonium	7.128E+03	5.698E+04	4.914E-01	3.928E+00
Total actinides	9.513E+05	5.795E+04	6.558E+01	3.995E+00

[a]VI-4 specimen contained 68.94 g U = 6.894E-5 MTU at 8.26% ^{235}U (= 5.69 g ^{235}U) before testing; all data at July 1, 1986.

the test apparatus was evacuated and purged with helium three times. The gas lines to the furnace, TGTs, and filter assemblies were preheated to at least 125°C.

As mentioned previously, this test was designed to duplicate as closely as possible the conditions of an in-pile test (ST-1) at SNL. Several months were devoted to a series of tests to obtain the desired temperature gradient in the induction furnace, and to insure accurate temperature measurement by optical pyrometer. The number and arrangement of induction coils and the thickness of thermal insulation around the upper end of the furnace were adjusted to approximate the desired temperature profile. The final arrangement of the induction coils is illustrated in Fig. 5. Three temperature profiles in the furnace, along with the target profile of the ST-1 test, are shown in Fig. 6.

This test was intended to investigate fission product release at fuel temperatures of 2000 to 2400 K under strongly reducing conditions. The operating conditions are summarized in Table 5, and the temperature history is shown in Fig. 7. The more important events during the test are listed in the Test Chronology, Table 6. The time periods for operation of the three collection trains, as shown in Table 6, were for Train A, 0 to 60.5 min; for Train B, 60.5 to 68.5 min; and for Train C, 68.5 min to end of test, including cooldown, at about 140 min. A preheat period was included to slowly heat the specimen to ~550 K, to approximate reactor operating temperature. Time zero was defined as that time when the controlled heating ramp was begun, with stable flow of the mixed gas (hydrogen + helium) established through the warm furnace. Temperature measurement and control were good throughout the test. The 23-min period at 1620 K was included to insure 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.

The test proceeded essentially as planned, with no unusual observations. A small change in the radiation monitor observing the fuel in the furnace occurred at a test time of ~56 min, shortly after the specimen reached the melting point of the Zircaloy cladding; this observation was later related to limited fuel relocation.

3.3 POSTTEST DISASSEMBLY AND EXAMINATION

After the test was completed, the apparatus was monitored for the distribution of radioactivity, and then disassembled. Initially, the filter assemblies and the top flange–TGT assembly, which contained the TGT liners, were removed, packaged, and transferred to another hot cell to avoid potential contamination from fuel handling. After removal of ceramic components from the outlet end of the furnace, the top end of the fuel specimen could be seen. It was observed that the top end of the specimen was at a lower elevation (by about 7 cm) than before the test, and some material, apparently broken fuel, was visible below. (The approximate elevation of the top end of the fuel specimen after the test is illustrated in Fig. 5.) However, no details of clad melting,

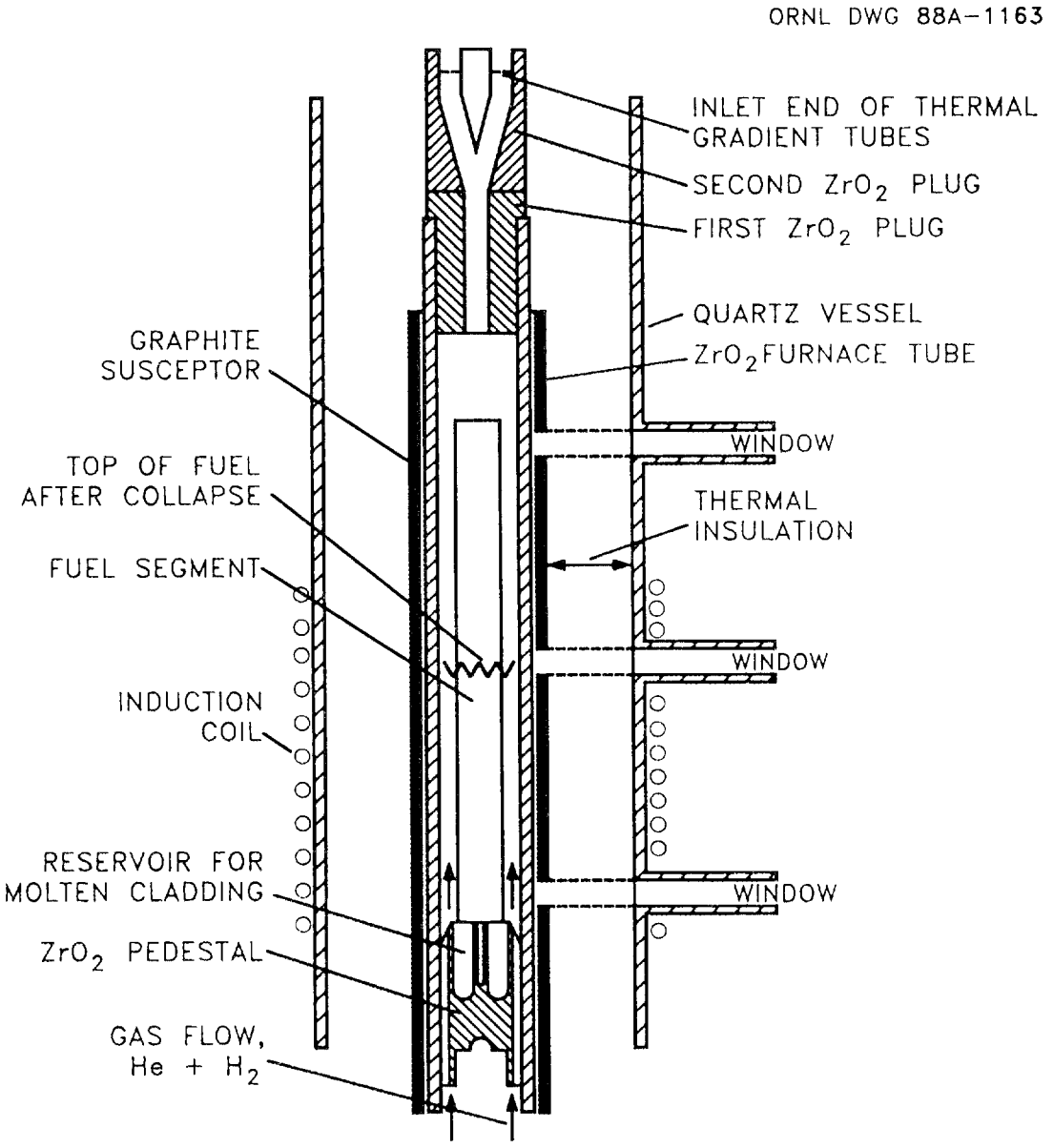

Fig. 5. Furnace details for test VI-4, showing locations of induction coils and windows for temperature measurement.

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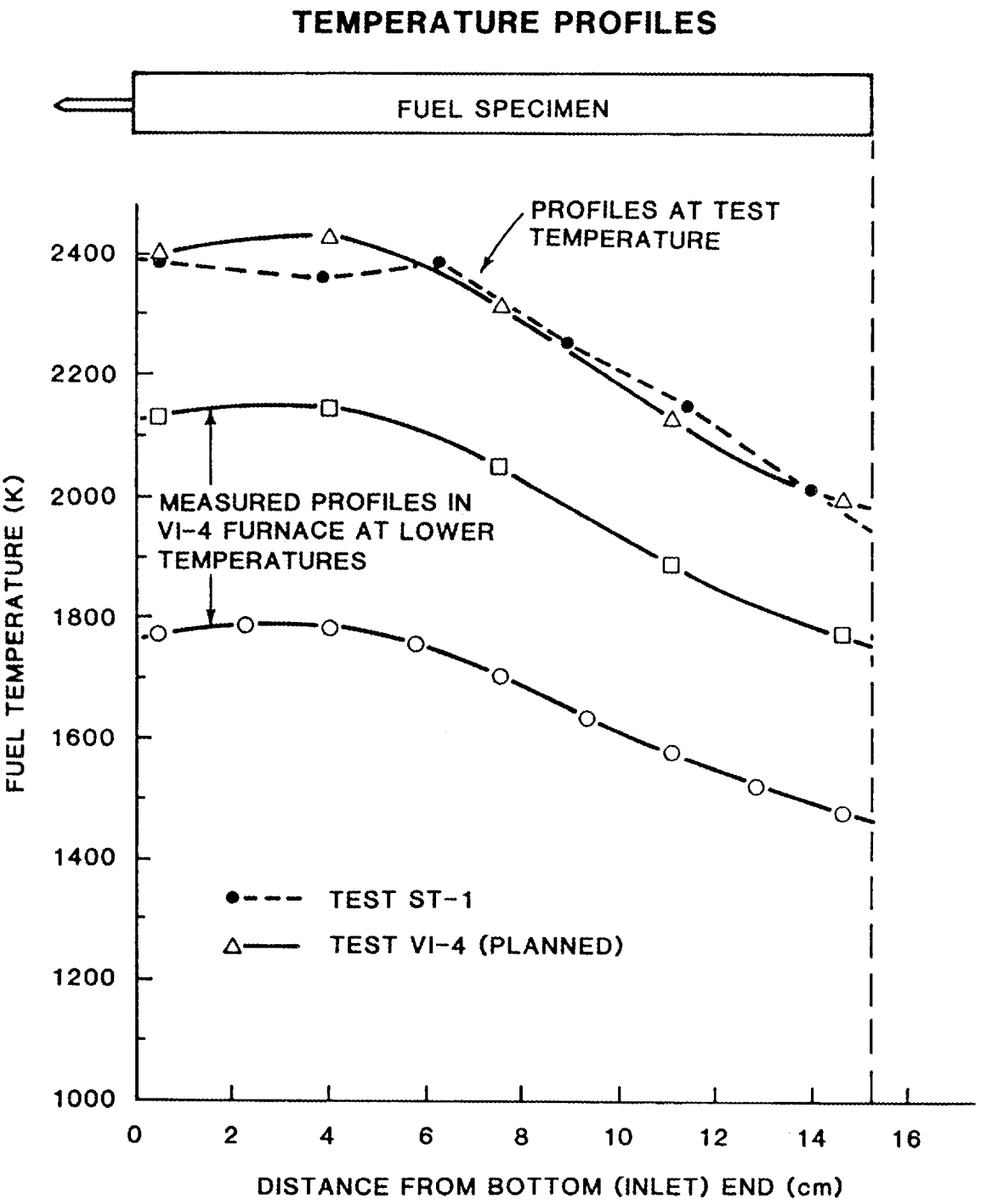

Fig. 6. Temperature profiles in the furnace during test VI-4, as developed to duplicate the profile of test ST-1.

Table 5. Operating data for test VI-4

Specimen temperature (K):	
At start of heatup ramp	~550
During 23-min period	1660
During 20-min period	2440
Heatup rate to 23-min plateau	0.7/s
Heatup rate to 20-min plateau	1.1/s
Cooldown rate	0.6/s
Time above 2000 K	35 min
Nominal gas flow rate data (L/min at 20°C, 1 bar):[a]	
50% H_2 + He recirculation system	0.10
50% H_2 + He to fuel specimen	0.81
Recirculation/purification system	1.5
Integral volume flow data (L):[b]	
H_2 + He to recirculation system	13.35
H_2 + He to fuel specimen[c]	102.54
Recirculation flow	90.8

[a]Measured by mass flowmeters.

[b]Measured by totalizers on mass flowmeters during the 125 min of gas flow into apparatus, at room temperature = 25°C.

[c]Absolute pressure in furnace during test was 99.1 kPa (743 mm Hg).

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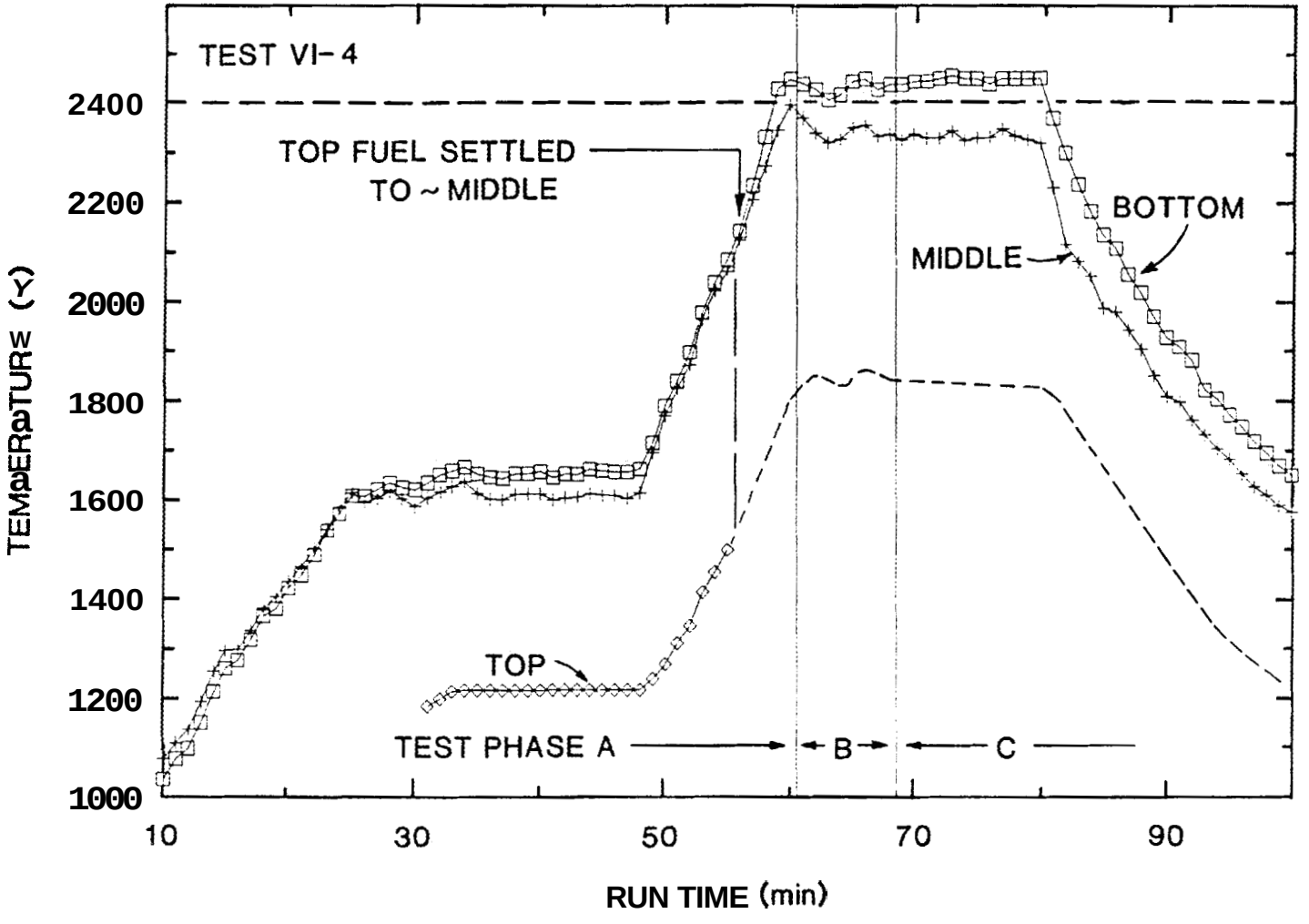

Fig. 7. Furnace temperatures during test VI-4, as measured at the top, middle, and bottom windows. Note time of fuel settling, -56 min.

Table 6. Chronology of test VI-4, May 11, 1988

Event/observation	Time		Furnace temperature corrected (K)
	Clock	Test (min)	
Load fuel into furnace	900		RT[a]
Evacuate and check	920		RT[a]
Preheat TGTs and filters	1100		RT[a]
Begin furnace preheat, with gas flow to furnace	1145		RT[a]
Stable flow and temperature	1300		~550
Test Phase A:			
Start ramp to ~1620 K	1315	0	~550
First optical pyrometer measurement	1324	9	1041
First Kr detected	1331	16	1292
First Cs on TGT, small	1333	18	1599
Reached ~1620 K plateau, hold 23 min	1340	25	1611
Resume ramp to ~2400 K	1403	48	1609
Rapid Kr increase	1412	57	2204
Reached ~2400 K plateau,	1415	60	2396
end Phase A, switch to Train B	1415.5	60.5	2383
Test Phase B:			
Begin Phase B	1415.5	60.5	2383
First Cs on filter	1418	63	2319
End Phase B, to Train C	1423.5	68.5	2438
Test Phase C:			
Begin Phase C	1423.5	68.5	2438
Kr and Cs on TGT leveling, Cs on filter still rising	1426	71	2328
End 2400 K plateau, begin controlled cooldown	1435.5	80.5	2317
Cs on filter still rising	1447	92	1755
Cs on filter leveling	1453	98	1602
Test essentially complete	1500	105	1440

[a]Room temperature, ~298 K.

fuel-cladding interaction, or fuel swelling could be distinguished by this visual inspection.

We believe that the appearance of the fuel specimen was similar to that of the unirradiated fuel in preliminary test VT-4; a posttest view of this specimen is shown in Fig. 8. The primary objectives of test VT-4 were to determine the axial temperature profile within a column of hollow UO_2 pellets, and to calibrate optical pyrometers viewing the outer surface of the ceramic furnace tube containing the specimen. It was conducted in an atmosphere of helium + 1% hydrogen at a maximum temperature of ~2200 K. As shown in Fig. 8, the cladding had melted and run down, dissolving some of the UO_2 fuel and accumulating in a crucible at the bottom end of the specimen. The region of porosity in the melted cladding is believed to have been caused by the gas flowing upward through the molten material.

To preserve the geometry of the fuel in test VI-4 for sectioning and subsequent microstructural examination, the ZrO_2 furnace tube was filled with epoxy resin before removal from the furnace. This procedure provides the added advantage of reducing the spread of highly radioactive dust during handling. Following extensive gamma spectrometric examination of the fuel-furnace tube assembly, which is discussed in Sect. 4.2.1, it was packaged and shipped to Argonne National Laboratory for sectioning (both radial and longitudinal) and detailed examination of the microstructures.

4. TEST RESULTS

4.1 TEST DATA

Furnace temperatures during the test and the three test phases are plotted as a function of test time in Fig. 7. As noted, fuel relocation (settling) occurred at ~56 min, at a temperature of ~2100 K in the lower half of the furnace and ~1500 K at the top of the specimen. Following this relocation of the fuel to the lower half of the test zone, the specimen was under nearly isothermal conditions. The release behavior of ^{85}Kr and ^{134}Cs, as functions of time and temperature, are shown in Fig. 9. As observed in previous tests, the release of cesium was delayed only slightly after that of krypton, and reached a total fractional release of ~95%.

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. 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

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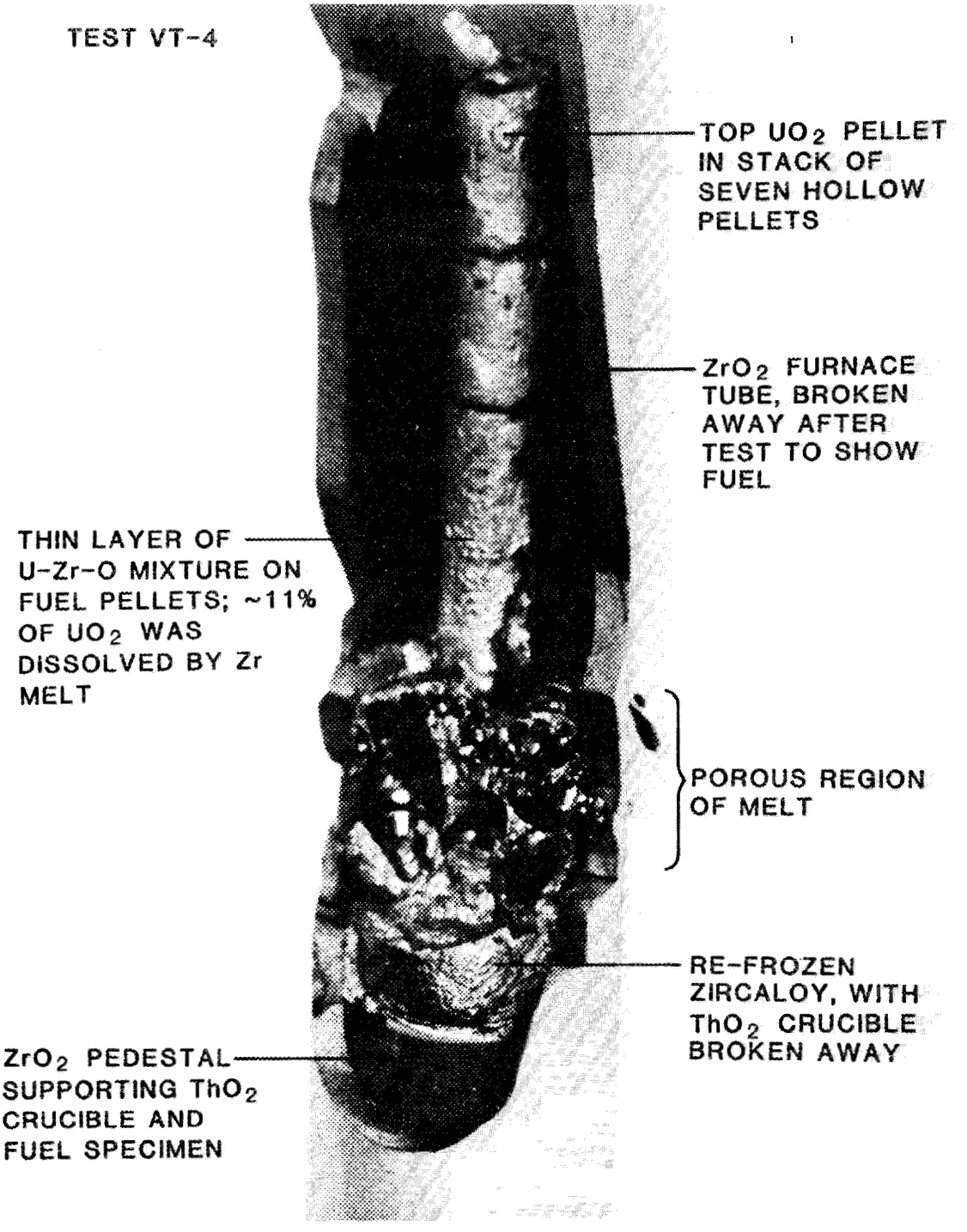

Fig. 8. View of unirradiated fuel specimen after test VT-4, showing details of cladding meltdown.

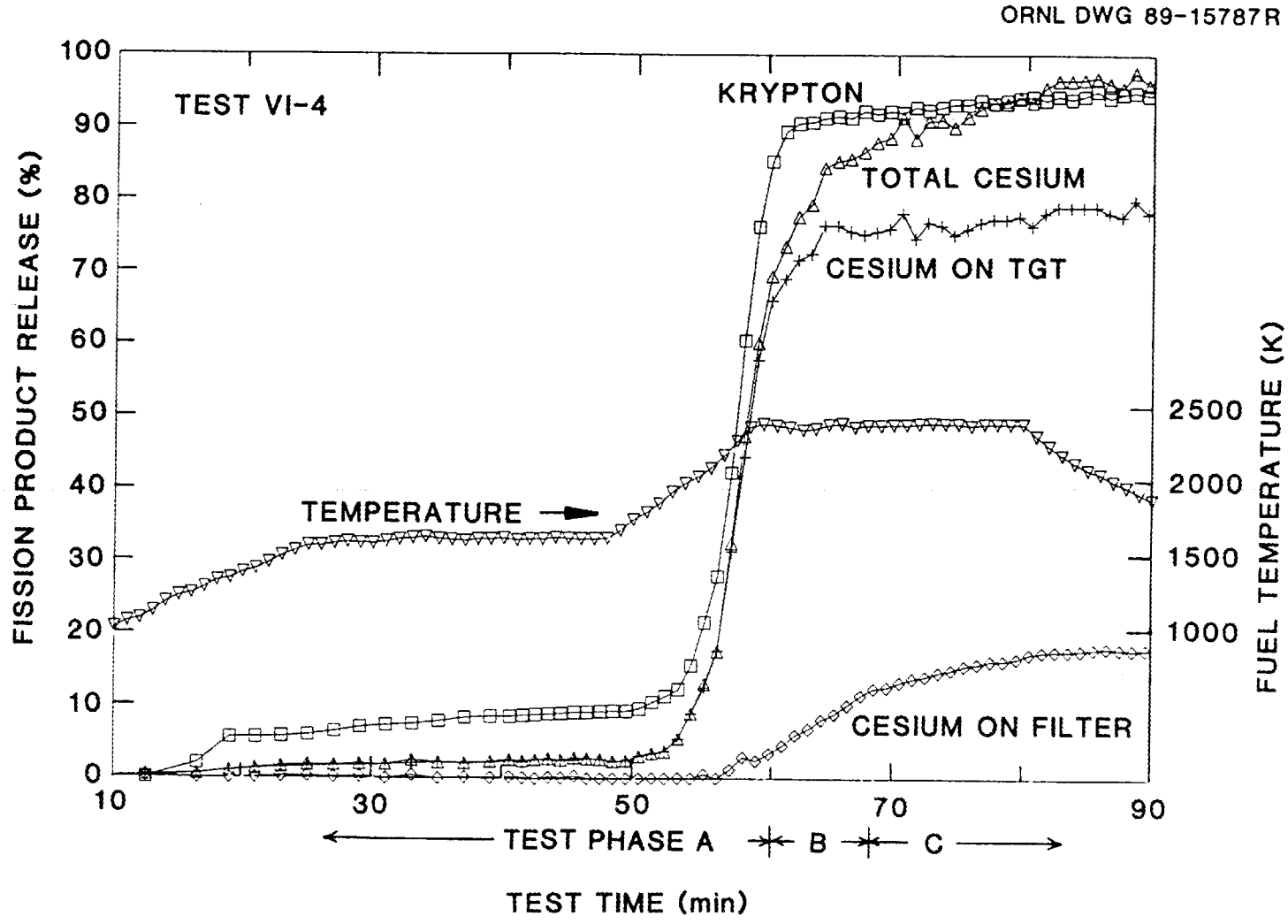

Fig. 9. Release behavior of krypton and cesium as functions of time and temperature in test VI-4.

the ubiquitous nature of cesium and the broad range of gamma-ray energies inherent to ^{134}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}Cs gamma-ray energies was developed. These shielding values were then applied to other nuclides, such as ^{106}Ru, ^{125}Sb, ^{144}Ce, and ^{154}Eu, to provide more accurate results.[11]

Pretest gamma spectrometric analysis of the 15.2-cm-long fuel specimen was used to determine the fission product inventories in the fuel. Long-lived gamma emitters – ^{106}Ru, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{154}Eu – were determined directly. A calculation by the computer program ORIGEN2 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. 10. These pretest data, based on measurements at 2- or 3-cm intervals, showed that the distributions of these major fission products were relatively uniform along the rod. Consequently, we conclude that (1) the burnup was similarly uniform and (2) the operating temperature was not high enough to cause significant migration of volatile species such as cesium during irradiation.

The posttest distribution of these same fission products, however, was entirely different. As shown in Fig. 11, all fission products (and presumably the fuel itself) were redistributed to a lower elevation. Most of the cesium had been released and there were concentrations of the other fission products near the lower end of the original fuel position. These concentrations appeared to be related to relocation of fuel and cladding, which was a combination of the processes of cladding melting, dissolution of UO_2, and rundown of the melt, and also including some disintegration and collapse of the fuel pellets. An unusual feature of the results shown in Fig. 11 is the high peak in the concentration of ^{125}Sb at the 11-cm position, combined with the very low level of ^{125}Sb at 13 to 17 cm. These results indicate that antimony was released from the fuel and combined with the molten cladding, rather than being released to the flowing hydrogen-helium gas as in the case of ^{85}Kr and ^{137}Cs in this test or as antimony would be expected to behave in a steam atmosphere. This behavior of antimony is discussed further in Sect. 4.2.1.2. Because of its low volatility, the distribution of cerium in Fig. 11 is believed to be a good indication of the distribution of the UO_2 fuel. In general, these conclusions were confirmed by the metallographic examinations conducted at Argonne National Laboratory, which are presented in Sect. 4.2.8.

Concentrations of fission product radioactivity in the region from near the top end of the furnace to the inlet end of TGT A are shown in Fig. 12. A sharp peak of ^{154}Eu (96% of that released from the fuel) was located on the ZrO_2 furnace tube immediately above the furnace specimen. Similar high concentrations of ^{125}Sb and of ^{134}Cs and ^{137}Cs were located in the two ZrO_2 plugs. As indicated, the concentration of cesium in these plugs was higher than in TGT A.

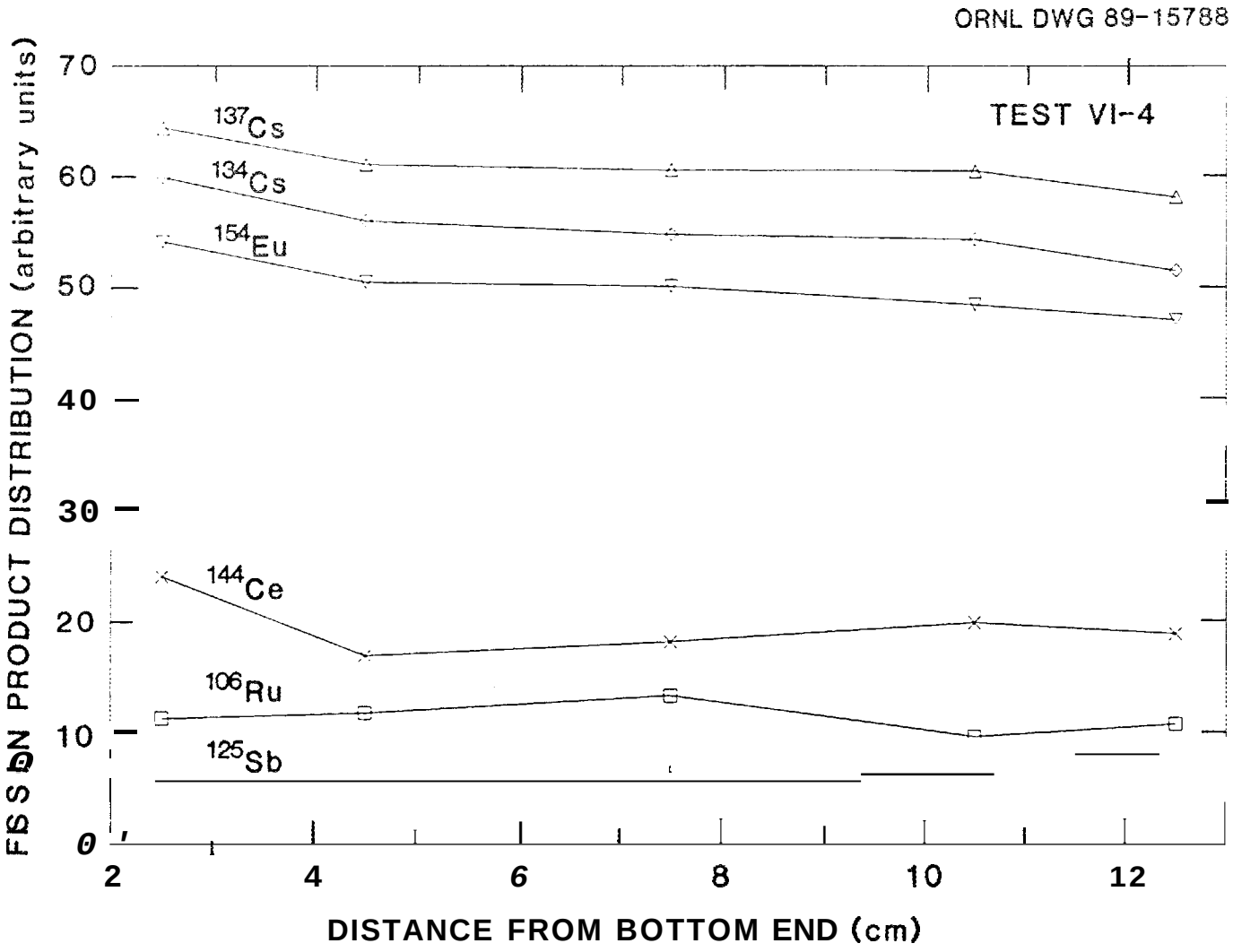

Fig. 10. Distribution of major fission products in fuel before test VI-4.

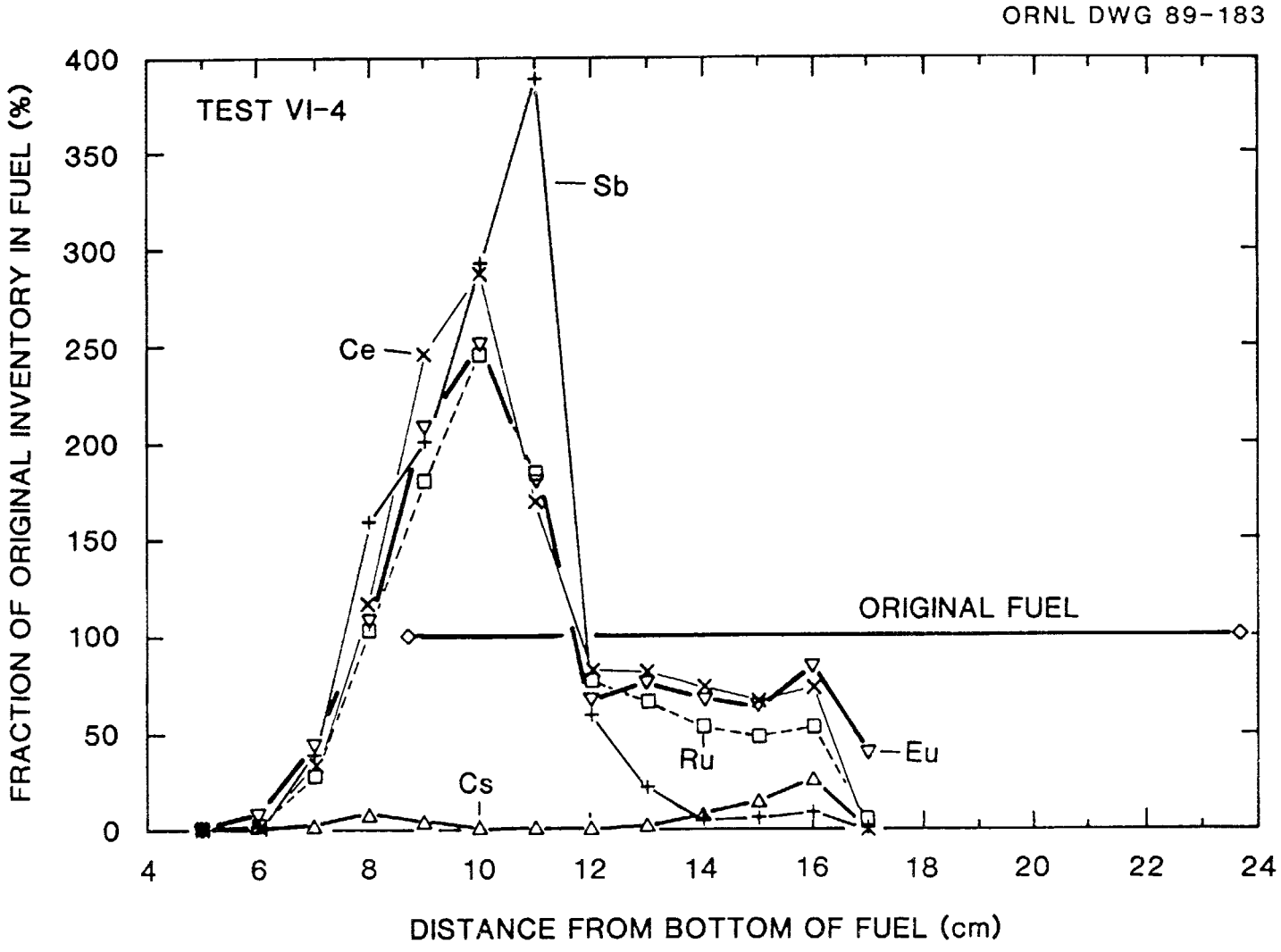

Fig. 11. Distribution of major fission products in fuel after test VI-4.

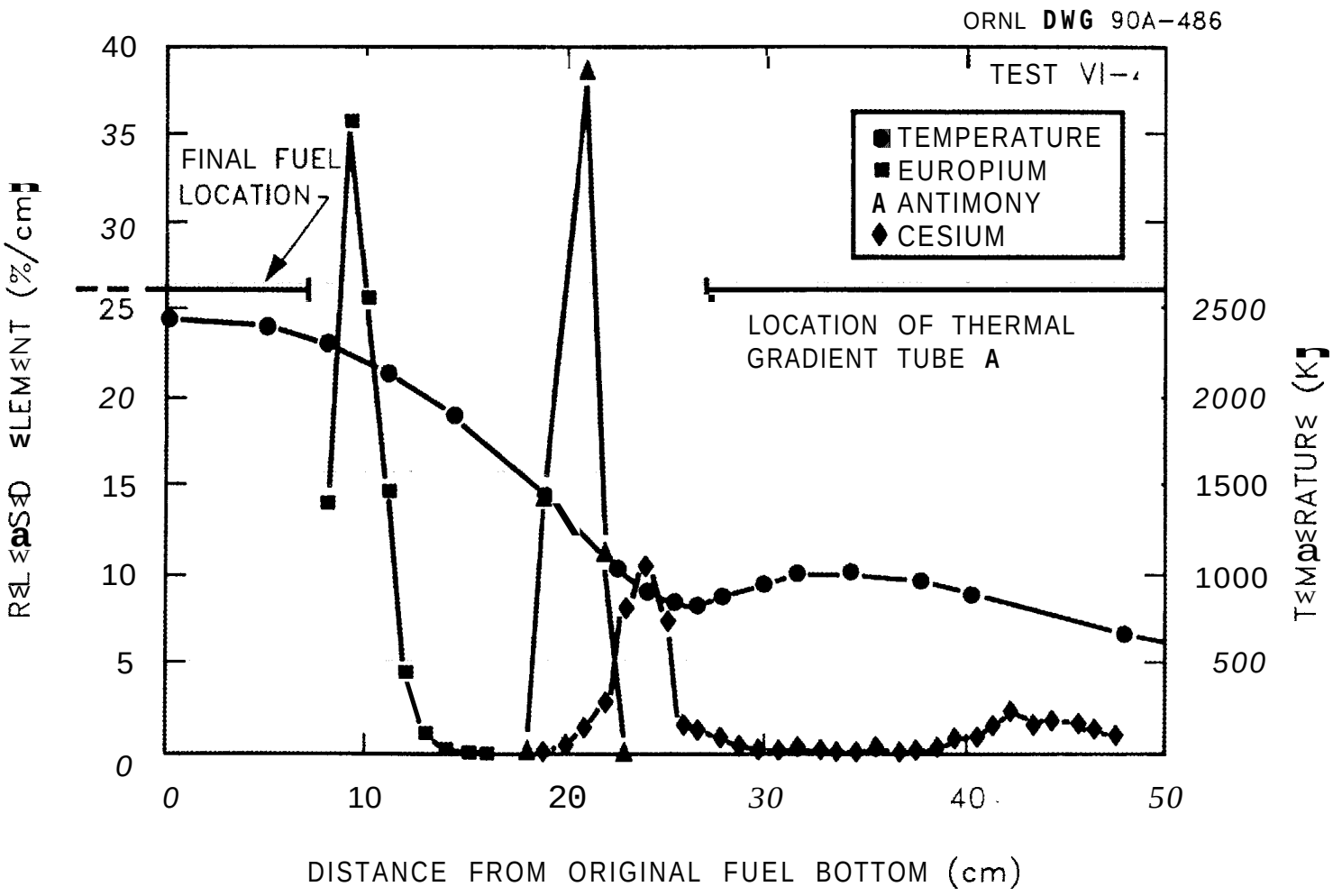

Fig. 12. High concentrations of antimony, cesium, and europium in the top end of furnace and TGT A inlet, shown with approximate temperature profile.

A continuous scan of the gross gamma radioactivity of the fuel-furnace tube assembly, which is dominated by ^{154}Eu in this case, is shown in Fig. 13. The highest concentration of radioactivity, at the top end of the posttest fuel, was cesium. Although this concentration was only a very small fraction of the original, it was still greater than the concentrations of other fission products. The smaller peak above the posttest fuel, at about 20 cm, was shown to be primarily ^{154}Eu, presumably deposited from the gas phase onto the ZrO_2 furnace tube after being released from the fuel at a lower elevation. The distribution of gamma radioactivity within the ZrO_2 plugs at the top end of the furnace is shown in Fig. 14. As shown in Tables 7 and 8, most of this radioactivity was cesium, apparently deposited and/or reacted primarily with the second (three-hole) plug, which was at a somewhat lower temperature than the first (one-hole) plug.

As has been typical of these tests of high burnup, long decayed fuel, ^{137}Cs and ^{134}Cs were the dominant gamma radioactivities in almost all samples, and interfered with the analysis of less abundant fission products. The release behavior of krypton and cesium as functions of time and temperature is illustrated in Fig. 9. These curves show that very little release of krypton and cesium occurred during the last half of the high-temperature period. A summary of the fractional release results for Kr, Sb, Cs, and Eu, as determined by gamma spectrometry, is presented in Table 9. On-line analysis indicated that the cumulative krypton release value reached 95% of the maximum about 3 min after the test temperature of 2400 K was attained. (An estimated 10% of inventory was released from the UO_2 to the pellet-cladding gap during irradiation and was released when the rod was sectioned.)

4.2.1.1 Behavior of cesium

Although no data for ^{134}Cs are shown, the agreement with ^{137}Cs was consistently good. The distribution of cesium within the test apparatus is shown in detail in Table 7. The largest fraction of the cesium (48%) was retained in the furnace, principally on the ZrO_2 ceramics at the furnace outlet, where the temperature was ~1200 to 1500 K. In earlier tests in steam at comparable temperatures, much less cesium was retained in this region, suggesting that the reducing conditions in the hydrogen atmosphere provided more opportunity for reaction between the cesium and the ZrO_2. Some 29% of the released cesium was collected during test Phase A, the heatup period to 2400 K, and smaller fractions (~11% each) were collected during Phases B and C. Unlike the tests in steam, the cesium fractions collected on the filters were not consistently larger than the fractions collected in the TGTs. During Phase A, 89% of the cesium was collected on the TGT, and thus is concluded to have been in vapor form. Conversely, during the first 8-min period at 2400 K (Phase B), only 21% of the cesium was collected as vapor. The reason for this apparently drastic change in cesium form is not clear, but most likely reflects a similarly drastic change in atmospheric conditions. Following meltdown of the strongly oxidizing Zircaloy cladding, the oxidation potential at the surface of the fuel probably increased significantly with a resulting shift toward a larger fraction of the

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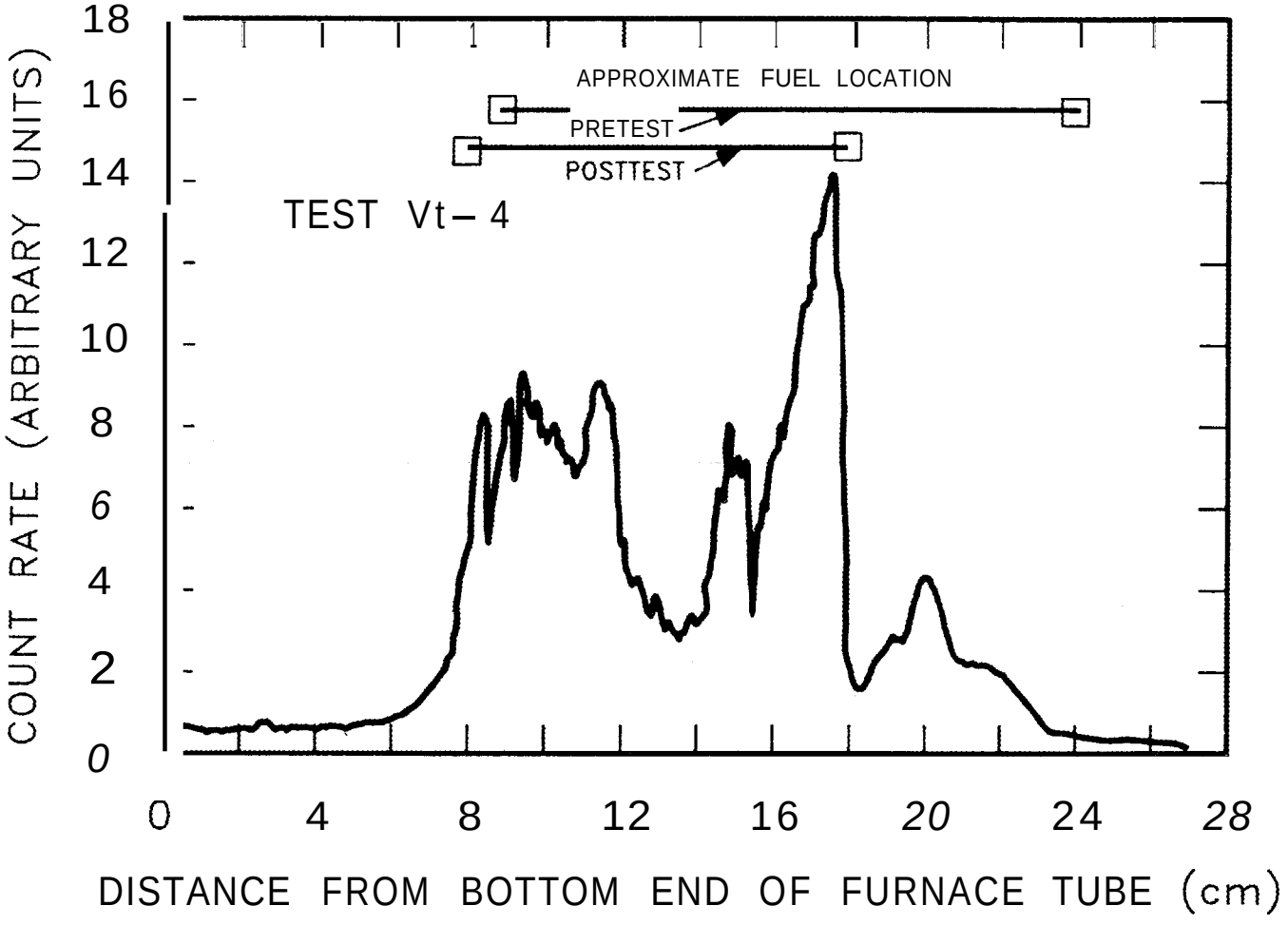

Fig. 13. Detailed profile of gross gamma radioactivity, which was dominated by ^{137}Cs and ^{154}Eu, in the fuel region after test VI-4.

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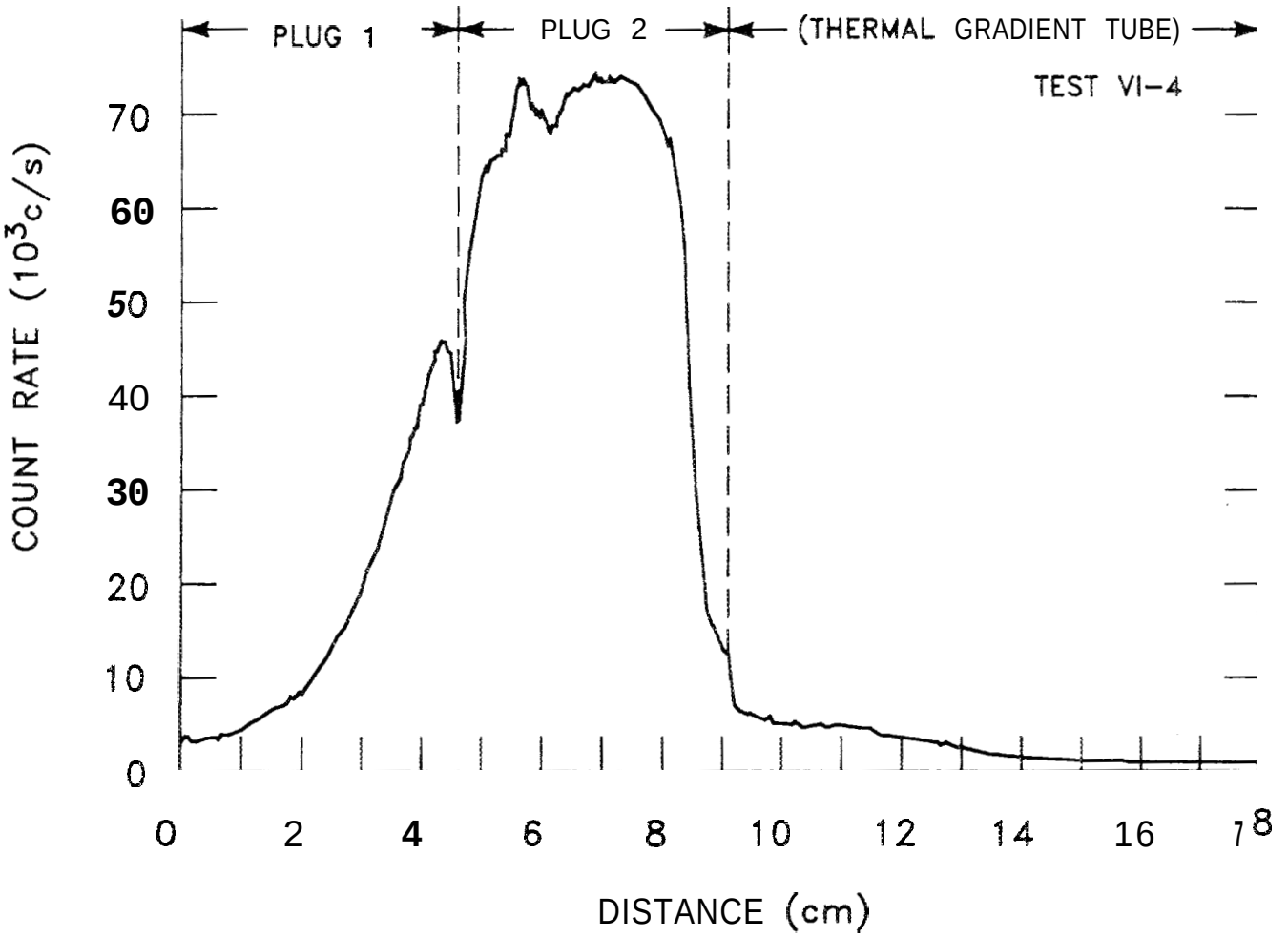

Fig. 14. Relative radioactivity in ZrO_2 plugs at top end of furnace after test VI-4.

Table 7. Fractional release and distribution of cesium in test VI-4

Location	Cesium found at each location			
	Amount (mCi ^{137}Cs)	Amount (mg Cs)[a]	Inventory[b] (%)	Released (%)
Furnace components:				
Furnace bottom	92	2.62	1.06	1.11
Furnace tube	15	0.43	0.17	0.18
First ZrO_2 plug	230	6.56	2.66	2.77
Second ZrO_2 plug	3470	99.00	40.12	41.81
ZrO_2 donut	140	3.99	1.66	1.69
Exit flange	59	1.69	0.68	0.71
Total furnace	4006	114.29	46.32	48.26
Collection train A:				
TGT A	2148	61.28	24.84	25.88
TGT – filter line	42	1.19	0.48	0.50
First prefilter	191	5.45	2.21	2.30
Second prefilter	37	1.06	0.43	0.45
HEPA filters	6	0.16	0.07	0.07
Total train A	2423	69.14	28.02	29.20
Collection train B:				
TGT B	192	5.48	2.22	2.31
TGT – filter line	70	2.00	0.81	0.84
First prefilter	540	15.41	6.24	6.51
Second prefilter	99	2.82	1.14	1.19
HEPA filters	14	0.40	0.16	0.17
Total train B	915	26.10	10.58	11.03
Collection train C:				
TGT C	510	14.55	5.90	6.15
TGT – filter line	23	0.66	0.27	0.28
First prefilter	340	9.70	3.93	4.10
Second prefilter	74	2.11	0.86	0.89
HEPA filters	8	0.23	0.09	0.10
Total train C	955	27.25	11.04	11.51
Total found outside fuel	8299	236.78	95.96[c]	100.00
Total in fuel after test	350	9.99	4.05	4.22
Total in fuel before test	8649	246.76	100.00	

[a]Based on analysis of ^{137}Cs in the fuel and ORIGEN calculations of other cesium nuclides: 0.02853 mg Cs/mCi ^{137}Cs.

[b]Inventories based on measured ^{137}Cs data.

[c]Comparison of pretest and posttest fuel analyses indicated a release of 96.1%.

Table 8. Fractional release and distribution of antimony and europium in test VI-4

Location	^{125}Sb (μCi)	^{125}Sb Inventory[a] (%)	^{154}Eu (μCi)	^{154}Eu Inventory[a] (%)
Furnace components:				
Furnace tube	23	0.023	31300	12.776
First ZrO_2 plug	2400	2.400	75	0.031
Second ZrO_2 plug	900	0.900	80	0.033
ZrO_2 donut	0	0.000	0	0.000
Exit flange	0	0.000	0	0.000
Total furnace	3323	3.323	31455	12.839
Collection train A:				
TGT A	5	0.005	260	0.106
TGT – filter line	11	0.011	629	0.257
First prefilter	113	0.113	320	0.131
Second prefilter	0	0.000	37	0.015
HEPA filters	0	0.000	7	0.003
Total train A	129	0.129	1253	0.511
Collection train B:				
TGT B	22	0.022	37	0.015
TGT – filter line	62	0.062	14	0.006
First prefilter	31	0.031	0	0.000
Second prefilter	0	0.000	0	0.000
HEPA filters	0	0.000	0	0.000
Total train B	115	0.115	51	0.021
Collection train C:				
TGT C	190	0.190	88	0.036
TGT – filter line	18	0.018	1	0.000
First prefilter	1	0.001	2	0.001
Second prefilter	96	0.096	2	0.001
HEPA filters	0	0.000	0	0.000
Total train C	305	0.305	93	0.038
Total found outside fuel	3,872	3.872	32,852	13.409
Total in fuel after test	68,000	68.000	182,000	74.286
Total in fuel before test	100,000[a]	100.000	245,000[a]	100.000

[a]Inventories based on measured data.

Table 9. Summary of fission product release data for test VI-4[a]

Location	Operating time at T >2000 K (min)	Fraction of fission product inventory found (%)			
		^{85}Kr	^{125}Sb	^{137}Cs	^{154}Eu
Furnace components	35	0	3.32	46.3	12.8
Collection train A:	8				
TGT A		0	0.005	24.8	0.106
Filters		0	0.124	3.20	0.405
Total train A		0.00	0.13	28.0	0.51
Collection train B:	8				
TGT B		0	0.022	2.22	0.015
Filters		0	0.093	8.36	0.006
Total train B		0.00	0.12	10.58	0.02
Collection train C:	19.5				
TGT C		0	0.19	5.90	0.036
Filters		0	0.115	5.14	0.002
Total train C		0.00	0.31	11.04	0.04
Cold charcoal	35	85	0	0	0
Total for test	35	85	3.87	95.9	13.4

[a]Inventories based on pretest gamma analysis of the fuel and ORIGEN2 calculations.

released cesium being in aerosol form, rather than predominantly vapor form.

4.2.1.2 Behavior of antimony and europium

The indicated release values for ^{125}Sb and ^{154}Eu are shown in Table 8. Although these nuclides are of relatively low yield and are not among the most hazardous, they provide additional information about fission product release which is potentially valuable. Both of these nuclides have intermediate half-lives and strong gamma rays, which make them easily detected. The total release of antimony (3.87%) was at least a factor of 10 lower than would have been expected in steam at this temperature. (Antimony releases in tests VI-1, VI-2, and VI-3 varied between 33 and 89%.)[11-13] In addition, 86% of the released antimony was retained in the furnace, as opposed to an average of ~20% in the aforementioned tests in steam, providing evidence that the released antimony was in a different chemical form in this test in hydrogen. We believe that antimony behaves much like tellurium, in that it is released from the fuel but combines with metallic Zircaloy if it is available.[19]

Whereas much less antimony was released in test VI-4 than in the previous tests in steam, much more europium (13.4 vs ~0%) was released in test VI-4. Almost all (96%) of the released europium remained in the furnace, mostly on the ZrO_2 furnace tube before reaching the ZrO_2 end plugs where the antimony was concentrated. This behavior suggests that, in this test in hydrogen, europium was released in the elemental form, which is more volatile than the oxide which would have been expected in a steam atmosphere. Furthermore, both elements tended to be deposited within the furnace in temperature regions consistent with their vapor pressures, showing little tendency to form more transportable, and potentially more hazardous, aerosol forms.

4.2.2 Analysis for Iodine

Since iodine has no long-lived, gamma-emitting nuclides, analytical methods other than gamma spectrometry must be used. Neutron activation of ^{129}I to ^{130}I, which can be counted easily, is a proven, sensitive technique. Because iodine forms dissolve readily in basic solutions to form stable iodides, the collector components from this test were leached with a basic solution ($NH_4OH + H_2O_2$) to remove this iodine for analysis. Iodine adsorbed on the heated charcoal in the filter packages is analyzed by direct activation of the charcoal adsorber. Unfortunately, no facilities for activation analyses to determine the amount of iodine in these leach solutions and on the charcoal have been available at ORNL since the test was conducted. These analyses will be carried out as soon as possible, and the results of several tests (VI-2, VI-3, and VI-4) will be reported and evaluated later.

4.2.3 Thermal Gradient Tube Deposits

After removal from the TGTs, the platinum liners were analyzed by gamma spectrometry. Each tube liner was counted through 1.27-cm- and 3.81-cm-thick lead plates to determine the total activity. The radioactivity profiles were obtained by successively counting 1-cm sections through a 1.0-cm-wide collimator, which permitted nuclide analysis, then followed by continuous scanning through a 0.2-cm-wide collimator to accurately determine the distribution of gross gamma radioactivity.

The abundant ^{134}Cs and ^{137}Cs were easily measured in all cases. Detailed data for ^{137}Cs are shown in Table 7. The data from the 1-cm scans without any lead shielding provided the best sensitivity; the ^{137}Cs data for the three TGTs are shown in Fig. 15. As opposed to the small amounts of ^{125}Sb which were found in most of the liners from Trains B and C in tests VI-1, VI-2, and VI-3, no ^{125}Sb was found in the TGT liners of this test. As discussed in Sect. 4.2.1.2, the very low release of antimony in test VI-4 is attributed to the hydrogen atmosphere, which prevented oxidation of the Zircaloy cladding, compared to the steam atmospheres used in the earlier tests.

The detailed gross gamma profiles in the three TGTs are compared in Fig. 16. As noted previously, Figs. 15 and 16 show clearly that much more cesium was collected during test Phase A than during the subsequent phases. The largest concentrations of cesium occurred at deposition temperatures of 700 to 850 K in TGTs A and B. Earlier work in steam atmospheres showed that CsI and CsOH would be deposited in this temperature range.[20] Although CsOH would not be expected to form in the hydrogen atmosphere in test VI-4, CsI would be expected. Since the available results from spark source mass spectrometric (SSMS) analyses showed high concentrations of iodine and Cs-to-I ratios approaching one in this same temperature region for both TGT A and TGT B, we concluded that large fractions of the deposits were CsI. In other regions of the TGTs, the Cs-to-I ratios were $\gg 1$, indicating a large excess of cesium.

4.2.4 Results of SSMS Analyses

Small smear samples of the material deposited on the TGTs and the filters were collected on graphite electrodes and analyzed by SSMS. This technique has advantages in that it is very sensitive, not only for the radionuclides but also for the stable elements, including structural materials and any impurities in the system. Its disadvantage is relatively low precision, a factor of about 2. These data may be converted to absolute values by comparison with either the ^{137}Cs data or known standards of erbium injected during the vaporization step. Although the samples from the TGTs were collected over a length of ~1 cm and those from the filters were intended to be representative of the entire surface, it was recognized that obtaining a truly representative sample was very unlikely.

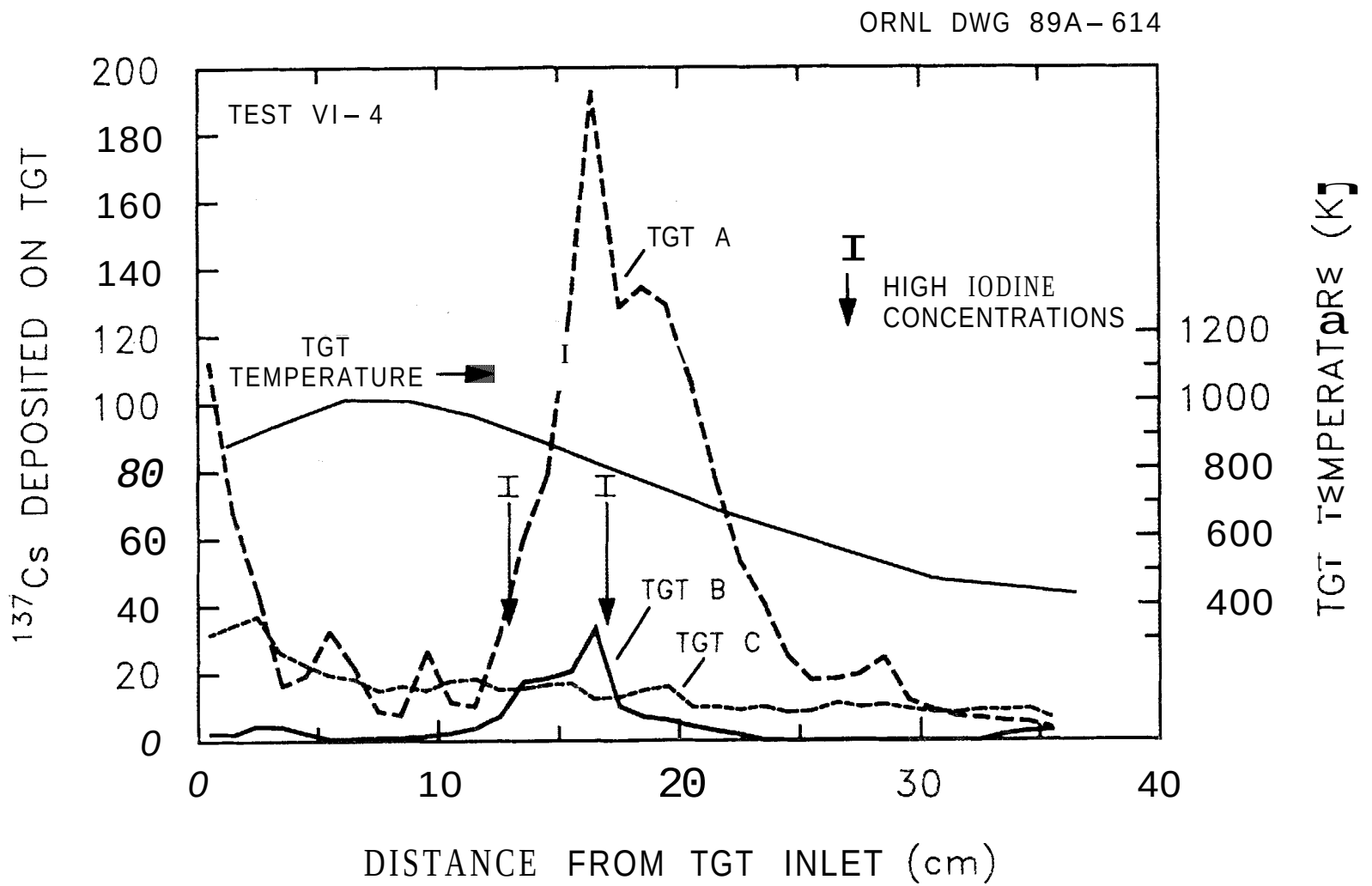

Fig. 15. Distributions of ^{137}Cs in TGTs A, B, and C from test VI-4. Note the locations where high concentrations of iodine were found.

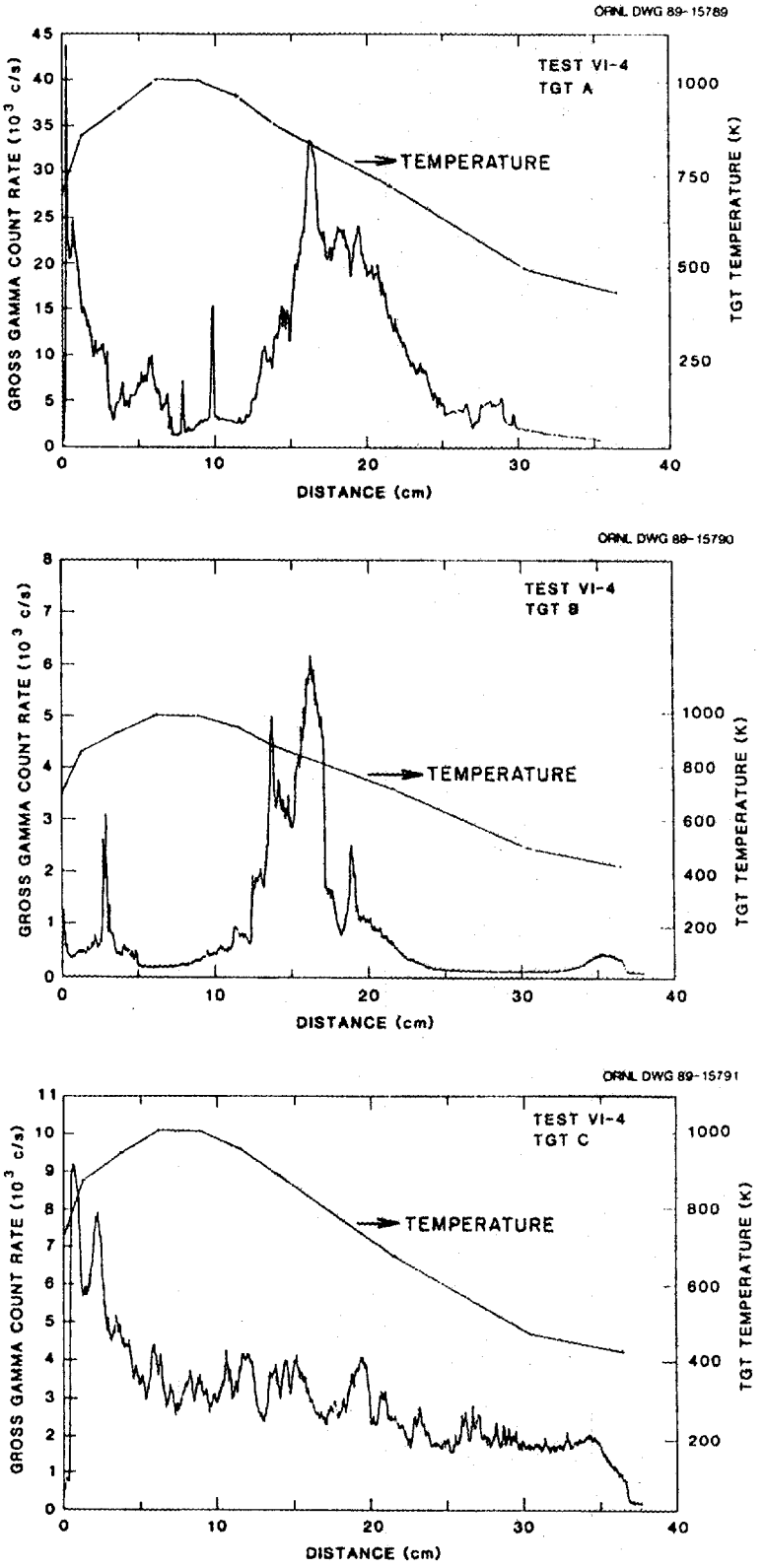

Fig. 16. Detailed distributions of gamma radioactivity, primarily ^{134}Cs and ^{137}Cs, in TGTs from test VI-4.

The results of SSMS analyses of samples from test VI-4 are summarized in Table 10. These data were normalized to the masses of material determined by weighing (see Sect. 4.2.7). Milligrams of element and percent of total are shown for each sample. During test Phase A (heatup to 2400 K, including meltdown of the Zircaloy cladding), the largest amounts of material and the broadest range of elements were collected. The fission products Br, Rb, Sr, Ag, Te, I, Cs, and Ba were found in varying amounts in these deposits. Although there was generally an excess of Cs compared to I, the Cs/I ratio in the central region of TGT A was near 1 (see Fig. 15). This is significant because it suggests the presence of CsI at the expected deposition temperature, ~700 to 850 K. The presence of Ba and Sr, and also U, in Train A deposits, and not in those from Trains B and C, which experienced more time at the highest temperature, seems surprising initially. We concluded that the strongly reducing conditions, and also the dissolution and rundown of some UO_2 with the melt, enhanced the release (vaporization) of these elements, all of which are much more volatile in the elemental form compared to the oxide form. Following meltdown, more oxygen, perhaps from the UO_2, must have been available to increase the oxidation potential. In addition, zirconium was present in relatively large amounts in Train A deposits, but only in a small amount in Train B, and not at all in Train C deposits. The presence of more tellurium in Train C deposits than in Train A and B deposits is consistent both with these conclusions and with the known behavior of tellurium.[19]

The observations of small amounts of Fe, Al, and S generally throughout the collection apparatus is in agreement with previous test results. All have been shown to be present as impurities in the structural ceramics of the furnace, and the Zircaloy cladding contained a small fraction of iron. Of these elements, only iron would be expected in a reactor coolant circuit. The presence of aluminum and sulphur impurities at these low concentrations is not believed to have a significant effect on fission product behavior in our tests.

4.2.5 Results of ICP-ES Analyses

Samples of the acidic leach solutions from the filters were submitted for inductively coupled plasma-emission spectrometry (ICP-ES) analysis for non-gamma-emitting elements. This technique appears to be well suited for measuring a number of the fission products. Although it is, unfortunately, not useful for I analysis, it appears to be promising for the measurement of Sr, Mo, Te, Ba, and perhaps for U. Because of the high levels of radiocesium in all samples, large dilutions were required to avoid excess radiation dose to the analyst. Unfortunately, these large dilutions resulted in poor sensitivities for measuring the elements of interest, and the results from ICP-ES analysis of test VI-4 samples, which are summarized in Table 11, are disappointing. In many samples, the concentrations of molybdenum and tellurium were below the limit of detection. Consequently, we are investigating ways of eliminating a major fraction of this cesium interference, so that sensitivities for measurement may be improved.

Table 10. Spark source mass spectrometry data for test VI-4[a]
(data normalized to total mass on TGTs and filters)

	Collection train A						Collection train B						Collection train C					
Sample No.	TGT A1		TGT A2		AG1		TGT B1		TGT B2		BG1		TGT C1		TGT C2		CG1	
Location (cm)[b]	13.5		17.5		Prefilter		13.5		17.5		Prefilter		13.5		17.5		Prefilter	
Temperature (K)	900		790		400		900		800		400		900		790		400	
	(mg/cm)	(%)	(mg/cm)	(%)	(mg)	(%)	(mg/cm)	(%)	(mg/cm)	(%)	(mg)	(%)	(mg/cm)	(%)	(mg/cm)	(%)	(mg)	(%)
Fission products:																		
Cs	1.4	32	5	19	5	17	0.4	51	0.7	32	16	50	0.5	62	0.4	52	11	55
Rb	0.4	9	1.7	6	1	3	0.08	10	0.13	6	3	9	0.11	14	0.09	12	1.8	9
I	1.6	36	4	15	1.6	6	0.1	13	1.1	50	0.3	1	0.05	6	0.1	13	0.16	1
Br	0.5	11	1.2	4	1.4	5	0.08	10	0.2	9	1.4	4	0.02	2	0.03	4	0.7	4
Te	0	0	0	0	2	7	0	0	0	0	0.9	3	0.1	12	0.06	8	2	10
Ag	0	0	0.02	0	1.4	5	0	0	0	0	0.0	0	0	0	0	0	0	0
Ba	0	0	0.8	3	3	10	0	0	0	0	0.0	0	0	0	0	0	0	0
Sr	0	0	0.2	1	0.5	2	0	0	0	0	0	0	0	0	0	0	0	0
Total	3.9	89	13	48	16	55	0.66	85	2.1	95	22	69	0.78	96	0.68	88	16	80
Cs/I	0.9		1.3		3.5		4		0.62		58		9		4		70	
Reactor materials:																		
Sn	0	0	0.2	1	0.8	3	0	0	0	0	0.0	0	0.01	1	0	0	0	0
Zr	0	0	4	15	5	17	0.01	1	0	0	3	9	0.001	0	0	0	0	0
U	0	0	9	33	2	7	0	0	0	0	0	0	0	0	0	0	0	0
Fe	0.3	7	0.2	1	0.2	1	0.01	1	0.04	2	0.5	1	0.004	0	0.01	1	0.8	4
Total	0.3	7	13	48	8	28	0.02	3	0.04	2	3.5	11	0.015	1	0.01	1	0.81	4
Other materials:																		
Al	0.01	0	0.04	0	0.5	2	0.01	1	0.07	3	1.4	4	0.004	0	0.03	4	0.4	2
S	0.2	5	0.2	1	5	17	0.11	14	0.11	5	5	16	0.04	5	0.06	8	2	10
Total	0.21	5	0.24	1	5.5	19	0.12	15	0.18	8	6.4	20	0.044	5	0.09	12	2.4	12
Total all materials	4.4	100	26	97	29	101	0.80	102	2.3	105	32	100	0.84	102	0.78	101	19	96

[a]Precision of all data is about a factor of 2.
[b]Distance from TGT inlet.

Table 11. Summary of ECP-ES data for test VI-4

Location	Mass found (mg)			Fraction of inventory found (%)[a]		
	Ba	Mo[b]	Te[b]	Ba	Mo[b]	Te[b]
Furnace components:						
Furnace tube	18.5	2.3	19.7	1.16	0.07	4.85
ZrO_2	5.28	10.26	87.98	0.33	0.30	21.67
Total	24	13	108	1.5	0.4	26.5
Collection train A:						
TGA A	3	0.060	0.520	0.19	0.002	0.13
Filters	1.44	0.460	3.920	0.09	0.014	0.97
Exit block fraction[c]	0.765	0.128	0.409	0.05	0.004	0.10
Total train A	5.3	0.6	4.8	0.3	0.020	1.2
Collection train B:						
TGT B	3.49	0.060	0.53	0.22	0.002	0.13
Filters	3.35	7.22	61.92	0.21	0.214	15.25
Exit block fraction[c]	0.011	0.002	0.309	0.00	0.000	0.08
Total train B	6.9	7.3	62.8	0.4	0.22	15.5
Collection train C:						
TGT C	3.76	0.06	0.54	0.24	0.002	0.13
Filters	1.3	1.3	11.12	0.08	0.039	2.74
Exit block fraction[c]	0.004	0.001	0.382	0.00	0.000	0.09
Total train C	5.1	1.4	12.0	0.3	0.04	3.0
Total for test	41	22	187	2.6	0.6	46.1

[a]Inventories based on ORIGEN2 calculations: 159 mg Ba, 337 mg Mo, and 40.6 mg Te in fuel.

[b]All molybdenum and tellurium values are less than, based on limits of detection.

[c]Exit block masses: 0.78 mg Ba, 0.13 mg Mo, and 1.1 mg Te; assumes ^{154}Eu distribution for Ba and Mo, and ^{125}Sb distribution for Te.

4.2.6 Uranium and Plutonium Data

The determination of fuel release is important, 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. (The fission products are, in general, closely associated with the fuel and may be carried along with the heavier atoms.) The data, obtained by fluorimetric analysis for uranium and alpha particle energy analysis for plutonium, are summarized in Table 12. The precision of measurement for uranium is significantly better (~10%) than that for plutonium (~30%).

These data show that in samples of material from the furnace, the TGTs, and the filters, neither uranium nor plutonium could be detected at all locations. However, both elements were measured in all three samples from the furnace, and neither was detected in collection Train C. The largest fraction of uranium (>90% of that released), but no plutonium, was found on the Train B filters. These data indicate that, in a hydrogen atmosphere, the transport behavior of the two elements is quite different. On the other hand, the results from test VI-2, at 2300 K in excess steam, showed rather similar behavior (i.e., a close association of the two elements.)[12] In both cases, however, the total fractional release of uranium was ~10 times greater than that for plutonium.

4.2.7 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 for any unusual appearances, then packaged for weighing. The masses of material collected at the various locations are listed in Table 13 and illustrated in Fig. 17. The masses of material collected during Phases B and C were each about half the mass collected during Phase A (0.202 g), and 65% of this amount (0.131 g)was found in TGT A. Furthermore, the total masses deposited were much less than those in any of the VI tests in steam.

Most of the material deposited in the TGT liners is believed to be a result of vapor condensation, and the material deposited on the filters probably was transported primarily as aerosol. The average mass concentration of total vapor and aerosol during each test phase was calculated from the masses collected and the total gas flow during that period (see Table 13). To avoid the distortion of the heatup and cooldown periods, it was assumed that no aerosol was formed at temperatures below 2050 K. These data show that the average mass concentration of the airborne material was highest during test Phase A, and that this average mass concentration declined rapidly during the test as the supply of the more volatile material was depleted.

Table 12. Release of uranium and plutonium in test VI-4

Location	U found[a]		Pu found[a]	
	(mg)	(%)[b]	(mg)	(%)[b]
Furnace components:				
ZrO_2 tube above fuel	2.0	0.0031	0.3	0.0611
First ZrO_2 plug	9.2	0.0142	0.015	0.0031
Second ZrO_2 plug	4.1	0.0063	0.017	0.0035
Total in furnace	15.3	0.0230	0.33	0.0676
Train A:				
TGT A	24.5	0.0377	0.013	0.0026
Filters	<0.2	0.0000	0.09	0.0183
Train B:				
TGT B	<0.06	0.0000	<0.0001	0.0000
Filters	457	0.7031	<0.003	0.0000
Train C:				
TGT C	<0.06	0.0000	<0.0001	0.0000
Filters	<0.6	0.0000	<0.006	0.0000
Total all	<498	0.7638	<0.435	0.0886

[a]Uranium analyses by fluorimetric, plutonium analyses by alpha pulse height, based on ^{238}Pu; precision ~10% for U and ~30% for Pu.

[b]Based on ORIGEN2 calculations, postirradiation inventories in the fuel were 65.0 g U and 0.491 g Pu.

Table 13. Vapor and aerosol deposits[a] in test VI-4

	Train A (g)	Train B (g)	Train C (g)	Totals (g)
Thermal gradient tube	0.130	0.002	0.029	0.161
Filters:				
Prefilter 1[b]	0.048	0.060	0.056	
Prefilter 2	0.015	0.018	0.016	
HEPAs	0.000	0.007	0.000	
Connecting tubes	0.009	0.008	0.003	
Total filters	0.072	0.093	0.075	0.240
Total: TGT and filters	0.202	0.095	0.104	0.401
Average aerosol concentration[c] (g/m^3)	30	9.7	5.1	11

[a]Precision = ±0.003 g.

[b]Includes estimated mass of deposits in connecting tubes, based on ^{137}Cs data.

[c]Assumes all aerosol was formed at temperatures ≥2050 K, and that the average temperature in the TGTs and filters was 423 K (150°C).

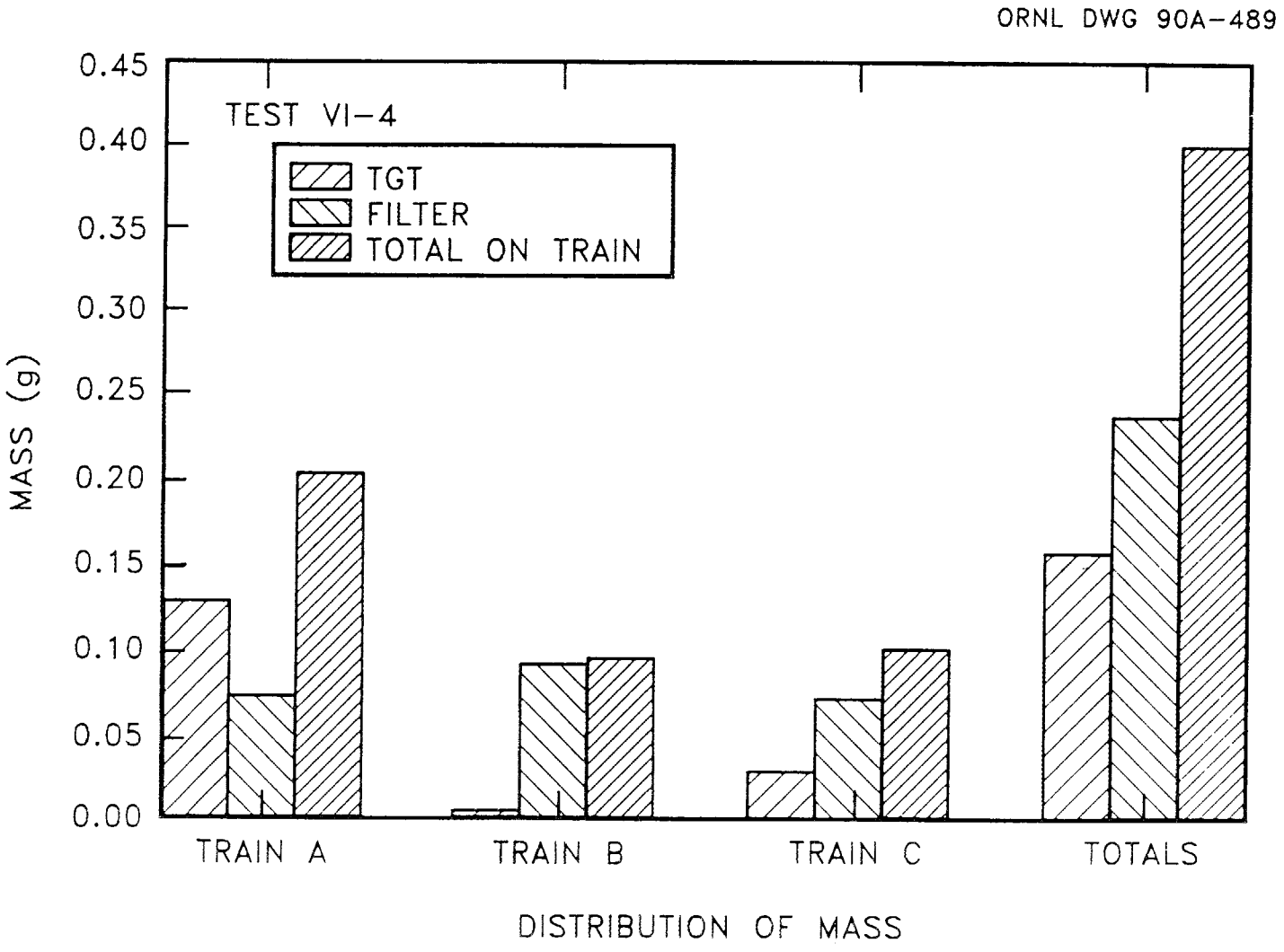

Fig. 17. Distribution of mass deposits in collection trains of test VI-4.

4.2.8 Fuel Examination

As noted previously, the fuel specimen was cast in epoxy resin to preserve the test geometry before removal from the furnace. Following detailed gamma spectrometry, the entire specimen was shipped to Argonne National Laboratory, where it was cut with an abrasive saw for examination of the fuel cross sections.* Using the bottom end of the ZrO_2 furnace tube as a reference point, transverse cuts were made at seven elevations: at 7.3, 9.5, 9.8, 10.8, 11.4, 12.1, and 14.6 cm. In addition, the section from 7.3 to 9.5 cm was cut axially, approximately along the centerline. Each section was ground smooth, inspected, and photographed. Also, the axial section was successively ground, polished, and photographed six times, providing a series of views of the same sample at various radial locations.

Examination of the radial sections confirmed that no fuel remained in the region where the upper end of the rod was originally located. That is, significant collapse or subsidence of the fuel had occurred, as indicated by the radioactivity measurements during the test (Sect. 3.2) and posttest gamma scanning of the fuel (Sect. 4.2.1). Macroscopic features such as the regions of fuel collapse and deposits of molten cladding were well defined. Agreement between fuel concentrations and the gamma scans was generally good, indicating the retention of gamma-emitting fission products in the degraded fuel.

The axial section from the lower end of the fuel region, ~7.3 to ~9.5 cm, was photographed after each of six successive grind and polish operations. These views, shown in Fig. 18, reveal much void space in the bottom end of the fuel region. The UO_2 was present in pieces of various sizes, usually associated with some cladding material, with many regions of mixed (U,Zr)O where fuel-cladding interactions had occurred. Also, numerous areas of interaction of the fuel-cladding melt with the ZrO_2 furnace tube were apparent.

Several chunks of UO_2 surrounded by previously molten cladding were examined and found to exhibit extensive porosity in the metallic phase. One such region is shown in Fig. 19. This porosity was concluded to have been caused by gases (Kr, Xe, and/or the vapors of the more volatile fission products, such as Cs, Rb, I, Te, and Sb) escaping from the UO_2 at high temperature and being trapped in the molten fuel-cladding layer. Similar regions of highly porous material were examined in the fuel specimens from test ST-1.

Also, several areas in the region of fuel-cladding interaction were examined by electron microprobe. The original 1.5% tin content in the Zircaloy had been reduced significantly, as would be expected from the observed release of tin (Table 10, Sect. 4.2.4). A radial section

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*Y. Y. Liu, Argonne National Laboratory, private communication, 1989.

ORNL-PHOTO 3595-90

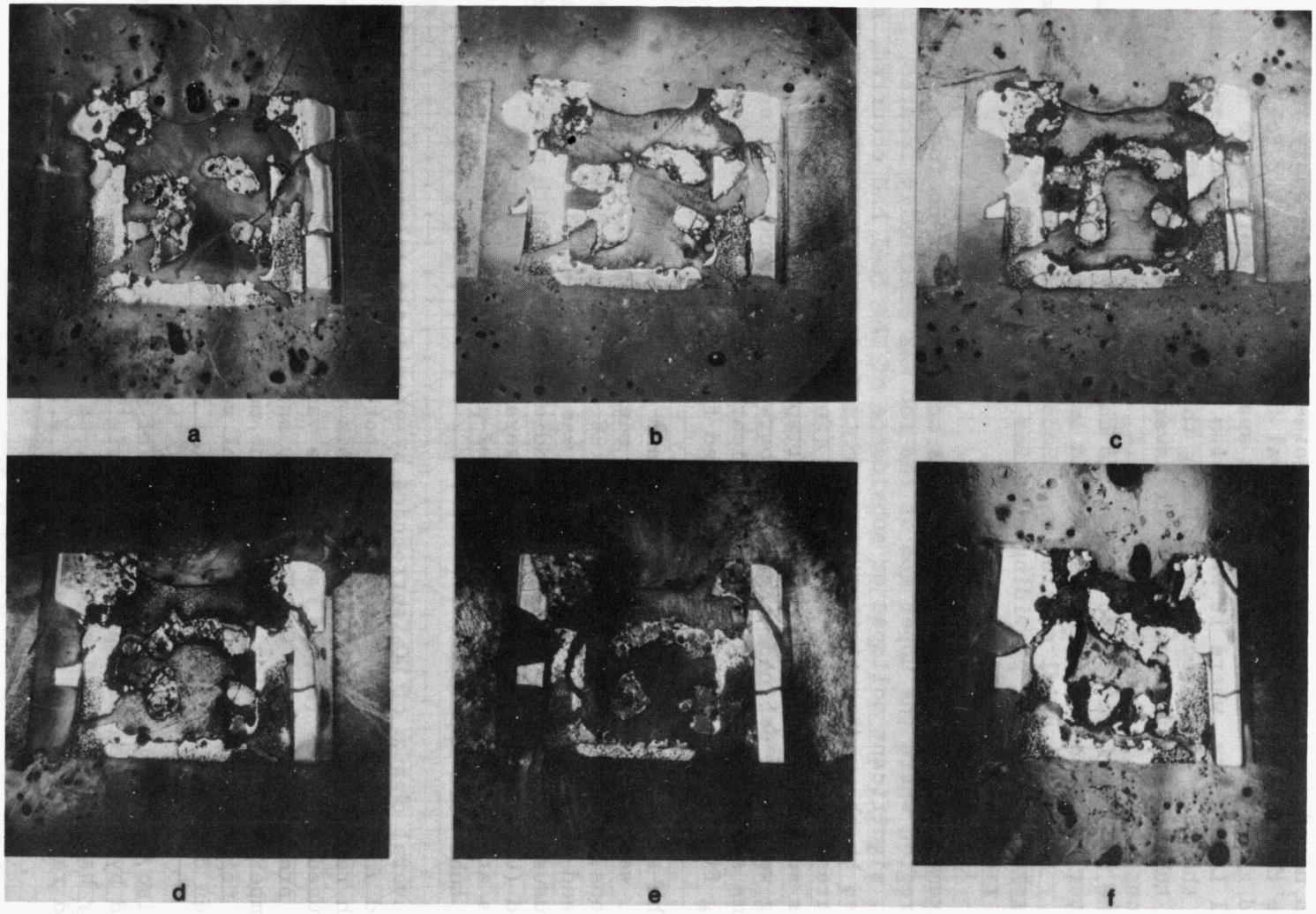

Fig. 18. Longitudinal section from lower end of fuel region in test VI-4, showing appearance after six successive grind-and-polish operations, with (a) after first grind-and polish, progressing to (f) after sixth.

ORNL-PHOTO 7933-90

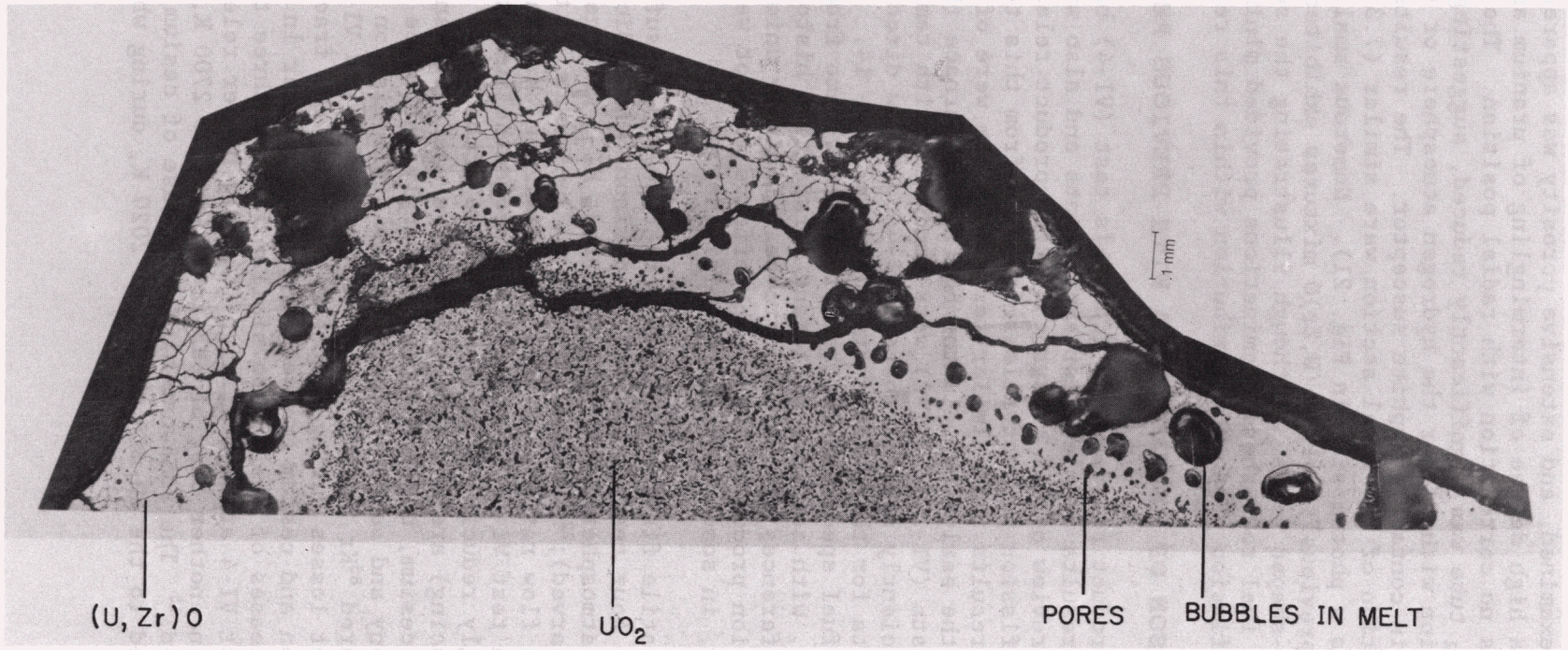

Fig. 19. Region of UO_2 fuel surrounded by molten (U,Zr)O, showing extensive and growing porosity in the molten phase (250×).

at the 9.5 cm elevation is shown in photomosiac form in Fig. 20. As indicated, areas from near the center, mid-radius, and near the ZrO_2 furnace tube were examined, and extensive porosity was apparent. The microprobe showed a high degree of intermingling of uranium and zirconium, but there was no correlation with radial position. The outer surface of the ZrO_2 tube was significantly reduced, suggesting some loss of oxygen by reaction with either the hydrogen atmosphere or the adjacent (but not in contact) graphite susceptor. The results of a microprobe examination of the axial section were similar (7.3 to 9.5 cm elevation, shown as a photomosiac in Fig. 21). Numerous small chunks of UO_2 surrounded by previously-molten (U,Zr)O mixtures exhibited very large, irregularly-shaped voids. Although illustrating the severe degradation of the fuel rod, these examinations provided minimal useful information about fission product distribution within this region.

5. COMPARISON OF RELEASE DATA WITH PREVIOUS RESULTS

The fission product release data from this test (VI-4) have been compared with the results from earlier experiments and also with a comprehensive NRC review of all relevant fission product release data.[21] Comparison of the fission product release results from this test in hydrogen with the results from similar tests in steam were of particular interest. One of the early tests in the horizontal furnace (HI-3) and two more recent tests (VI-1 and VI-2) were conducted with temperature/time regimes sufficiently similar to test VI-4 to allow direct comparison. Important data for these tests are shown in Table 14. It should be noted that the fuel specimens in these four tests came from three different reactors, with somewhat different irradiation histories. Although these differences are believed to be relatively unimportant influences on fission product release, the effects are not well understood and may, in some cases, be significant.

Only the volatile fission products were released in sufficient fractions in these four tests to permit comparison. As indicated by the steam flow rates, atmospheric conditions in test HI-3 were generally reducing (steam-starved);[22] whereas conditions, by virtue of the five times higher steam flow rate, were strongly oxidizing throughout tests VI-1 and VI-2. In test VI-4, however, the hydrogen-helium atmosphere resulted in strongly reducing conditions. These atmospheric effects (oxidizing vs reducing) are not expected to have a significant effect on either krypton or cesium, but they should strongly influence the behavior of antimony and several other less-volatile fission products. Although the measured ^{85}Kr release data for tests VI-1 and VI-2 are low because of apparent losses during posttest transfer, the fractional releases of krypton and cesium are believed to be similar in each test. The fractional releases of cesium are similar for the three tests at 2300 K, but in test VI-4 at 2400 K, significantly higher release was observed. Also, in another test of BR3 fuel, VI-3 at 2700 K, 99% of the cesium was released.[13] The somewhat higher release of cesium in test VI-1 may be related to the 20-min period at 2020 K, during which 39% was released.

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Fig. 20. Radial section at 9.5 cm elevation, showing extensive porosity and fracturing. Locations examined by electron microprobe are identified.

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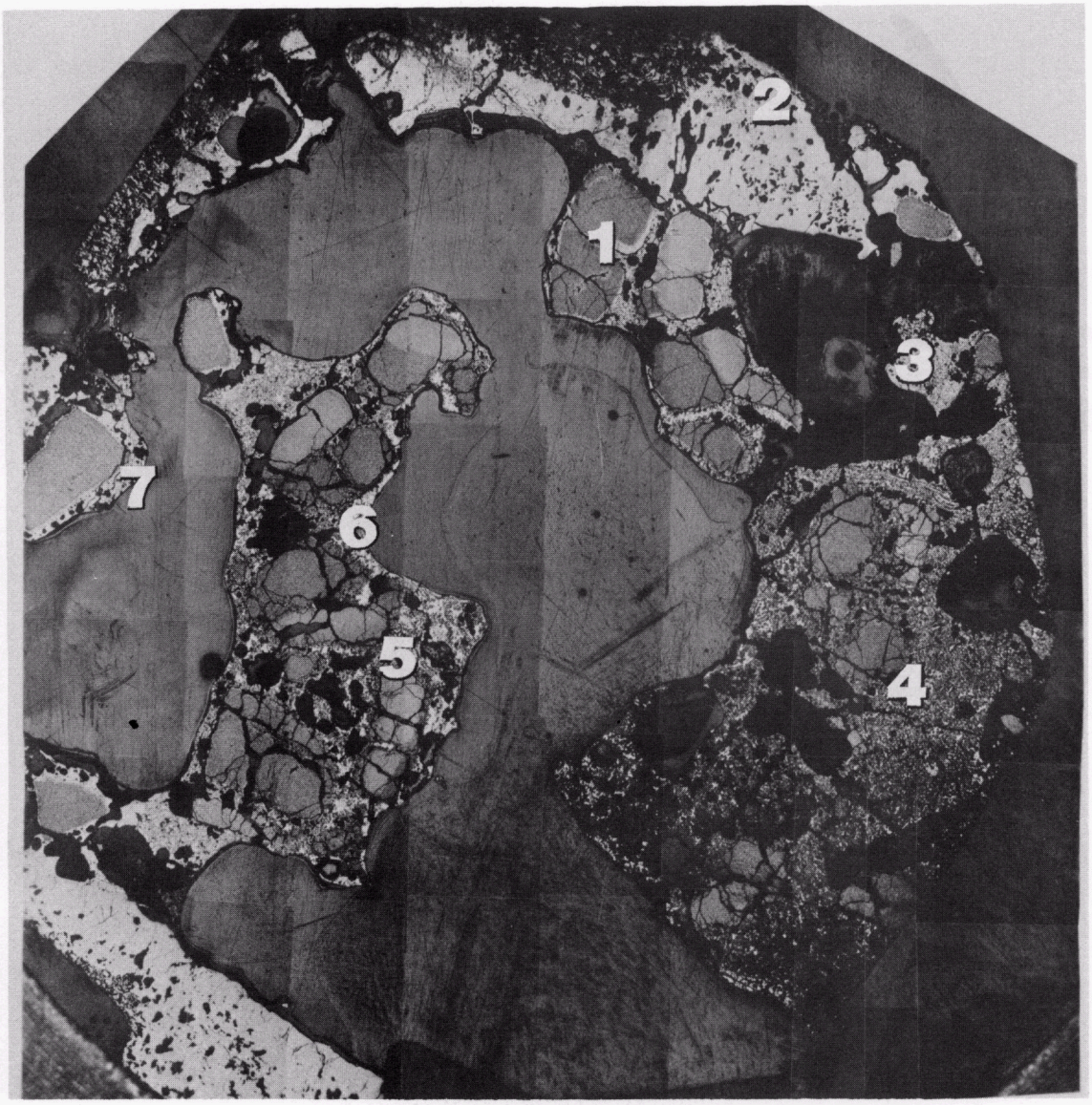

Fig. 21. Axial section from 7.3 to 9.5 cm elevation, showing voids. Locations examined by electron microprobe are identified.

Table 14. Comparison of release data from tests at similar conditions

Test No. (phase)	HI-3	VI-1 (A+B+C)	VI-2 (A+B)	VI-2 (A+B+C)	VI-4 (A+B+C)
Fuel characteristics					
Reactor	HBR 2	Oconee 1	BR3		BR3
Burnup (MWd/kg)	25.2	40	44		47
Maximum heat rating (W/cm)	326		251		270
Test conditions					
Fuel temperature (K)	2275	2300	2300	2300	2400
Effective time (min)[a]	21	30	25	72	31
Test atmosphere (fraction)	0.7 H_2O +0.3 He	0.8 H_2O +0.2 He	0.8 H_2O +0.2 He	0.8 H_2O +0.2 He	0.5 H_2 +0.5 He
Gas flow rate (L/min)	0.3 H_2O 0.15 He	1.5 H_2O 0.4 He	1.5 H_2O 0.3 He	1.5 H_2O 0.3 He	0.4 H_2 0.4 H_2
Fission product release (%)					
^{85}Kr	59	54[b]	31[b]	31[b]	85
^{125}Sb	0.001	33	49	68	3.9
^{137}Cs	59	74	54	63	95

[a]Includes contribution for heatup and cooldown periods.
[b]Parts of Kr released during tests VI-1 and VI-2 were lost during posttest transfer; release fractions for Kr are believed to be similar to those for Cs.

Unlike the data for Kr and Cs, the Sb release varied widely for the four tests compared (Table 14). The retention of antimony by metallic Zircaloy is believed to be the dominant effect. Although Sb (like Te) appears to be released from the UO_2 fuel at rates similar to those for Kr and Cs, our experiments have shown that both Sb and Te are retained as long as the Zircaloy cladding remains metallic. Because test HI-3 was steam-starved (see Table 13), the cladding was only partially oxidized; hence the antimony was retained almost entirely. In tests VI-1 and VI-2, however, the excess steam resulted in complete oxidation of the cladding, causing the high release of antimony late in the test. In the reducing atmosphere of test VI-4, most of the antimony released from the fuel was retained in the melted and rundown cladding. As shown in Table 9, most of the small fraction of antimony released from the cladding was deposited in the furnace, probably as the elemental vapor. The releases of cesium and antimony in all four VI tests are illustrated graphically in Fig. 22. (It should be recalled that tests VI-1, VI-2, and VI-3 were conducted in steam, and test VI-4 in hydrogen.) As shown, although large fractions of cesium were found in Train A of all tests, no antimony was found in Train A in any test. The antimony release in the first three tests was delayed until the cladding had become oxidized by the steam, but in Test VI-4, the hydrogen atmosphere prevented any significant oxidation; hence, there was very little antimony released at all.

Total cesium release data during the heatup phase to ~2300 K for three ORNL tests and SNL test ST-1 are compared in Fig. 23. Although the final release values (including at least ~20 min at test temperature) for these four tests are not drastically different (in the range 60 to 95%), it is apparent that the release occurred much earlier in some tests than in others. Test atmosphere may be a factor in these differences; HI-3 was steam-starved, VI-2 had excess steam, and VI-4 and ST-1 were in hydrogen. Fuel characteristics are another possible explanation for the different cesium behavior; fuel from the H. B. Robinson 2 reactor was heated in Test HI-3, whereas BR3 fuel was heated in the other three tests. In any event, the tests of most similar fuel and conditions, VI-4 and ST-1, both (1) of high enrichment BR3 fuel of the same heat rating and (2) tested in hydrogen, showed the most similar cesium release behavior. This suggests that any influence of the mode of testing, fission heating in-pile for ST-1 vs radiant heating in a hot cell for VI-4, was secondary to the above experimental variables.

In Fig. 24, the release rate coefficients for krypton and cesium, as observed on-line in test VI-4, are compared with the coefficient as calculated by CORSOR-M as a function of test time;[23] the test temperature is also shown. The erratically higher release rate coefficients for krypton and cesium at very low temperatures, as have been observed previously, are attributed to poor counting statistics and to fission products previously released but adsorbed or deposited on the fuel surfaces.* During the ~1620 K period, the krypton and cesium coefficients tended somewhat above and below the CORSOR-M value, respectively, but followed it quite closely during the heatup from 1620 to

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*T. Nakamura, private communication, ORNL, 1988.

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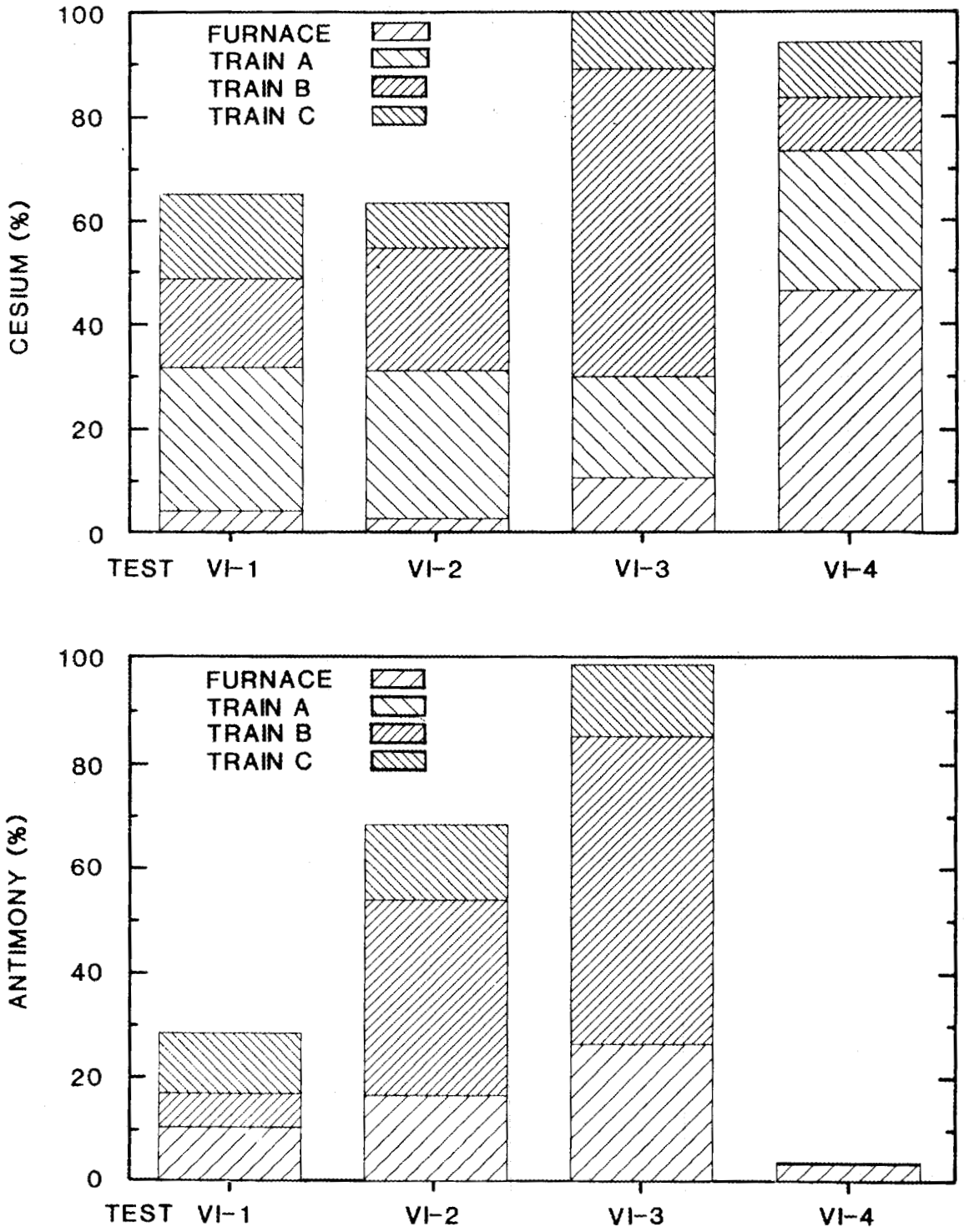

Fig. 22. Comparison of the distributions of cesium and antimony in the furnaces and collection systems of four tests; tests VI-1, VI-2, and VI-3 were conducted in steam and test VI-4 was conducted in hydrogen.

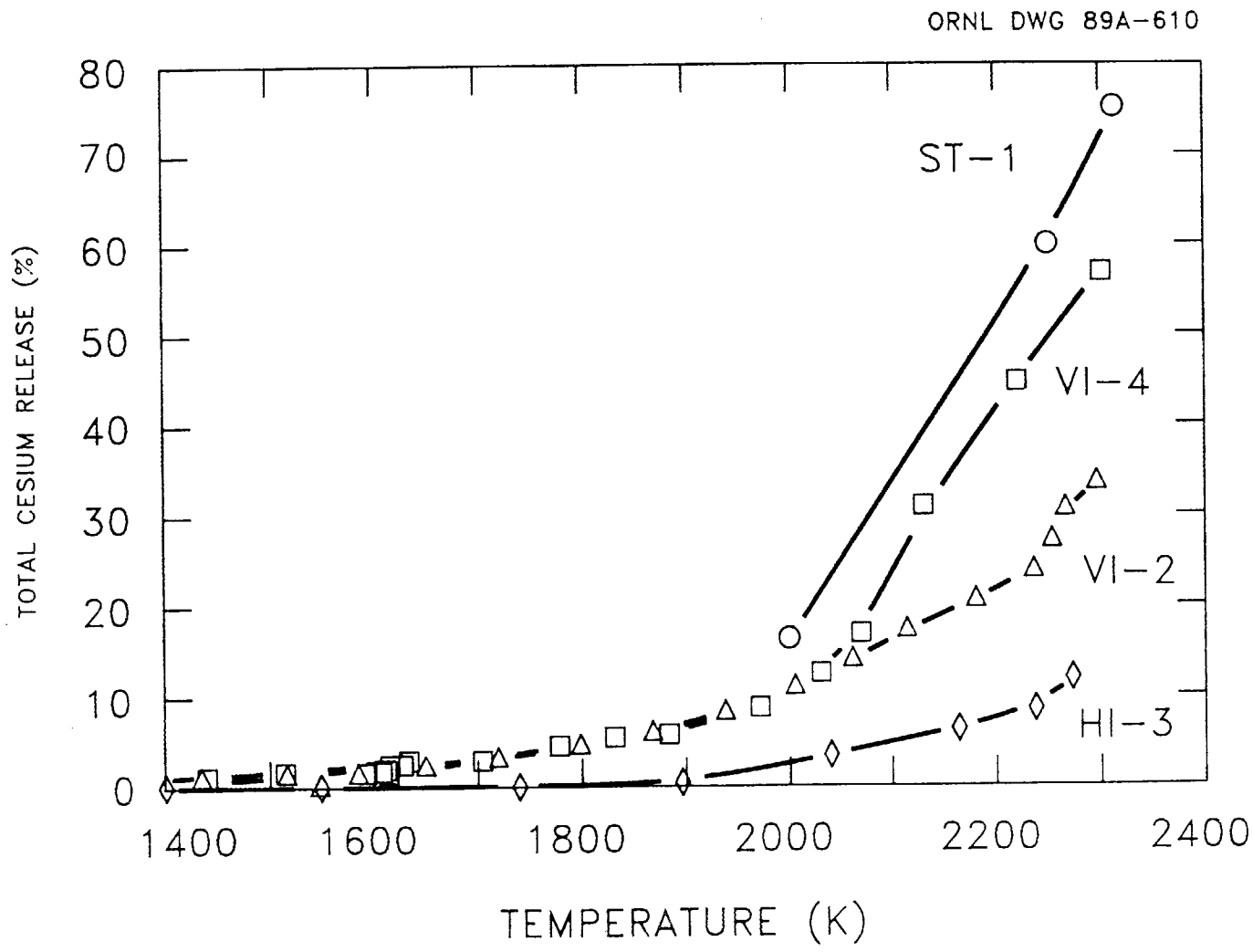

Fig. 23. Comparison of cumulative cesium release during heatup to ~2300 K in four tests, showing widely varying values.

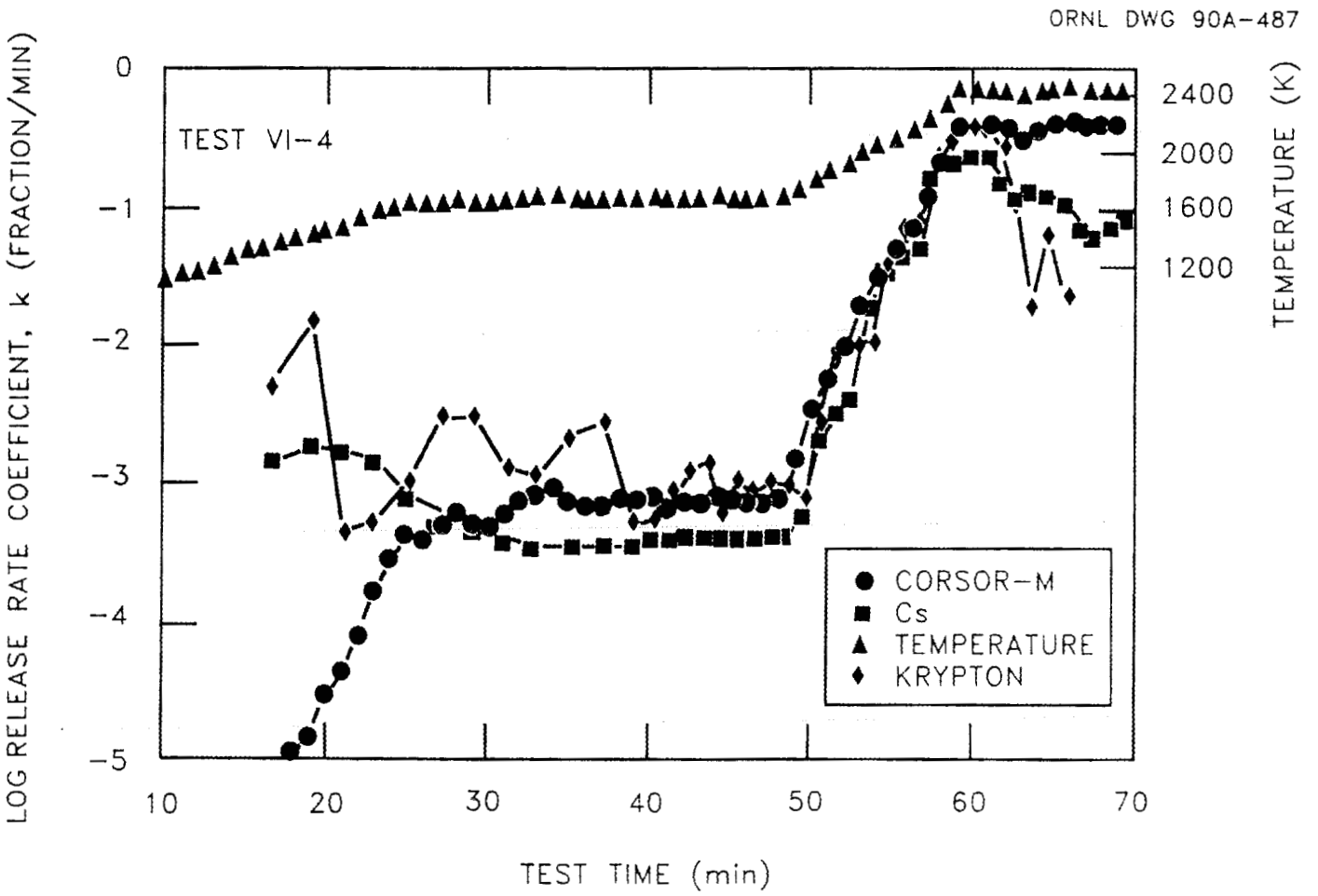

Fig. 24. Release rate coefficients for krypton and cesium in test VI-4, as compared with the coefficient calculated by CORSOR-M for the same temperature history.

2400 K. Shortly after reaching the test temperature of 2400 K, however, both measured coefficients began to fall significantly below the CORSOR-M curve. Such a decline in release rate with time (fraction released) has been observed and discussed previously.[12]

Diffusion coefficients for cesium release were calculated from the on-line release data and are compared with the ANS 5.4 Standard,[24] which is based on essentially the same data as the CORSOR-M model. As shown in Fig. 25, cesium behavior in test VI-4 was generally quite consistent with the ANS 5.4 Standard. The increase in the diffusion coefficient at ~2100 K appears to be the result of the fuel relocation (Sect. 4.1), in which a significant fraction of the fuel suddenly relocated to a higher temperature region, with a corresponding sudden increase in cesium release from that fuel.

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-4 is impossible 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 test equipment operated well and the specially-developed temperature gradient within the furnace was achieved. Although partial collapse of the fuel occurred, the test was successful, and valuable release data were obtained.

2. The total release values for the more volatile fission products – Kr, Sb, and Cs – were determined for the specific conditions of this test. The release values were ~85% for Kr, 95% for Cs, and 3.9% for Sb. Comparison of these data from test VI-4 with the results from previous tests at similar temperatures showed similar values for Kr and Cs, but drastically different release behavior for Sb, which appears to be released from the fuel but retained in the metallic Zircaloy cladding.

3. The high test temperature (2400 K) in hydrogen resulted in higher release of europium (13.4%) than in any previous test. Most of this europium was deposited on the furnace tube immediately above the fuel, apparently by vapor condensation. In addition, more Sr and Ba, and less Sb and Ru, were released in the this test in hydrogen than would be expected at the same temperature in steam.

4. Measurements of the masses of deposits collected on the TGTs and the filters showed that much less mass release occurred in the hydrogen atmosphere of this test than in the steam atmospheres of previous similar tests. In addition, the ratio of aerosol to vapor was much less in this test, suggesting that the different atmosphere had a strong reduction on the amount of oxide produced as aerosol, with less effect on vaporization of the elements.

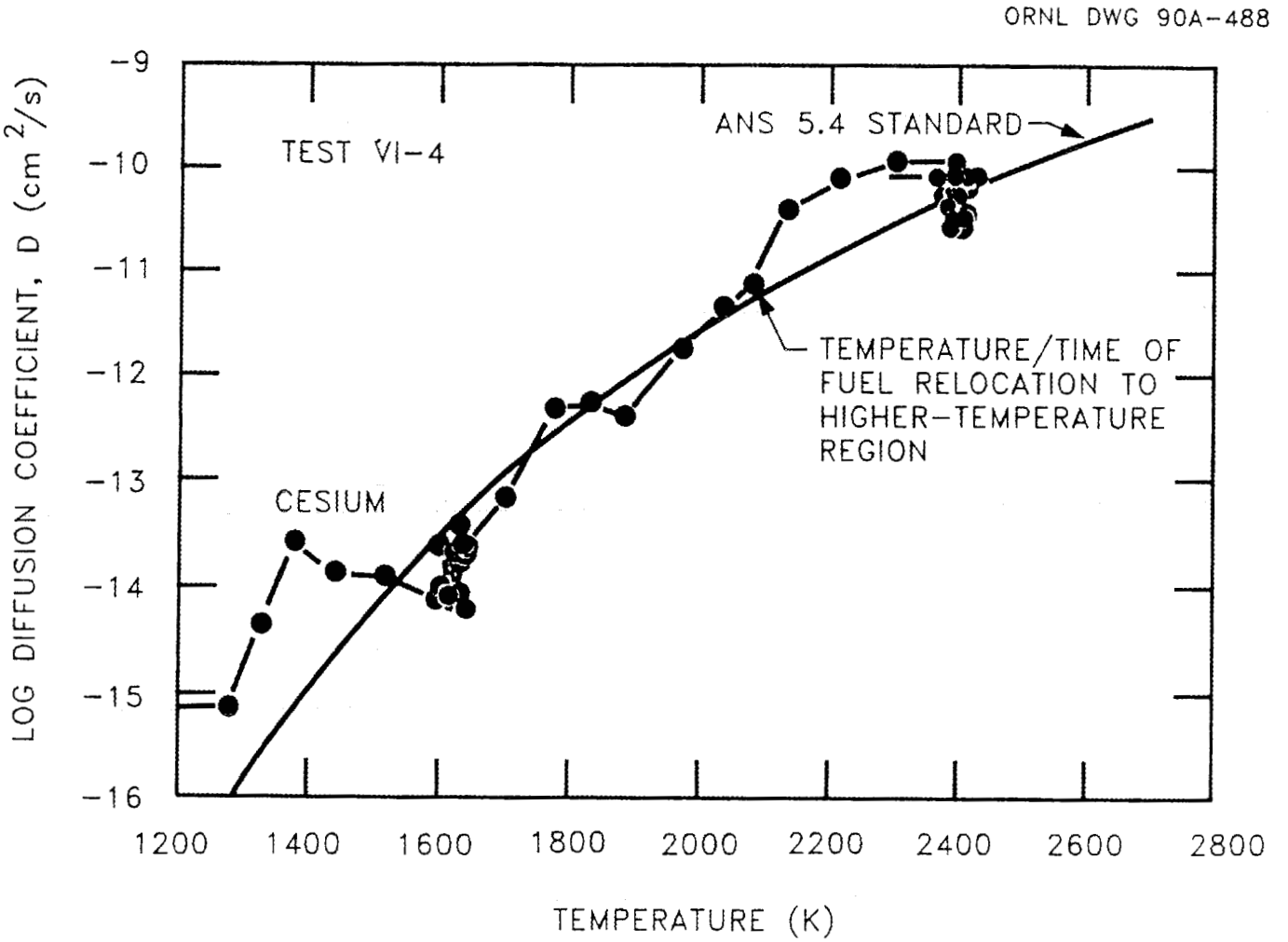

Fig. 25. Diffusion coefficients from cesium in test VI-4, compared with the ANS 5.4 standard curve.

5. Although all fission product release data are not yet available, the available results indicate good agreement, for the release of fission products, between this test (VI-4) and the principal objective for comparison, SNL Test ST-1. There was no indication that the mode of testing – fission-heated in-pile vs radiant-heated in a hot cell – caused a significant influence on fission product release. This finding tends to verify the applicability of results from previous ORNL hot cell tests for use in reactor accident analyses.

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13. ABSTRACT (200 words or less)

Test VI-4, the fourth in a series of high-temperature fission product release tests in the vertical test apparatus, was conducted in a flowing hydrogen-helium atmosphere. The test specimen was a 15.2-cm-long section of a fuel rod from the BR3 reactor in Belgium, which had been irradiated to a burnup of 47 MWd/kg. Using an induction furnace, it was heated under simulated LWR accident conditions to a test temperature of 2400 K for 20 min.

Radioactivity measurements showed that the fuel collapsed after the Zircaloy cladding melted during heatup. The total release of fission products from the fuel was 85% for ^{85}Kr, <1% for ^{103}Ru, 3.9% for ^{125}Sb, 96% for both ^{134}Cs and ^{137}Cs, and 13% for ^{154}Eu. Most of the Eu was retained in the furnace, but most of the other elements were released to the collection system. Small fractions of other fission products (Sr, Te, and Ba), as well as fuel (U and Pu) were released also. The total mass release from the 100 g fuel specimen was 0.40 g, with 40% of this as vapor and 60% as aerosol.

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