



NUREG/CR-3715  
ORNL/TM-9135

OAK RIDGE  
NATIONAL  
LABORATORY

MARTIN MARIETTA

**Design, Construction, and Testing  
of a 2000°C Furnace and Fission  
Product Collection System**

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

Prepared for the  
U.S. Nuclear Regulatory Commission  
Office of Nuclear Regulatory Research  
Under Interagency Agreement DOE 40-551-75

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

8411290160 841031  
PDR NUREG  
CR-3715 R PDR

#### NOTICE

##### Availability of Reference Materials Cited in NRC Publications

Most documents cited in NRC publications will be available from one of the following sources:

1. The NRC Public Document Room, 1717 H Street, N.W., Washington, DC 20555
2. The NRC/GPO Sales Program, U.S. Nuclear Regulatory Commission, Washington, DC 20555
3. The National Technical Information Service, Springfield, VA 22161

Although the listing that follows represents the majority of documents cited in NRC publications, it is not intended to be exhaustive.

Referenced documents available for inspection and copying for a fee from the NRC Public Document Room include NRC correspondence and internal NRC memoranda; NRC Office of Inspection and Enforcement bulletins, circulars, information notices, inspection and investigation notices; Licensee Event Reports; vendor reports and correspondence; Commission papers; and applicant and licensee documents and correspondence.

The following documents in the NUREG series are available for purchase from the NRC/GPO Sales Program: formal NRC staff and contractor reports, NRC-sponsored conference proceedings, and NRC booklets and brochures. Also available are Regulatory Guides, NRC regulations in the *Code of Federal Regulations*, and *Nuclear Regulatory Commission Issuances*.

Documents available from the National Technical Information Service include NUREG series reports and technical reports prepared by other federal agencies and reports prepared by the Atomic Energy Commission, forerunner agency to the Nuclear Regulatory Commission.

Documents available from public and special technical libraries include all open literature items, such as books, journal and periodical articles, and transactions. *Federal Register* notices, federal and state legislation, and congressional reports can usually be obtained from these libraries.

Documents such as theses, dissertations, foreign reports and translations, and non-NRC conference proceedings are available for purchase from the organization sponsoring the publication cited.

Single copies of NRC draft reports are available free, to the extent of supply, upon written request to the Division of Technical Information and Document Control, U.S. Nuclear Regulatory Commission, Washington, DC 20555.

Copies of industry codes and standards used in a substantive manner in the NRC regulatory process are maintained at the NRC Library, 7920 Norfolk Avenue, Bethesda, Maryland, and are available there for reference use by the public. Codes and standards are usually copyrighted and may be purchased from the originating organization or, if they are American National Standards, from the American National Standards Institute, 1430 Broadway, New York, NY 10018.

#### Notice

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

NUREG/CR-3715  
ORNL/TM-9135  
Dist. Category R3

Chemical Technology Division

DESIGN, CONSTRUCTION, AND TESTING OF A 2000°C FURNACE AND  
FISSION PRODUCT COLLECTION SYSTEM

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

Manuscript Completed — March 1984  
Date of Issue — September 1984

**NOTICE** This document contains information of a preliminary nature.  
It is subject to revision or correction and therefore does not represent a  
final report.

Prepared for the  
U.S. Nuclear Regulatory Commission  
Office of Nuclear Regulatory Research  
Washington, DC 20555  
under Interagency Agreement DOE 40-551-75

NRC FIN No. B0127

Prepared by the  
OAK RIDGE NATIONAL LABORATORY  
Oak Ridge, Tennessee 37831  
operated by  
MARTIN MARIETTA ENERGY SYSTEMS, INC.  
for the  
U.S. DEPARTMENT OF ENERGY  
under Contract No. DE-AC05-84OR21400

## ABSTRACT

In order to conduct fission product release tests on commercial LWR fuel specimens, an induction furnace was developed, capable of operation at 2000°C in steam. The test specimen and steam atmosphere are contained in a stabilized ZrO<sub>2</sub> furnace tube, which is heated by a concentric susceptor of either tungsten or graphite. A two-color optical pyrometer and high-temperature thermocouples are used for temperature measurement. The furnace has operated reliably for periods up to 30 min at test temperatures of 1400 to 2000°C with steam flowing at ~1 L/min.

The apparatus for collecting the released fission products includes a thermal gradient tube (TGT), an aerosol deposition sampler, a series of glass fiber filters, and heated charcoal. A steam condenser and cooled charcoal, for inert gas adsorption, are located further downstream. The thermal gradient tube, which varies from ~850°C at the inlet to 150°C at the outlet, is composed of either platinum or stainless steel.

The principal analytical techniques used for fission product identification and measurement are (1) gamma spectrometry, for all radionuclides on all test components; (2) spark-source mass spectrometry, for all elements, primarily deposits on the thermal gradient tube and filters; and (3) neutron activation, for iodine on selected test components. The apparatus and techniques have been shown to be effective in conducting the tests and in collecting an abundance of data on fission product release and behavior in this apparatus.



## CONTENTS

	<u>Page</u>
ABSTRACT .....	111
LIST OF FIGURES .....	vii
LIST OF TABLES .....	ix
1. EXECUTIVE SUMMARY .....	1
2. INTRODUCTION .....	2
3. APPARATUS DESIGN REQUIREMENTS AND CONSTRUCTION .....	3
3.1 FURNACE .....	3
3.2 FISSION PRODUCT COLLECTION SYSTEM .....	3
3.3 HOT CELL INSTALLATION .....	5
3.4 INSTRUMENTATION .....	12
3.4.1 Temperature Measurement .....	12
3.4.2 Gas Flow Rate and Pressure Measurement .....	12
3.4.3 Radiation Measurement .....	17
3.4.4 Chemical Analysis .....	17
3.4.5 Aerosol Sampling and Analysis .....	20
4. PRELIMINARY TESTING AND CALIBRATION .....	20
4.1 FURNACE TESTING .....	20
4.2 THERMAL GRADIENT TUBE TESTING .....	24
4.3 INERT GAS AND STEAM FLOW TESTING .....	27
4.4 RADIATION MEASUREMENT TESTING .....	27
5. CONTROL TESTS .....	27
5.1 TESTS C-1 AND C-2 .....	28
5.2 TEST C-3 .....	28
5.3 TEST C-4 .....	33
5.4 TEST C-6 .....	38
5.5 TESTS C-7 AND C-8 .....	45
6. REFERENCES .....	45
APPENDIX. COMMERCIALY SUPPLIED EQUIPMENT AND INSTRUMENTATION ....	49

LIST OF FIGURES

<u>Number</u>		<u>Page</u>
1	Fission product release furnace .....	4
2	Fission product collection system .....	7
3	Fission product release and collection system .....	8
4	Photograph of (a) fission product release furnace, (b) thermal gradient tube, and (c) filter package in steel containment box before test HI-3 .....	9
5	Hot cell installation with containment box closed .....	10
6	Condenser, dryer, and two cold charcoal traps mounted around NaI(Tl) detector for $^{85}\text{Kr}$ measurement .....	11
7	Data acquisition and processing system for fission product release test .....	13
8	Temperature instrumentation for fission product release apparatus .....	14
9	Steam generator and flow measurement/control instrumentation mounted outside hot cell .....	15
10	Simplified flow diagram for fission product release test apparatus .....	16
11	Multichannel analyzer (MCA) used to measure quantities of radionuclides on test components .....	18
12	Arrangement of apparatus for laser-induced fluorescence (LIF) spectrometry .....	19
13	Schematic diagram of miniature aerosol deposition sampler .....	21
14	Miniature aerosol sampler, showing path of gases/aerosol through interior .....	22
15	Temperature profiles along test fuel rod as measured during furnace testing with a 22 turn induction coil ....	23
16	Axial temperature profile indicated by interior platinum vs rhodium thermocouple during furnace testing .....	25
17	Comparison of temperatures indicated by optical pyrometer and thermocouple at inlet end in furnace test .....	26

<u>Number</u>		<u>Page</u>
18	Temperature and power histories of test C-1 .....	31
19	Temperature and power histories of test C-2 .....	32
20	Temperature and power histories of test C-3 .....	34
21	Comparison of the oxidized fuel specimen (Zircaloy-clad UO <sub>2</sub> ) from test C-3 with an untested specimen .....	35
22	Distribution of <sup>130</sup> I along thermal gradient tubes in tests C-3 and C-4 .....	37
23	Temperature and flow histories of test C-4 .....	39
24	Distribution of tin and antimony in test C-4 thermal gradient tube .....	42
25	Temperature and flow histories for test C-6 .....	43
26	Appearance of the fuel specimen from test C-6 .....	44

LIST OF TABLES

<u>Number</u>		<u>Page</u>
1	Analytical techniques for fission product release data .....	6
2	Construction and operating data for control tests .....	29
3	Comparison of temperature data in tests C-1 through C-6 ..	20
4	Distribution of $^{130}\text{I}$ in test C-3 .....	36
5	Distribution of $^{130}\text{I}$ in test C-4 .....	38
6	Distribution of radioactive tracers from irradiated Zircaloy in test C-4 .....	40
7	Distribution of radioactive tracers (from irradiated Zircaloy) in test C-6 .....	46

DESIGN, CONSTRUCTION, AND TESTING OF A 2000°C FURNACE AND  
FISSION PRODUCT COLLECTION SYSTEM

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

1. EXECUTIVE SUMMARY

The development and testing of the furnace and fission product collection apparatus discussed in this report were carried out over a period of about two years; some of the control tests were interspersed with early tests of LWR fuel. Preparation of this report has been delayed to permit the inclusion of as many of the apparatus changes and control tests as possible.

The performance of the high temperature furnace has been generally consistent with the design goals. The required temperature (2000°C) was reached easily, with heating rates  $>2^{\circ}/s$  being achieved. Although no effort was made to determine the maximum possible heating rate, we estimate that rates of 4 to 5 $^{\circ}/s$  are possible, but not necessarily recommended, because of the increased chance of breakage of the ZrO<sub>2</sub> furnace tube, which is the primary containment for the fuel specimen. Temperature measurement within the furnace required significant effort, and because of the high electromagnetic fields and the steam atmosphere (which required a protective ZrO<sub>2</sub> thermowell for the tungsten vs rhenium thermocouples), precision has been less than desired. We assume a precision of  $\pm 50^{\circ}C$  at the inlet end of the furnace, and the axial gradient has been determined to be 50 to 100 $^{\circ}C$ , depending on flow rate and maximum temperature. The ZrO<sub>2</sub> furnace tubes, which are stabilized with 3 wt % MgO, have performed very well. Minor fracturing has occurred in a few instances and a small amount of blistering was evident on the tube that was heated rapidly to 2200 $^{\circ}C$ .

Development of the TGT has included (1) increasing the length, from 30 to 36 cm; (2) doubling the number of heating elements, from 2 to 4, to improve temperature control; (3) increasing the number of thermocouples, from 6 to 12, to improve the temperature resolution; and (4) most recently, substituting a larger, stainless steel liner, in place of platinum, to provide a deposition surface similar to that in reactors. With the exception of one heater failure during a test, this component has operated quite reliably and has provided the desired linear temperature profile.

The filter package, which contains a series of filters and charcoal cartridges, the steam condenser, the dryer, and the cooled charcoal traps, have all performed suitably with only minor modifications. The miniature aerosol sampler has been used in only two tests; it appears to provide an adequate means of collecting very small samples of the aerosol as a function of time during the test, but may require further refinement.

The series of control tests proved the basic operation of the system, and provided needed experience in operation prior to the introduction of dangerous levels of radioactivity. The detailed results of three tests using tracer materials are included. The observed behavior of tracer levels of fission products/chemical species in the control tests under relatively simple conditions has been very helpful in interpreting the behavior of real fission products under the more complex conditions of LWR fuel tests. Those tests, for instance, showed that most (~95%) of the elemental iodine passed through the thermal gradient tube and more than half penetrated the filters to be deposited on the heated charcoal. Conversely, such fission product compounds as CsI and CsOH, which dominate the release from fuel into steam, were collected almost completely on the thermal gradient tube and the filters. In addition, the control tests provided much of our currently available data on the release and behavior of several structural elements, such as Cr, Co, Zr, and Sn.

## 2. INTRODUCTION

Experiments to determine fission product release from light water reactor (LWR) fuel under accident conditions are needed to assess the consequences of severely overheating the fuel. Such accidents might result from a loss of cooling water. In order to conduct tests of irradiated fuel, a highly specialized apparatus capable of remote operation in a hot cell is required.<sup>1</sup> A series of fission product release experiments, in which fuel specimens were heated in the range of 500°C to 1600°C, was conducted at Oak Ridge National Laboratory (ORNL) during 1975-1979; the results of these studies were reported by Lorenz et al.<sup>2-4</sup> In FY 1981 the U. S. Nuclear Regulatory Commission (NRC) requested further fission product release studies to higher temperatures. Because the previous furnace capability was limited to a maximum of ~1600°C, a new design was required. In addition, the importance of collecting more information per test (more fission product species, identification of particular chemical forms, aerosol characteristics, etc.) was recognized. Consequently, a program with the following objectives was initiated.

1. To design, construct, and test a furnace capable of heating a suitable fuel specimen to at least 2000°C in flowing steam. The ultimate objective was fuel melting, at ~2500°C, but a preliminary evaluation of the problem showed that development of a 2000°C furnace, using ZrO<sub>2</sub> ceramics, could be accomplished more quickly and cheaply than development of a 2500°C furnace which would require ThO<sub>2</sub> ceramics. Since a number of tests in the range of 1400°C to 2000°C were desired, we chose to build and test the 2000°C furnace first. Then, utilizing the experience gained from operation of this furnace, we planned to develop the higher temperature apparatus.
2. To improve the fission product collection and analysis techniques used in the previous tests, with particular emphasis on efforts to identify the chemical forms of the fission product species.



3. To improve the on-line data acquisition by the use of newer, improved instrumentation. Although the initial apparatus would include a single collection train and NaI(Tl) detectors as used previously, we planned to utilize multiple collection trains, Ge(Li) detectors, a cascade impactor, and, if available, a sophisticated vapor analyzer (laser Raman or laser-induced fluorescence spectrometer) with the higher temperature furnace.

### 3. APPARATUS DESIGN REQUIREMENTS AND CONSTRUCTION

The design requirements for the test apparatus and instrumentation depended heavily on the test parameters: temperature, atmosphere, and time; other major considerations were the feasibility of remote operation and safety.

#### 3.1 FURNACE

The ultimate objective of the new furnace was the reliable capability to isothermally heat a representative fuel specimen (min 100 g) to complete melting (min 2400°C) in a flowing steam atmosphere and maintain controlled conditions for at least 10 min. Additionally, we required two temperature measurement methods — thermocouples and optical pyrometry — and furnace materials as similar as possible to those found in a reactor core under accident conditions. Application of our experience with furnaces used in other fuel testing programs<sup>4-6</sup> to satisfy these requirements resulted in the design shown in Fig. 1. The principal features of this new furnace are an inductively heated tubular susceptor (both graphite and tungsten have been used successfully), a dense ceramic tube (ZrO<sub>2</sub>) that contains the fuel specimen and the flowing steam-helium atmosphere, temperature measurement at the inlet end using both optical pyrometry and thermocouples, a purified helium purge flowing into the annulus containing the susceptor, thermal insulation of fibrous ZrO<sub>2</sub> surrounding the high temperature zone, and a fused silica vessel that provides gas-tight containment for the assembly. The silica vessel is surrounded by an induction coil of .64-cm (1/4-in.)-diam copper tubing which extends ~1 cm beyond the 25-cm-long susceptor at each end. This furnace is powered by a radio frequency (RF) generator and has operated reliably in fuel tests up to 2000°C, which is near the limit for the ZrO<sub>2</sub> ceramics. This series of tests, utilizing commercial LWR fuel, began in March 1982; they are designated HI-1, HI-2, HI-3, etc. and are being reported separately.<sup>7-9</sup> Upon receipt of the ThO<sub>2</sub> ceramics, which are being fabricated at Los Alamos National Laboratory (LANL),<sup>10</sup> the furnace will be modified and tested at higher temperatures, up to 2400°C.

#### 3.2 FISSION PRODUCT COLLECTION SYSTEM

During the tests, the vapors and aerosols carried out of the furnace by the flowing steam-helium atmosphere are the sources of information on fission product release and behavior, which are the principal objectives of these experiments. Consequently, we sought to collect and analyze this



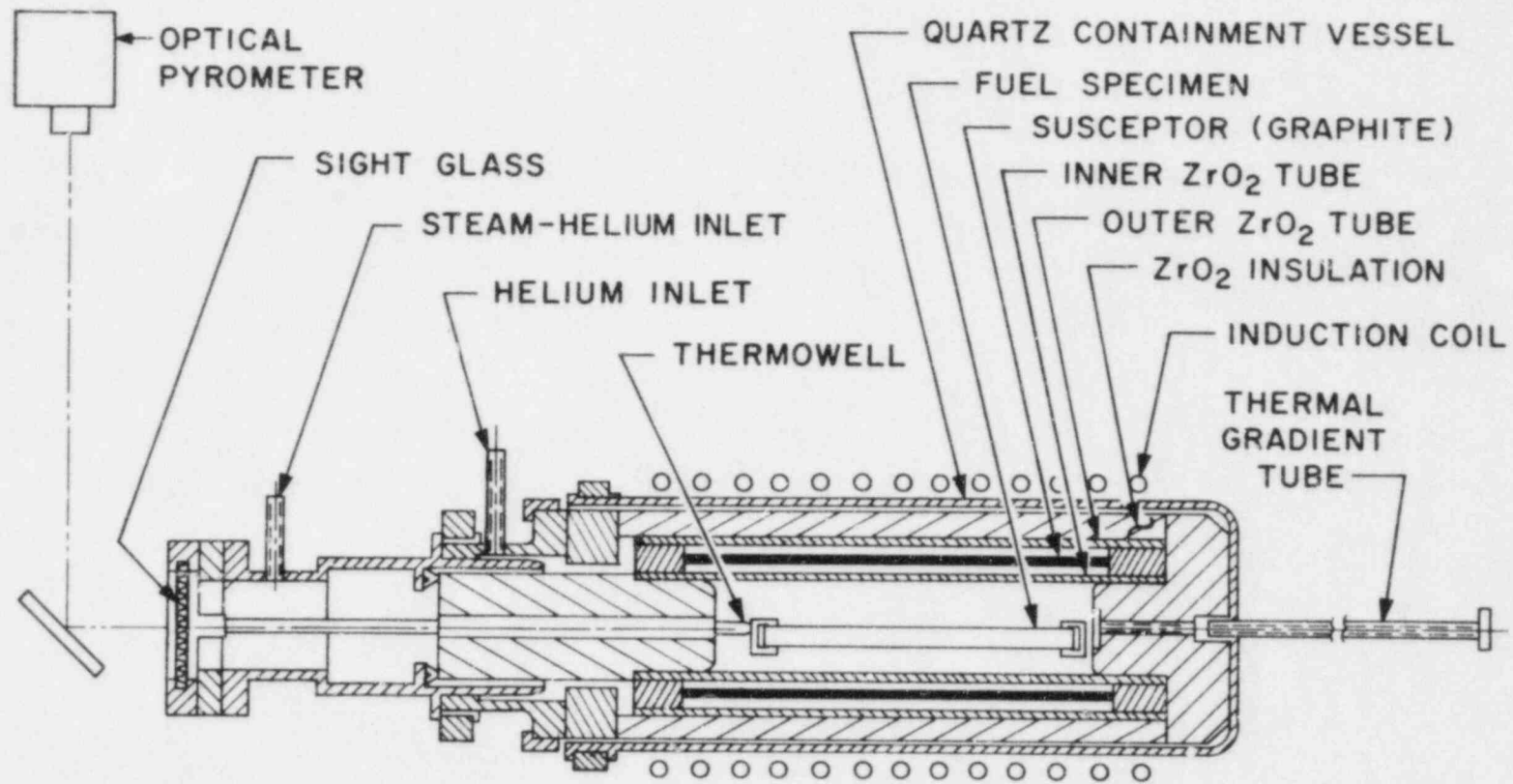


Fig. 1. Fission product release furnace.

material in ways that would provide as much specific information as possible. Techniques employed in previous experimental programs<sup>2,6</sup> constituted the basic tools for this effort; most of our data were obtained through use of the techniques listed in Table 1. The principal components, (as shown in Fig. 2) in succession downstream from the furnace, are (1) a thermal gradient tube providing an inert deposition surface (platinum) at temperatures of  $\sim 850^{\circ}\text{C}$  at the inlet, declining to  $\sim 150^{\circ}\text{C}$  at the outlet; (2) a filter package, maintained at  $\sim 130^{\circ}\text{C}$ , containing, in series, a glass wool prefilter, glass fiber filters of 96% and 99.7% efficiency in collecting  $0.3\text{-}\mu\text{m}$ -diam particles, and four cartridges of triethylenediamine (TEDA)-impregnated charcoal; (3) a condenser and dryer, operating at  $0^{\circ}\text{C}$  and  $-78^{\circ}\text{C}$ , respectively, to collect the water vapor and any other condensible species that might pass through the filter package; and (4) two successive charcoal traps at  $-78^{\circ}\text{C}$  for adsorbing the rare gases Kr and Xe, while allowing the He carrier gas, and any  $\text{H}_2$  formed by steam-Zr reaction, to pass through for measurement and release. In addition to the above collectors, two others have been considered, and may be used in some tests: a stainless steel TGT liner in place of the platinum tube, to provide a deposition surface more similar to that in a reactor system; and a miniature cascade impactor to facilitate particle size separation and analysis. Because of doubts about the applicability of a cascade impactor to our apparatus, a small graphite rod aerosol sampler was designed and has been used in two tests. Along with conversion of the furnace to  $\text{ThO}_2$  ceramics and the capability for  $2400^{\circ}\text{C}$  operation, installation of dual TGT/filter packages is planned for operation during different test periods.

### 3.3 HOT CELL INSTALLATION

Because both the fuel specimens and apparatus components from tests with significant fractional releases are highly radioactive, the furnace and fission product collection system were installed in a hot cell, as shown in Fig. 3. This hot cell is maintained in relatively clean condition (low level radioactive contamination) so that personnel access for apparatus assembly and checkout prior to an experiment is possible. The furnace, TGT, and filter package are all mounted inside a stainless steel containment box  $\sim 1.8\text{ m} \times 0.6\text{ m} \times 0.45\text{ m}$  ( $0.5\text{ m}^3$ ). The box has 3.18-mm-thick walls and is connected to the off-gas system; it operates at a negative pressure of -10-in. water relative to the hot cell, which, in turn, is -1-in. water relative to the surrounding laboratory. This container was designed to contain the debris in the event of severe damage to the furnace, and, in addition, the combination of volume, an air flow rate through the box of 42 L/min, and reduced pressure should prevent positive pressurization if a hypothetical "worst case" hydrogen detonation should occur. The test apparatus is shown, with the box open in Fig. 4 and with the box closed in Fig. 5.

The steam condenser, dryer, cold charcoal traps (two), and a  $\text{NaI}(\text{Tl})$  detector for  $^{85}\text{Kr}$  monitoring are located in a cubicle in the hot cell wall, as illustrated in Fig. 3, and pictured in Fig. 6. The effluent from this part of the collection system - the noncondensable gases helium (or argon) and hydrogen - are returned to the hot cell exhaust system.

Table 1. Analytical techniques for fission product release data

Technique	Time	Location	Elements detected
Gamma spectrometry	Pretest, posttest	Fuel specimen	Long-lived, high energy fission products - Cs, Sb, Ru, Eu, Ce,
	On-line	Thermal gradient tube, filters, gas traps	Cs, Kr
	Posttest	Furnace components, thermal gradient tube, filters	Cs, Ag, Ru, Ce, Eu, (CO)
Activation analysis	Posttest	Charcoal, solution from furnace, thermal gradient tube, filters	I, Br
Spark-source mass spectrometry	Posttest	Samples from furnace, thermal gradient tube, filters	All

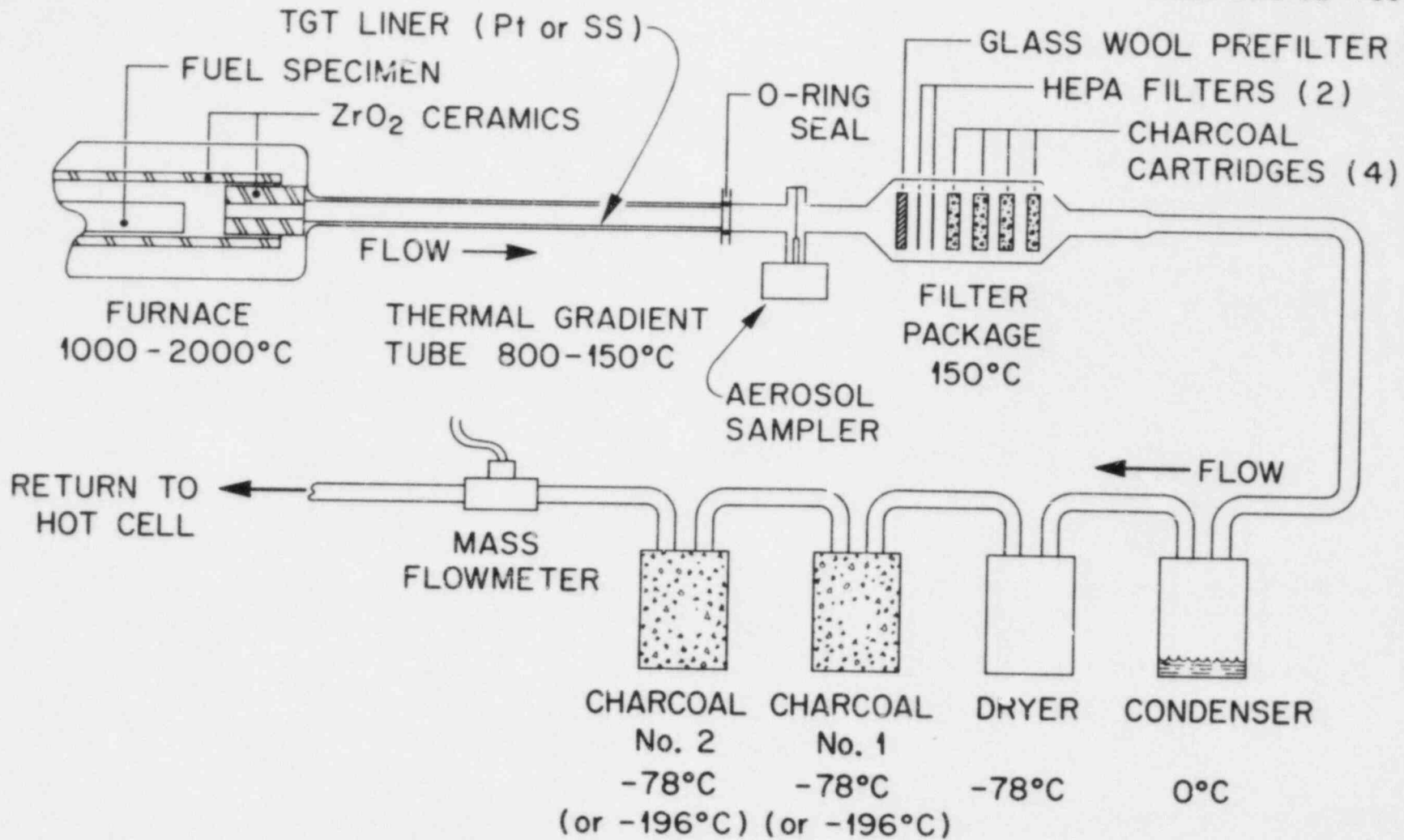


Fig. 2. Fission product collection system.

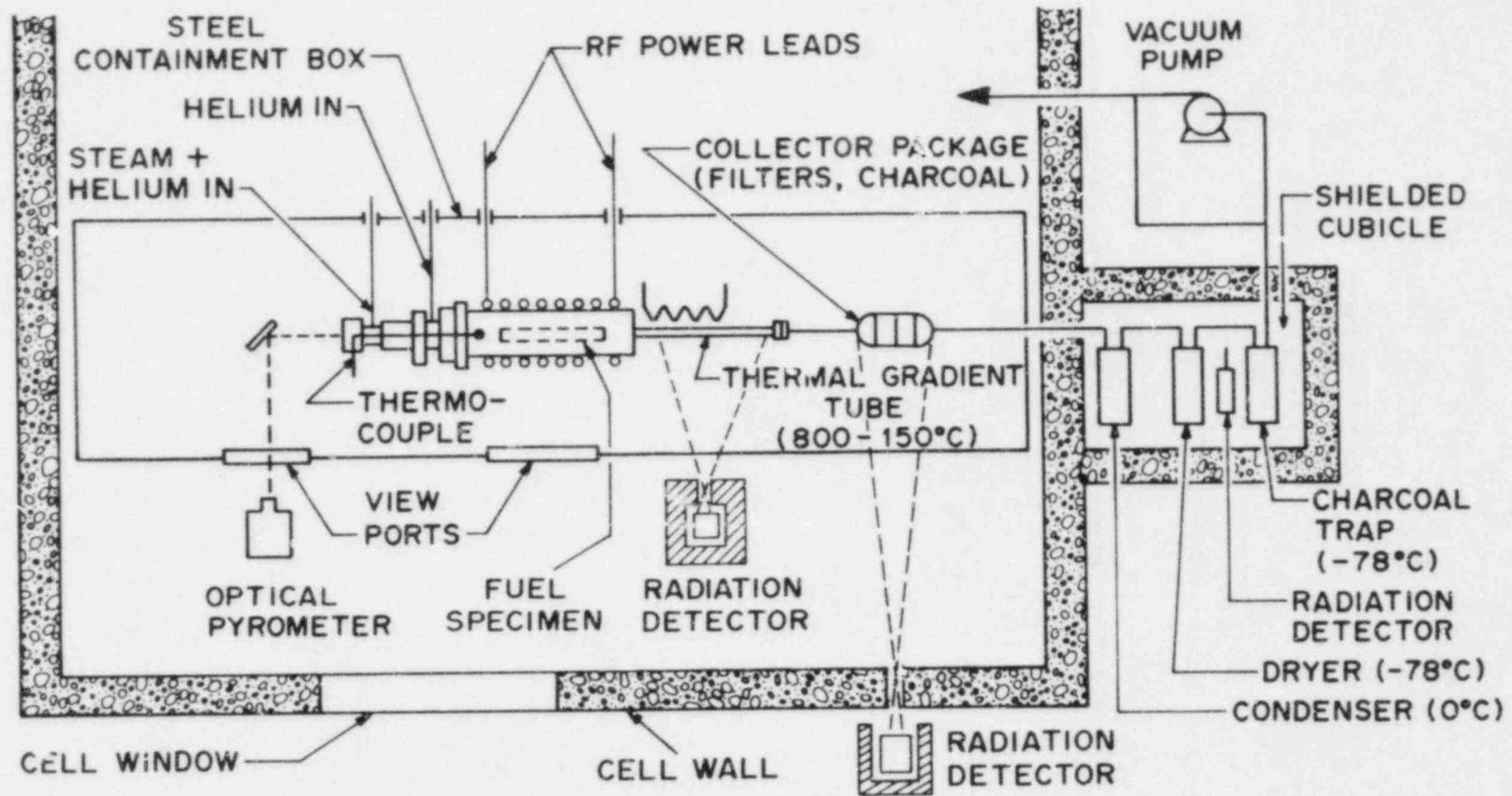


Fig. 3. Fission product release and collection system.





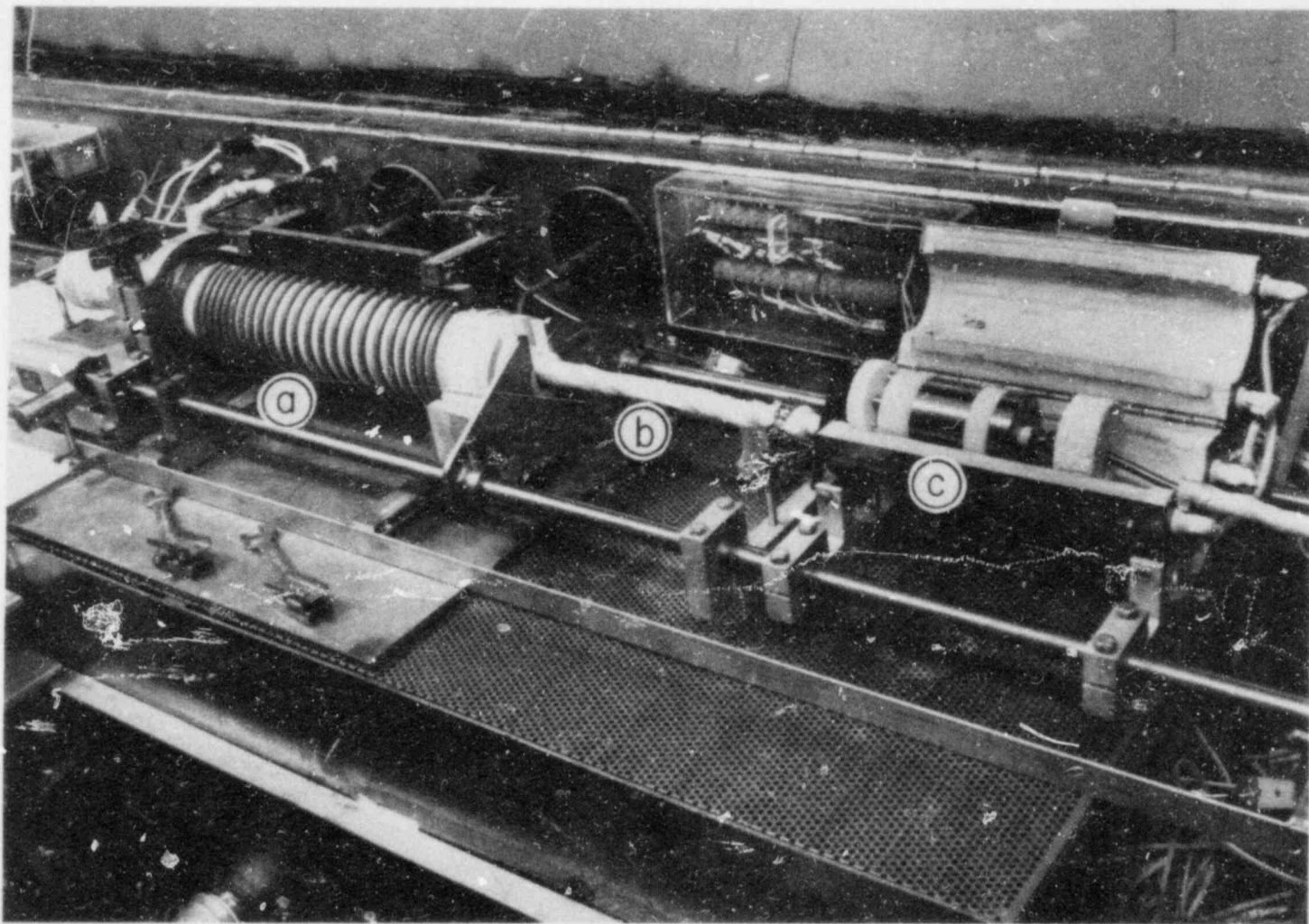


Fig. 4. Photograph of (a) fission product release furnace, (b) thermal gradient tube, and (c) filter package in steel containment box before test HI-3.

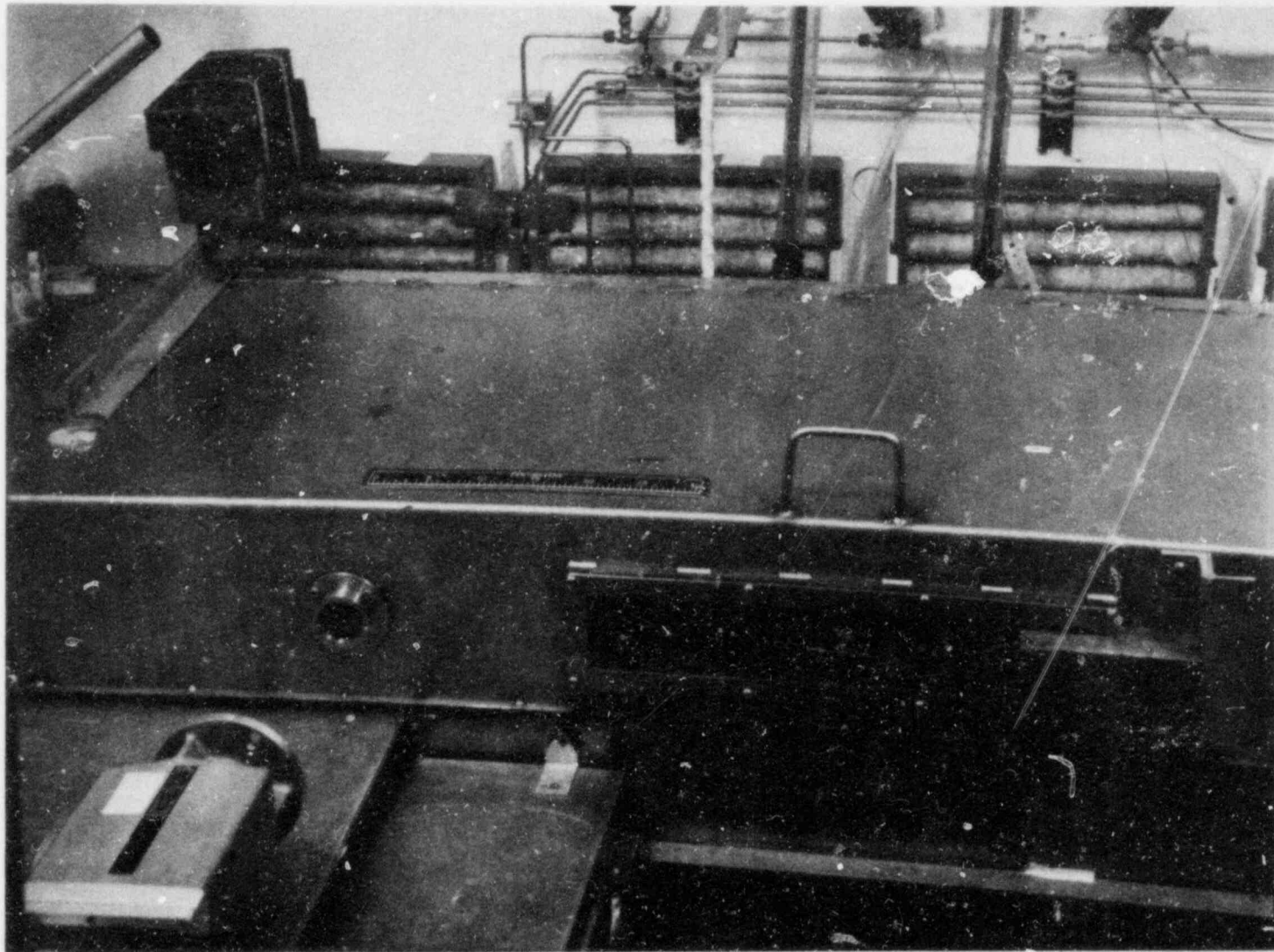


Fig. 5. Hot cell installation with containment box closed.



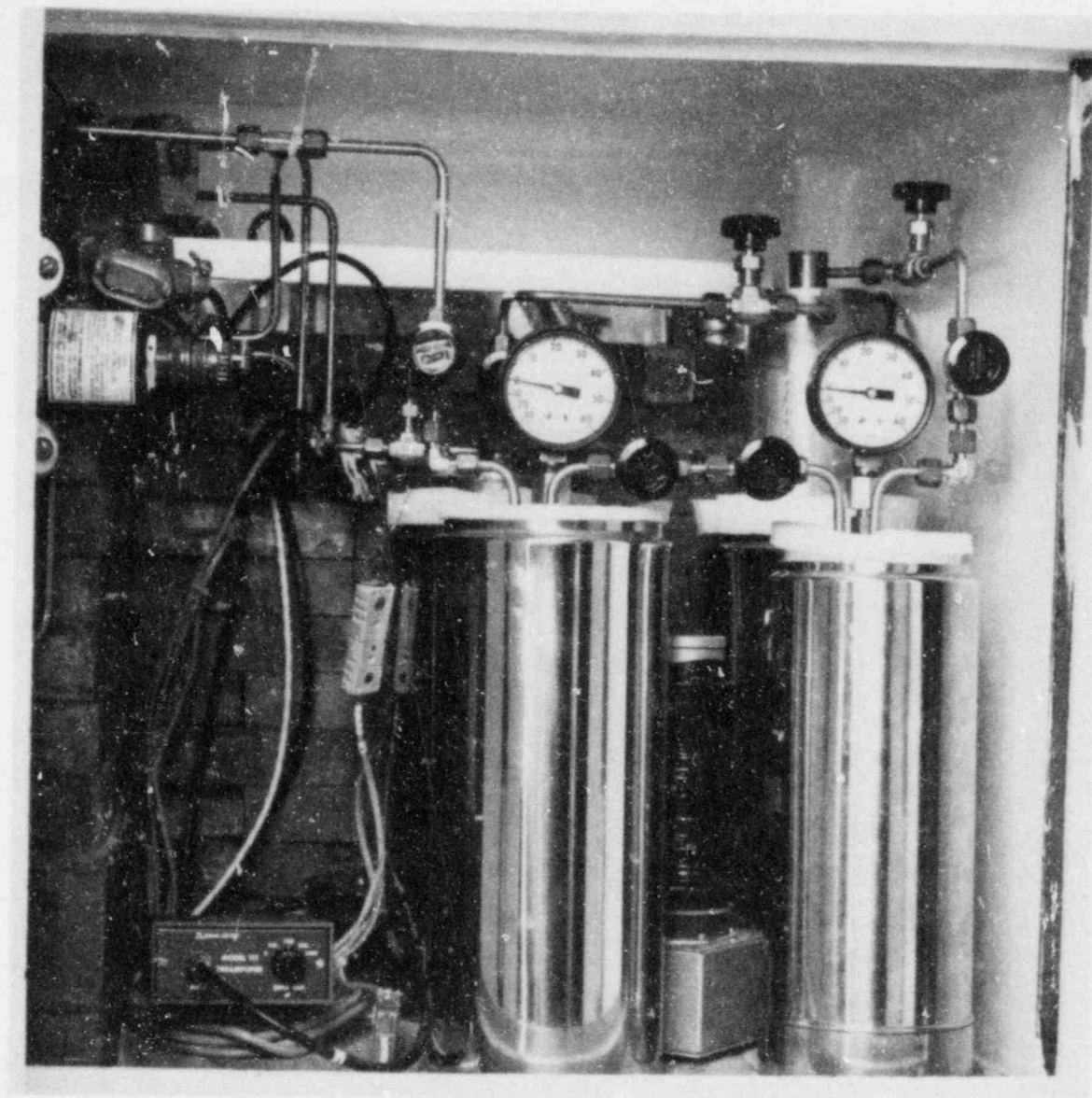


Fig. 6. Condenser, dryer, and two cold charcoal traps mounted around NaI(Tl) detector for  $^{85}\text{Kr}$  measurement.

### 3.4 INSTRUMENTATION

Instruments are used to continuously measure, control, and record temperature, to measure and record gas flow rates, to measure gas pressure, and to measure and record radioactivity during the course of each experiment. Although most of the sensors, and a few of the indicators, are located inside the hot cell, all devices for test control, operation, and data recording are located outside the hot cell as shown in Fig. 7. After the fuel specimen has been loaded into the furnace and the containment box closed, use of the manipulators to conduct a test is not required. The commercially supplied instruments and the RF power supply are listed in the Appendix.

#### 3.4.1 Temperature Measurement

Measurement, control, and recording of the furnace and fuel specimen temperature during a test were accorded top priority. The principal temperature instrumentation is illustrated in Fig. 8. Power input to the furnace is controlled manually by adjusting a large rheostat that controls plate voltage in the RF power supply via a saturable reactor. Both thermocouples — with bare Pt vs Pt-10% Rh for tests up to 1700°C or W-5% Re vs W-26% Re in ceramic thermowells for tests above 1700°C — and a two-color optical pyrometer are used to sense the temperature at the inlet end of the furnace. The optical pyrometer utilizes windows, a mirror, and the gas inlet hole through the ceramic end plug to view the end of the fuel specimen. The thermocouples are located in the same area, 1 to 2 cm from the end of the specimen. Calibration tests with unirradiated, instrumented fuel specimens were conducted to determine the temperature corrections and gradients for various geometries, temperatures, and gas flow rates.

Immediately downstream from the furnace, the temperature of the TGT (see Fig. 2) is monitored by 12 stainless steel clad Chromel-Alumel thermocouples and controlled by four heaters to provide a linear thermal gradient from ~850 to 150°C over its 35.6-cm length. Because gas flow rate and furnace temperature affect temperatures in the TGT, automatic controllers are used to operate the four heaters. The low temperature furnaces that heat the filter package and the steam lines, however, are insensitive to other test conditions and are operated manually. The steam generator, which is mounted outside the hot cell as shown in Fig. 9, is electrically heated; a resistance thermometer senses the temperature and supplies a signal to an automatic controller that maintains the desired temperature to a deviation of  $\pm 0.2^\circ\text{C}$ .

#### 3.4.2 Gas Flow Rate and Pressure Measurement

Gas flow in the test apparatus is shown schematically in Fig. 10. Purified helium is supplied to three regions: (1) the furnace thermowell, to protect the thermocouple and furnace inlet region from backflow of steam in the event of thermowell breakage; (2) the annulus in the furnace

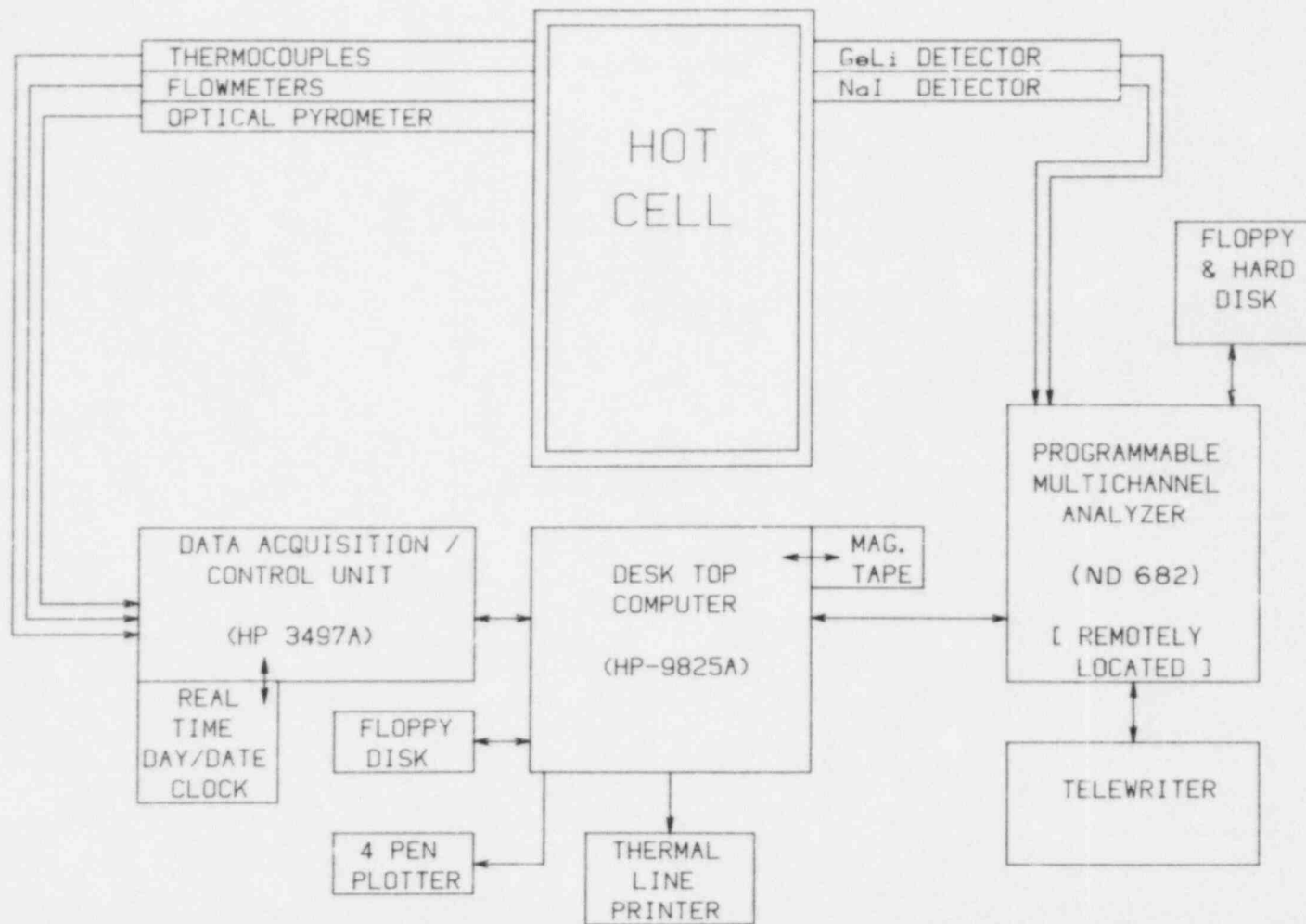


Fig. 7. Data acquisition and processing system for fission product release test.

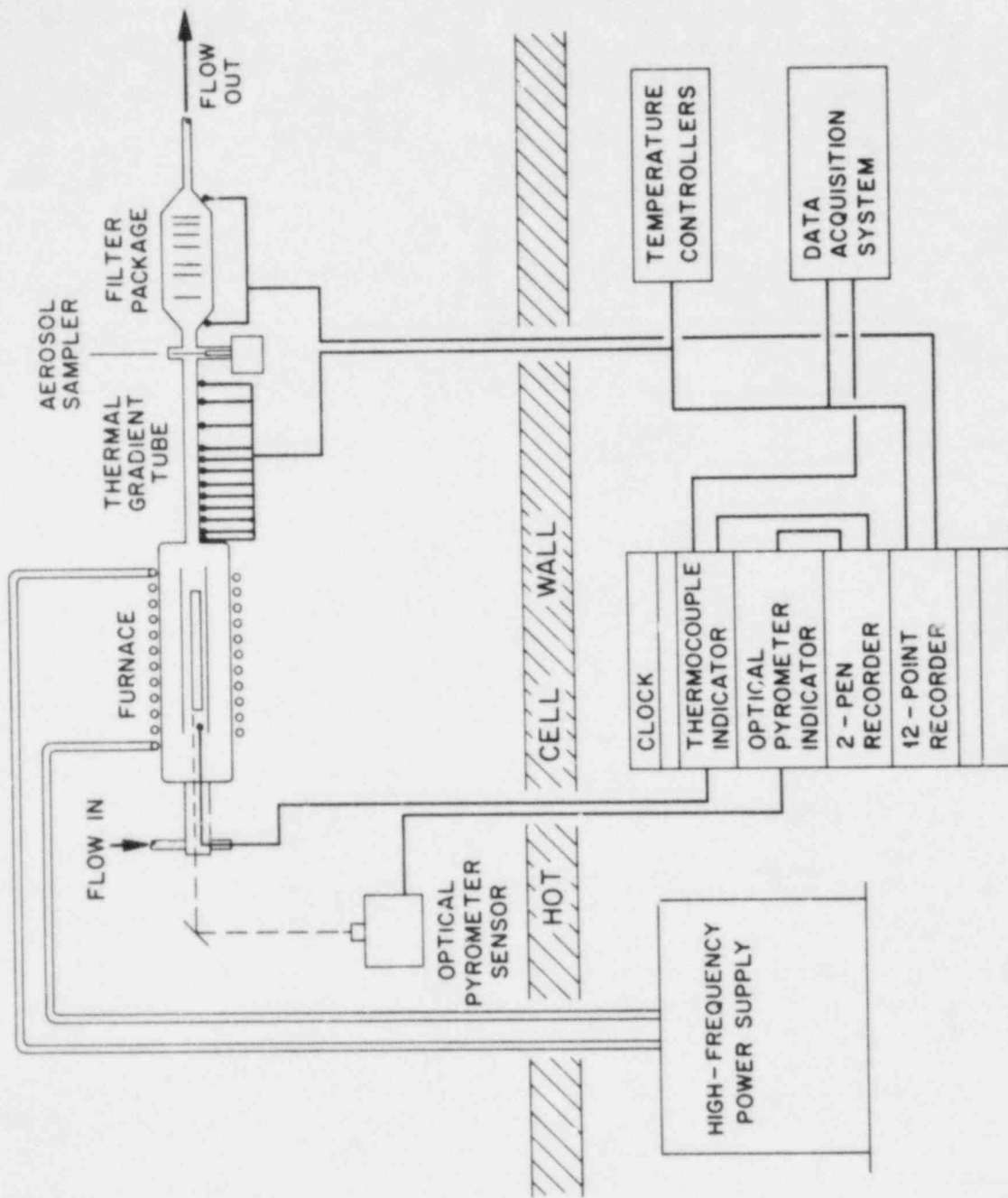


Fig. 8. Temperature instrumentation for fission product release apparatus.

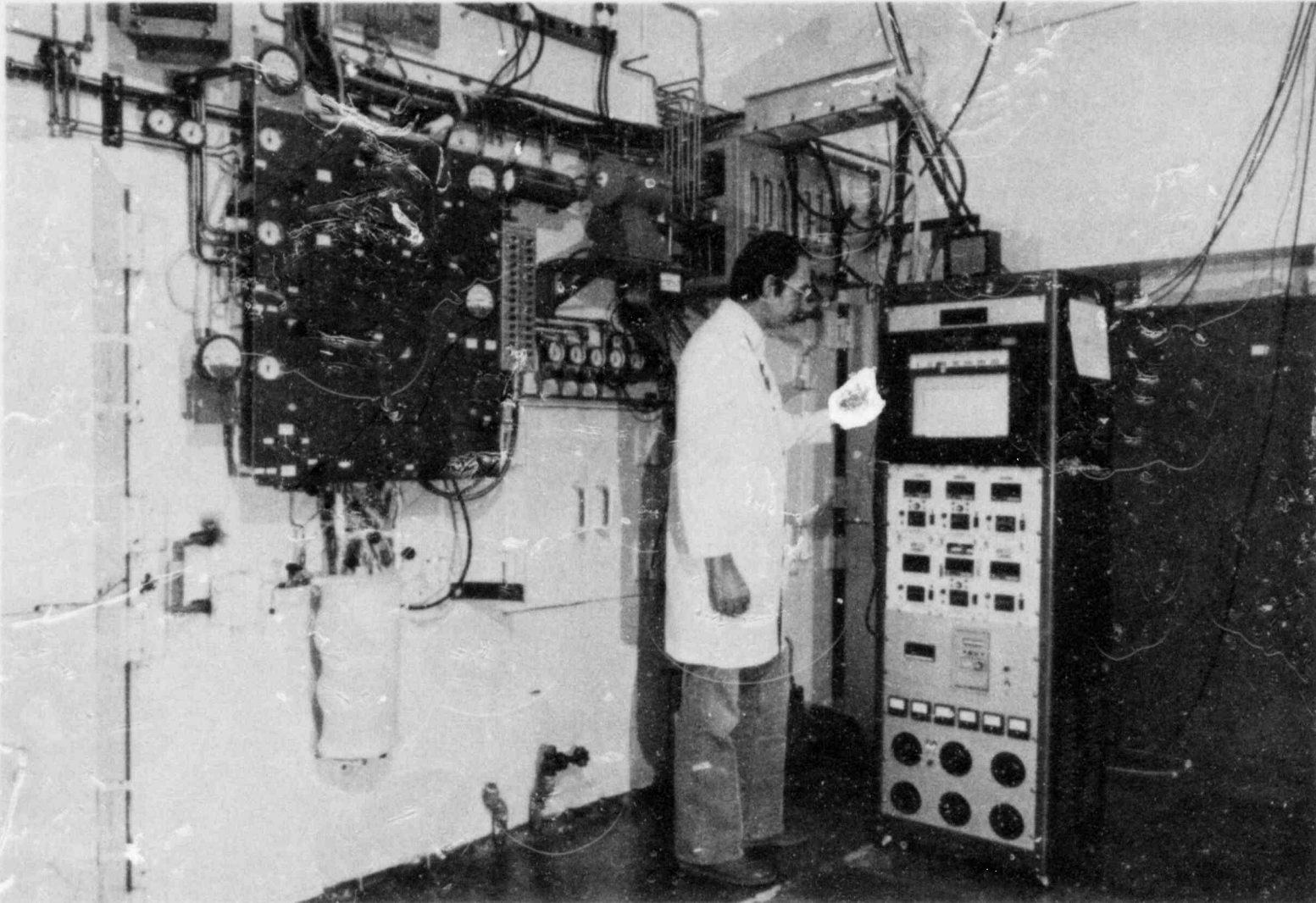


Fig. 9. Steam generator and flow measurement/control instrumentation mounted outside hot cell.



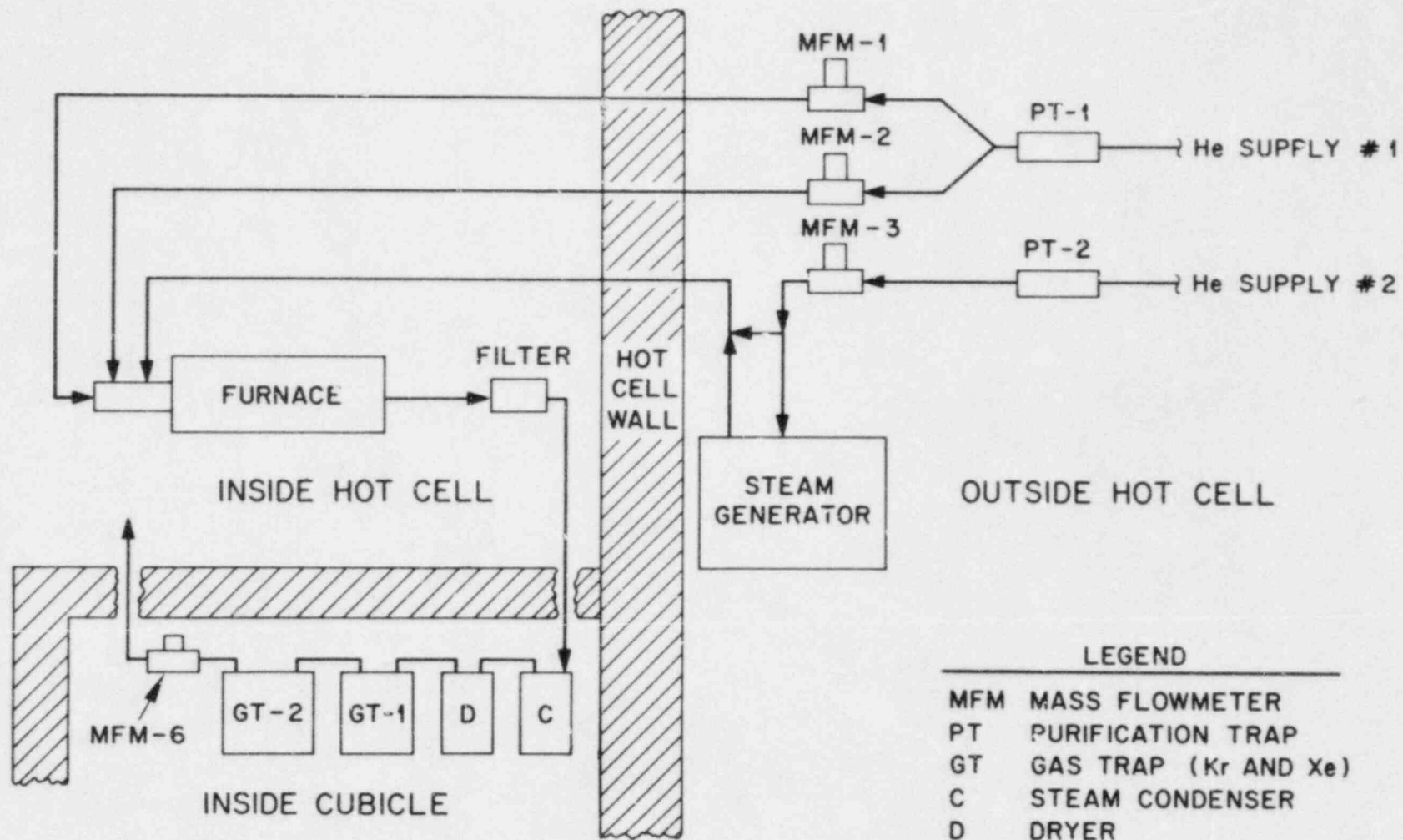


Fig. 10. Simplified flow diagram for fission product release test apparatus.

that contains the susceptor (either graphite or tungsten), again to guard against backflow of steam; and (3) the steam generator. (A small, but constant amount of helium is required to sweep the fission gases and any hydrogen produced in the furnace through the condenser and dryer to the gas collection traps.) These inlet helium flow rates and the outlet flow rate (a mixture of helium and hydrogen) are measured by mass flowmeters that operate on thermal conductivity principles (see Appendix). The pressures (~1.1 atm) are monitored at various points throughout the system during a test to ensure proper operation.

#### 3.4.3 Radiation Measurement

Two NaI(Tl) detectors and one Ge(Li) detector are used for on-line measurement of gamma-emitting fission products that are collected in the TGT, in the filter package, and in the cold charcoal traps (Fig. 3). The spectra are recorded at 1-min intervals during a test, and subsequently analyzed with a multichannel analyzer (MCA). The deposits in the TGT and the filter package are dominated by  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , whereas only  $^{85}\text{Kr}$  reaches the cold charcoal traps. In addition, qualitative data are obtained before, during, and after a test through use of a portable in-cell ion chamber and two Geiger-Mueller (G-M) tubes located in holes in the hot cell wall.

Following each test, the apparatus is completely disassembled and all components are carefully analyzed by Ge(Li) detector and MCA. This computer/MCA, shown in Fig. 11, is capable of simultaneously collecting data from three detectors and supplying quantitative data for the amount of fission products on each sample. Both long distances (up to 12 m) and lead attenuation (up to 6 cm) are used to analyze samples with very high levels of gamma activity, as high as 1000 R/h at contact.

#### 3.4.4 Chemical Analysis

Chemical species analysis in the gas phase at high temperatures (~700°C) has been considered. The species of particular interest would include CsI,  $\text{I}_2$ , HI, CsOH, and perhaps others as well. Two potential techniques, development of which has been supported in another laboratory on a limited basis, are laser Raman spectrometry and laser-induced fluorescence spectrometry.<sup>11</sup> Although the former initially appeared to be quite promising, preliminary experiments in vacuum showed a relatively low sensitivity for Raman compared with laser-induced fluorescence.<sup>12</sup> Further experiments using the fluorescence technique in an apparatus with steam flow are planned.

If this analysis method proves to be applicable to our apparatus, a special quartz or sapphire cell would be installed immediately downstream from the furnace (Fig. 2) as indicated in Fig. 12. Other components of such an analyzer would include a laser mounted outside the hot cell, a light-pipe to conduct the beam to the cell, a detector to sense the scattered radiation, and an MCA to record and analyze the signals. While such



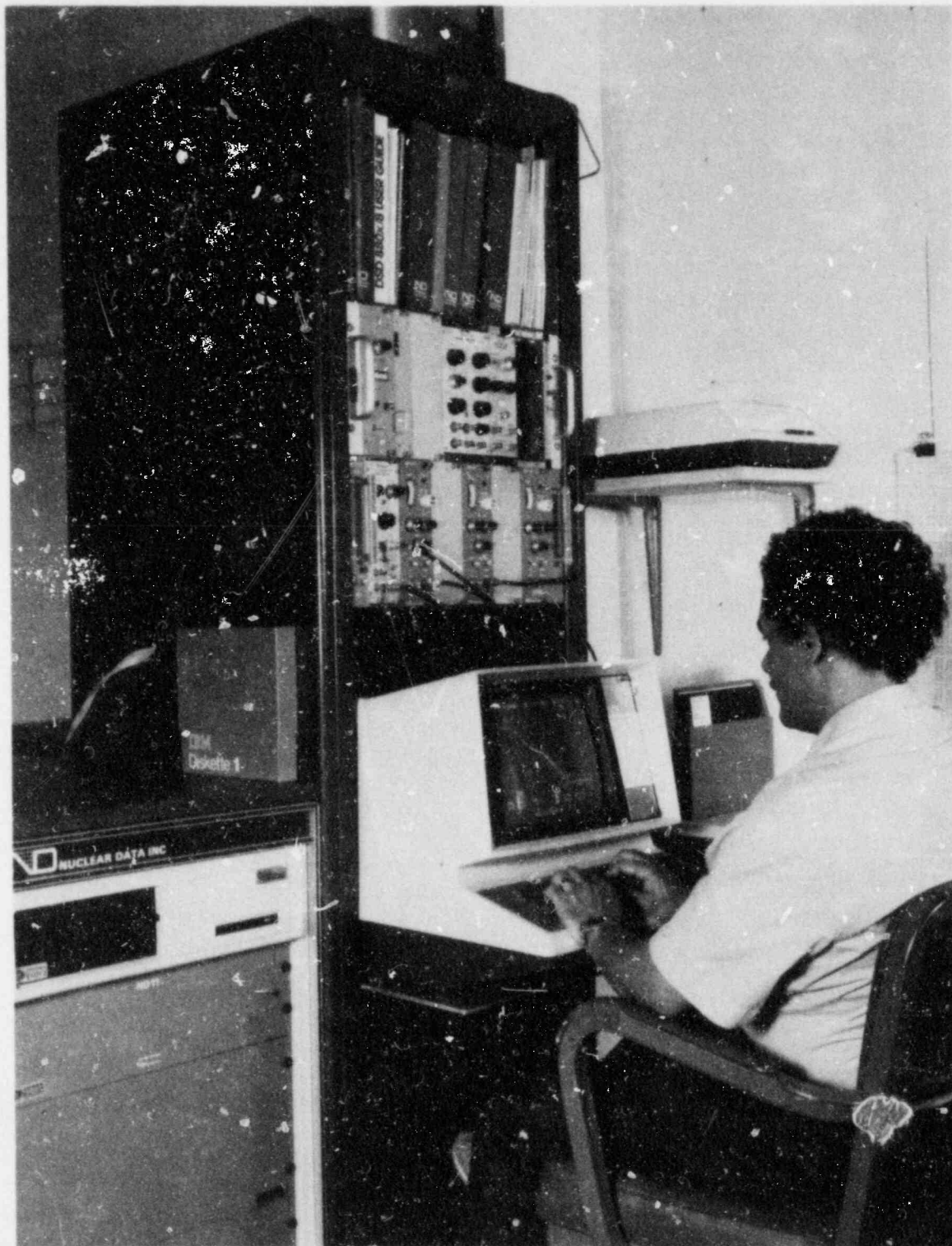


Fig. 11. Multichannel analyzer (MCA) used to measure quantities of radionuclides on test components.

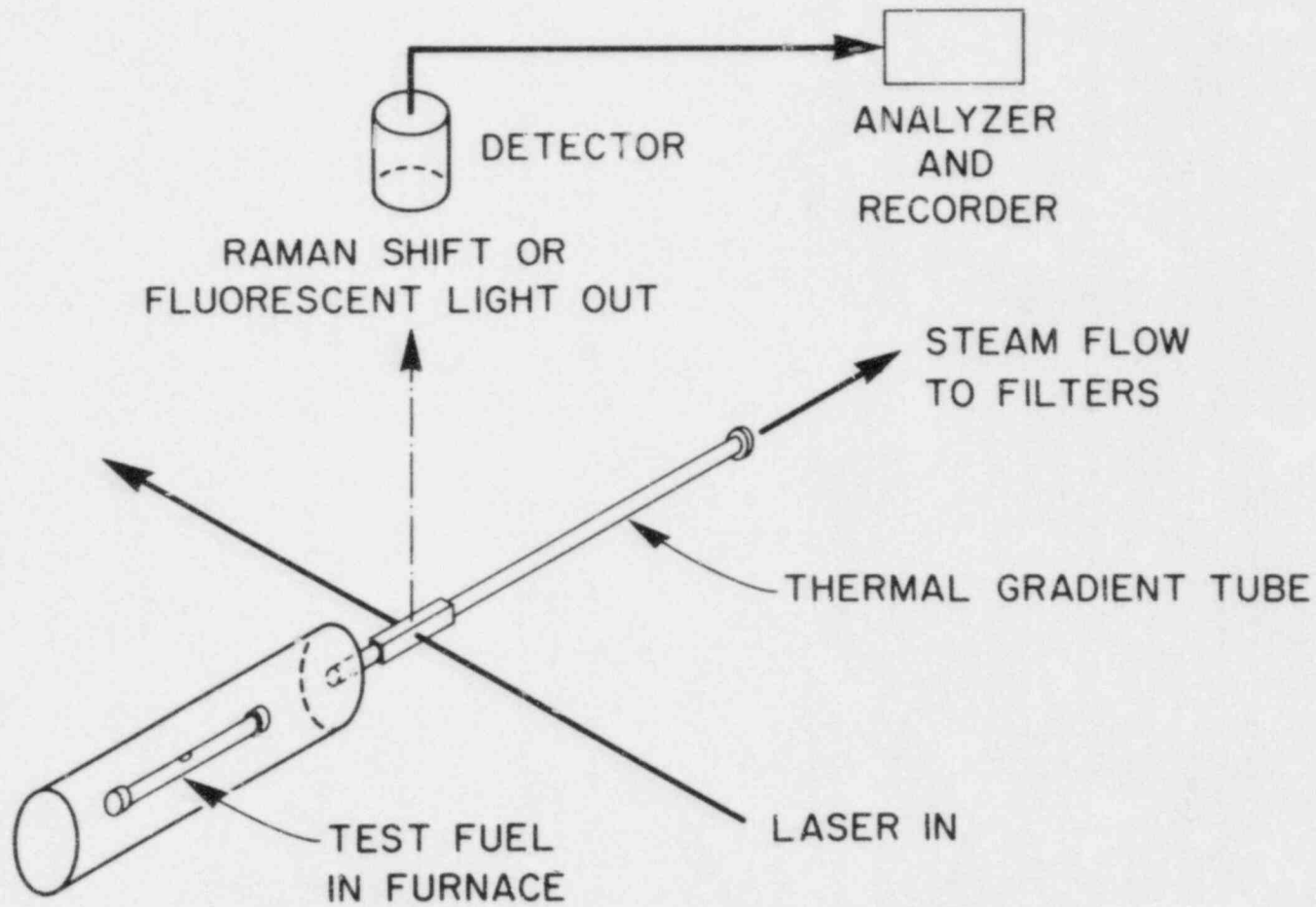


Fig. 12. Arrangement of apparatus for laser-induced fluorescence (LIF) spectrometry.

a system offers the potential for greatly expanding our analytical capability, a significant development and implementation effort will be required once applicability to our test conditions has been proven.

#### 3.4.5 Aerosol Sampling and Analysis

Data on the mass and the composition of the released aerosol as a function of time during tests were recognized as being of considerable interest. Accordingly, a sampling device was designed to provide continuous, differential aerosol collection. However, it was installed relatively late in the development phase, first being tested in release test HI-5. This collector, which is a miniature graphite rod driven slowly by a motor-operated screw across a narrow gap in the gas stream, is illustrated in Fig. 13; a view in the direction of gas flow through the sampler is shown in Fig. 14. The deposited aerosol particles trace a spiral path along the graphite rod, thereby allowing posttest examination of the aerosol sample at any particular time during the test. Graphite was selected as the deposition surface because of its low atomic number (6), which causes no interference with elemental analysis by the energy dispersive x-ray method (EDX) in a scanning electron microscope. The very small size (0.8 mm diam) and narrow deposition area (1 mm) were selected to avoid the high levels of radioactivity inherent in larger samples. This collector is installed between the TGT and the filter package, and is heated to 150°C to prevent steam condensation.

### 4. PRELIMINARY TESTING AND CALIBRATION

Following assembly of the test apparatus, the components were tested, both separately and together, to ensure proper operation and calibration. Some of these tests have been repeated at intervals (between fuel heatup tests) to account for apparatus modifications or to verify stable performance.

#### 4.1 FURNACE TESTING

Several tests of the furnace and induction coil were conducted to determine the axial thermal profile in the test zone for various coil configurations, heatup rates, and gas flow rates. (The inflowing steam-helium mixture tends to cool the inlet region.) The induction coils were concentrated at the ends to counteract end losses; the coil spacing varied from 0.95 cm (3/8 in.) at the ends to 1.59 cm (5/8 in.) at the center. Two additional coils were added at the inlet end to counteract the cooling effect of the incoming gas.

Two techniques were used to measure the axial thermal profile. In one case, six Pt vs Pt-Rh thermocouples were installed on a Zircaloy test specimen, thereby affording simultaneous temperature measurement at six locations in addition to the normal measurements by thermocouple and optical pyrometer at the inlet end. Two temperature profiles (at different argon flow rates) from this test, shown in Fig. 15, indicate a gradient of

ORNL DWG 84-613

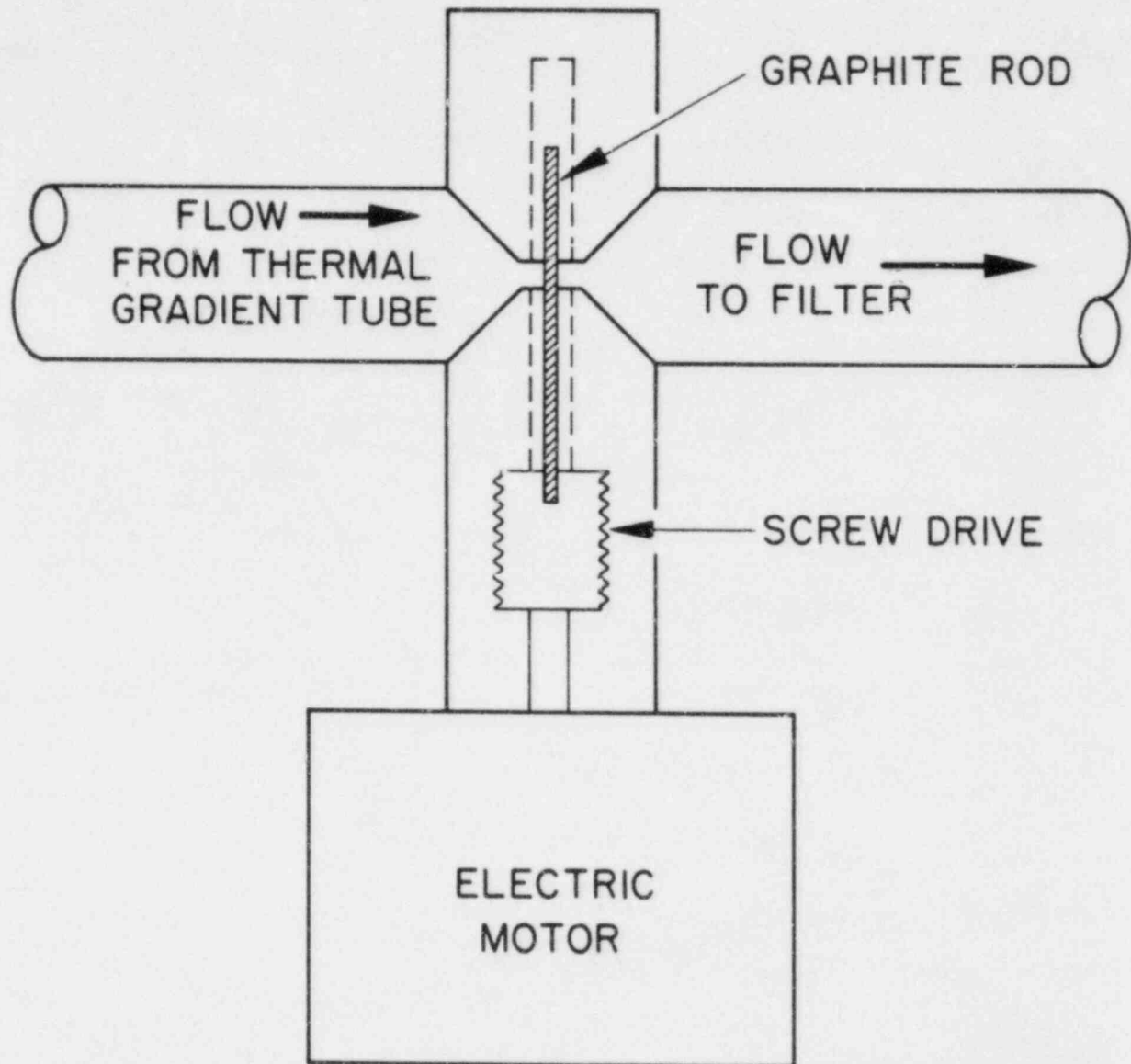


Fig. 13. Schematic diagram of miniature aerosol deposition sampler.





Fig. 14. Miniature aerosol sampler, showing path of gases/aerosol through interior.

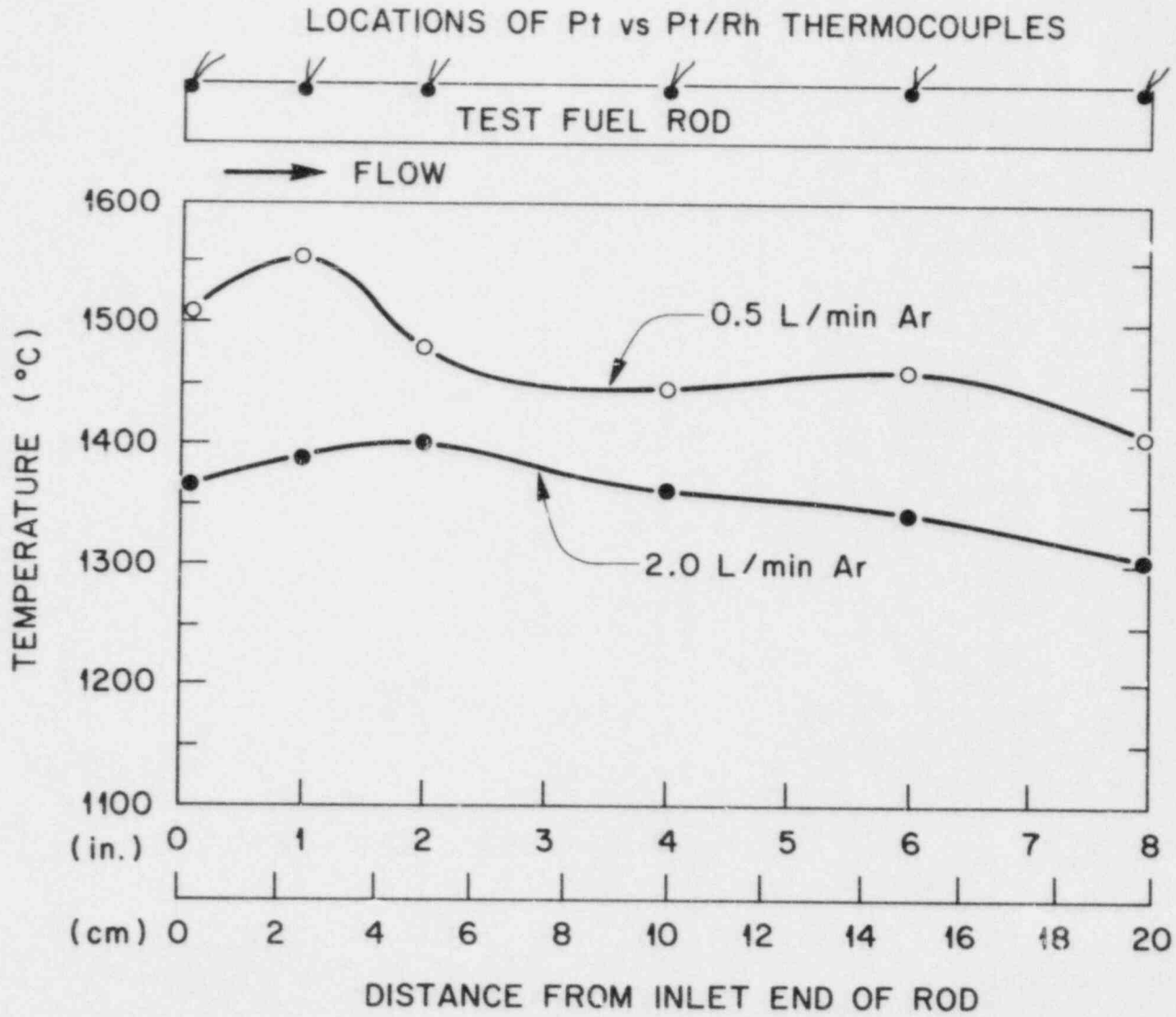


Fig. 15. Temperature profiles along test fuel rod as measured during furnace testing with a 22-turn induction coil.

some 100°C over the 20-cm specimen length. Compared to the inlet thermocouple and optical pyrometer, the maximum specimen temperature was ~75 and ~125°C higher, respectively.

In a later test, using a single movable Pt vs Pt-Rh thermocouple inside a Zircaloy specimen, the axial profile was measured at two different temperature levels. Using the same 22-turn coil configuration, but a different flow rate, 1.0 L/min steam plus 0.3 L/min He vs 2.0 L/min Ar in the previous test, a somewhat different axial profile was found, as shown in Fig. 16.

Several tests comparing the response of a W-5% Re vs W-26% Re thermocouple with the two-color optical pyrometer were conducted in order to select the best combination of mirror and windows used in the light path to the pyrometer. As may be seen in Fig. 3, the radiation must (1) pass through a small window at the inlet end of the furnace, (2) be reflected 90°, and (3) pass through a window in the stainless steel containment box before reaching the pyrometer. The windows and mirror are made of high purity fused silica (quartz), while back-silvering was found to be better than front-silvering for the mirror.

The results of a test to 2200°C are displayed in Fig. 17. Using a graphite rod as a test specimen, the pyrometer viewed the bottom of an axial hole in the inlet end and the thermocouple was installed in a similar hole in the outlet end. The furnace was heated slowly (~50°C/min) in a helium-hydrogen atmosphere. As indicated by the figure, the thermocouple and pyrometer agreed well up to ~1900°C; thereafter, the pyrometer indicated a significantly lower temperature. Since the immediate objective was conducting tests to a maximum of 2000°C, we concluded that this arrangement was adequate for the first series of tests using ZrO<sub>2</sub> ceramics, but would require further development for higher temperature tests using ThO<sub>2</sub> ceramic. (Not unexpectedly, the ZrO<sub>2</sub> furnace tube was damaged by the 2200°C temperature of this brief test.)

Another aspect of furnace testing included the installation and positioning (by trial and error) of a 5-cm-long platinum tube in the inlet end plugs of the furnace to serve as a gas preheater. Sufficient power from the induction coil was obtained to heat this tube, and the gas flowing through it, to ~800 to 1000°C during high-temperature operation, thus reducing the cooling effect of inflowing gas on the test specimen.

#### 4.2 THERMAL GRADIENT TUBE TESTING

Based on previous experience<sup>2</sup> and the performance of several test heaters in this program, a suitable method of heating the TGT was developed. First, two layers of quartz insulating tape (~0.1 mm thick) are wound on the fused quartz tube. Twelve Chromel-Alumel thermocouples, located at specific positions, are strapped to the tube, and bound with three layers of quartz tape. Next, the nickel-chromium heating wire is wound in place, and secured with additional layers of quartz tape. The



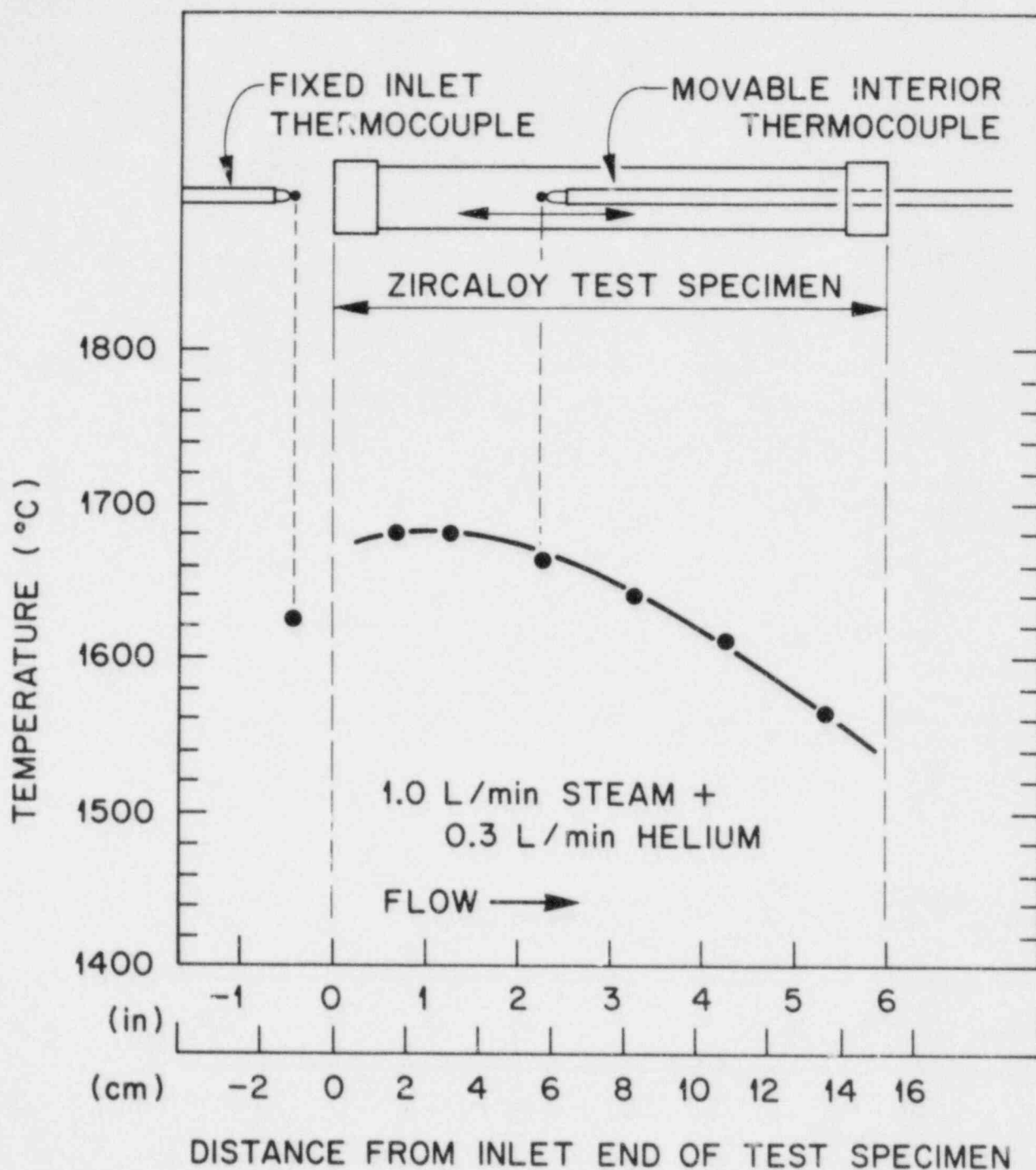


Fig. 16. Axial temperature profile indicated by interior platinum vs rhodium thermocouple during furnace testing.

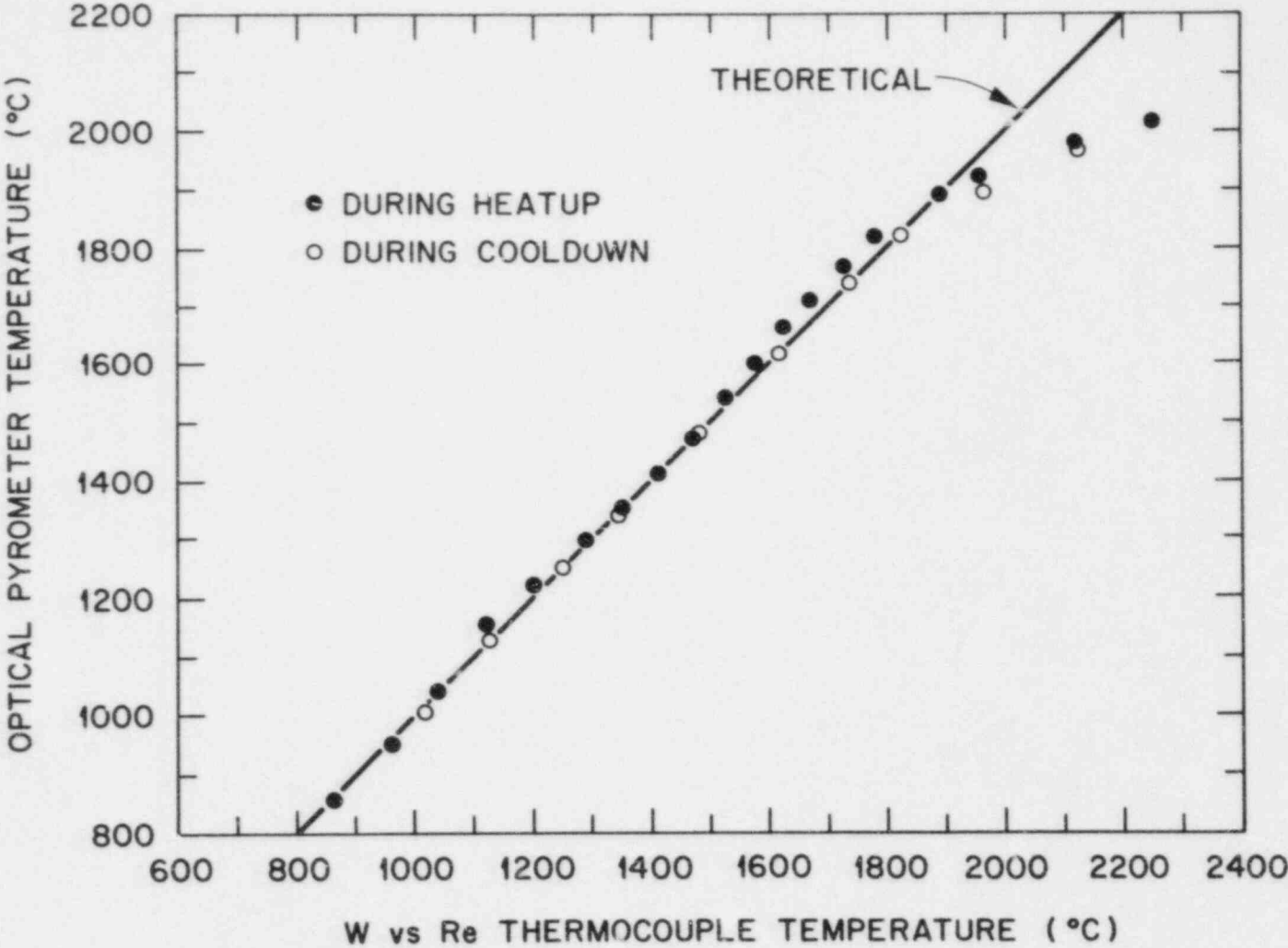


Fig. 17. Comparison of temperatures indicated by optical pyrometer and thermocouple at inlet end in furnace test.

35-cm-long tube is divided into four sections, each ~9 cm long, with the spacing of the windings gradually increasing toward the outlet end. Each heating section is individually controlled, and at least two thermocouples monitor the temperature.

Because a new assembly is required for each experiment, it is tested for the proper power input, heater response, and axial temperature profile prior to final test assembly.

#### 4.3 INERT GAS AND STEAM FLOW TESTING

The mass flowmeters used to measure helium flow through the apparatus were tested and certified as accurate by the ORNL Instrumentation and Controls Division. The entire system was pressure- and vacuum-tested to ensure gas purity. Standard test operating procedure includes rechecking for system integrity prior to each test.

The flow of steam from the steam generator was carefully measured as a function of both temperature and argon flow in preliminary testing. These calibration tests showed that, for an inert gas flow of 0.15 L/min, temperatures of 95 and 85°C provided the desired steam flows of 1.0 and 0.35 L/min, respectively. These values have been verified in control tests with tracers and in fuel tests.<sup>7-9</sup>

#### 4.4 RADIATION MEASUREMENT TESTING

As described in Sect. 2.4.3, NaI(Tl) and Ge(Li) detectors are used to obtain on-line fission product release data during each test. Because an accurate quantitative calibration of these detectors/spectrometers would be very difficult, we rely on posttest analysis with calibrated Ge(Li) detectors and MCAs for quantitative measurements. The <sup>85</sup>Kr counting system was initially a single-channel analyzer with teletype recording, precisely adjusted to detect the 514-keV gamma ray by use of a <sup>85</sup>Kr source. More recently, the <sup>85</sup>Kr detector has been connected to the MCA. This detector, along with the detectors that monitor the TGT and filter package, are checked prior to each test for position and measurement, using sources of <sup>85</sup>Kr and <sup>137</sup>Cs. The MCA is regularly calibrated using NBS standard sources.

The in-cell ion chamber and the G-M counters mounted in the hot cell wall are not calibrated, but are checked for proper operation before each test. One G-M counter monitors the filter pack and the other monitors the low temperature end of the TGT. Both are located inside 1-in.-diam holes that penetrate the cell wall.

### 5. CONTROL TESTS

After assembly of the test apparatus, a series of control or verification tests were conducted to ensure proper operation of the entire system, to calibrate certain instruments, and to determine the effects of

test conditions on unirradiated fuel specimens. Minor changes and adjustments in both the hardware and the operating procedures were made during these tests; these changes are summarized in Table 2 and will be discussed in the order in which they were conducted. Test C-5 was aborted because of experimental problems and has been omitted. Three of the tests contained radioactive tracers ( $^{130}\text{I}$  and/or Zircaloy activation products:  $^{51}\text{Cr}$ ,  $^{60}\text{Co}$ ,  $^{95}\text{Zr}$ ,  $^{113}\text{Sn}$ , and  $^{125}\text{Sb}$ ), which were used to test radioactivity measurement and to study the behavior of these elements in our test apparatus. Temperature data for tests C-1 through C-6 are summarized in Table 3.

### 5.1 TESTS C-1 AND C-2

These two tests had as their common objectives testing the furnace with steam flow, investigating the effect of the high temperature steam on a Zircaloy specimen, and determining the steam flow rate under conditions expected for the tests with radioactive fuel specimens. Construction and operating data for both tests are listed in Table 2, while temperature and power histories are shown in Figs. 18 and 19. As determined in temperature calibrations, the optical pyrometer data require a correction factor of  $\sim 150^\circ\text{C}$  at  $1700\text{--}2000^\circ\text{C}$  because of absorption in the windows and the mirror. Because the furnace thermocouple is mounted in the inlet end of the furnace and does not make contact with the Zircaloy specimen, it requires a correction that is strongly dependent on the distance from the end of the specimen. The Pt-10% Rh vs Pt thermocouple in test C-1 was mounted  $\sim 1$  cm from the end of the Zircaloy specimen; it indicated a maximum of  $1690^\circ\text{C}$  before failing, perhaps by melting, whereas the optical pyrometer indicated a maximum of  $1700^\circ\text{C}$ . Because the Zircaloy specimen in test C-1 was partially molten, the melt may have contacted the thermocouple and caused failure. (The melting point of pure Zircaloy has been reported to be  $\sim 1750^\circ\text{C}$ ,<sup>13</sup> but increases rapidly with oxidation.) In test C-2, the thermocouple was mounted  $\sim 2$  cm from the end of the specimen, which was typical Zircaloy-clad  $\text{UO}_2$ . In this case, the thermocouple indicated  $\sim 150^\circ\text{C}$  less than the optical pyrometer, or  $\sim 300^\circ\text{C}$  less than the specimen temperature. Since gas (argon and steam) flow conditions were similar in the two tests, we concluded that, at specimen temperatures of  $\sim 1800^\circ\text{C}$ , a temperature correction for the thermocouple of  $150^\circ\text{C}/\text{cm}$  from the specimen was reasonable. Temperature data for the five control tests are summarized in Table 3.

As shown in Table 2, the steam flow rate into the furnace was slightly more than 1 L/min in each test, and the combined argon flow rate (to steam generator and to susceptor purge) varied between 0.30 and 0.60 L/min. The steam mass flowmeter used in these tests provided inconsistent readings; the data shown in Table 2 were derived from the masses of water collected in the condenser and dryer and from measurements of the total gas flow rate (including hydrogen) exiting the apparatus.

### 5.2 TEST C-3

This test was similar to tests C-1 and C-2 except that a source of elemental iodine, traced with  $^{130}\text{I}$ , was included to study the behavior of

Table 2. Construction and operating data for control tests<sup>a</sup>

Property or condition	Test No.				
	C-1	C-2	C-3	C-4	C-6
Test specimen	Zircaloy	Zircaloy-UO <sub>2</sub>	Zircaloy-UO <sub>2</sub>	Zircaloy-UO <sub>2</sub>	Zircaloy-UO <sub>2</sub>
Tracer material	None	None	<sup>130</sup> I <sub>2</sub>	<sup>130</sup> I <sub>2</sub> and irradiated Zircaloy	Irradiated Zircaloy
Furnace thermocouple	Pt/Rh	Pt/Rh	Pt/Rh	Pt/Rh	W/Re with thermowell
Susceptor material	Tungsten	Tungsten	Tungsten	Tungsten	Graphite
Test atmosphere	Ar-steam	Ar-steam	Ar-steam	Ar-steam	He-steam
Flow rates (L/min):					
Ar to steam generator	0.30	0.30	0.30	0.30	0.15
Ar to susceptor purge	0.30	0.30	0.20	0.15	0.15
Steam out	0.88	1.03	0.99	0.92	0.54
Steam in	~1.1	1.19	1.21	1.30	0.89
Specimen temperature (°C)	~1850	1820	1830	1700	2100
Heatup rate (°/min)	42	48	72	75	140
Time at T <sub>max</sub> (min)	~20	30	20	20	20

<sup>a</sup>Test C-5 was aborted because of experimental problems and produced no valid data.



Table 3. Comparison of temperature data in tests C-1 through C-6

Test No.	Thermocouple location <sup>a</sup> (cm)	Maximum indicated temperatures (°C)		Estimated maximum specimen temperature (°C)
		By thermocouple	By optical pyrometer	
C-1	1	1690 <sup>b</sup>	1700	~1850
C-2	2	1526	1670	1820
C-3	1	1692	1680	1830
C-4	1	1564	1555	1700
C-6	1	1940	1940	2100

<sup>a</sup> Approximate distance from thermocouple to end of fuel specimen.

<sup>b</sup> Thermocouple failed during test.

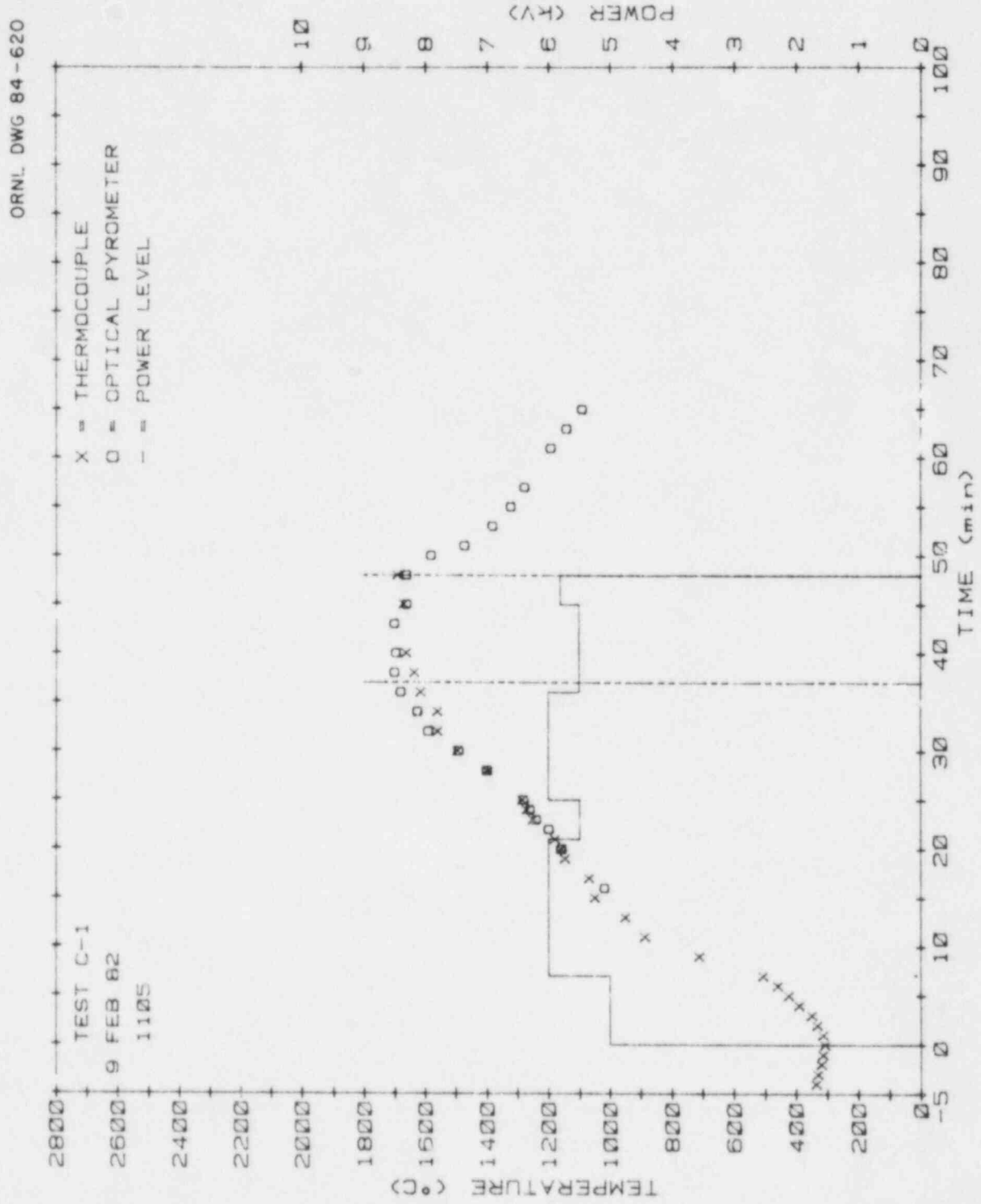


Fig. 18. Temperature and power histories of test C-1.

ORNL DWG 84-619

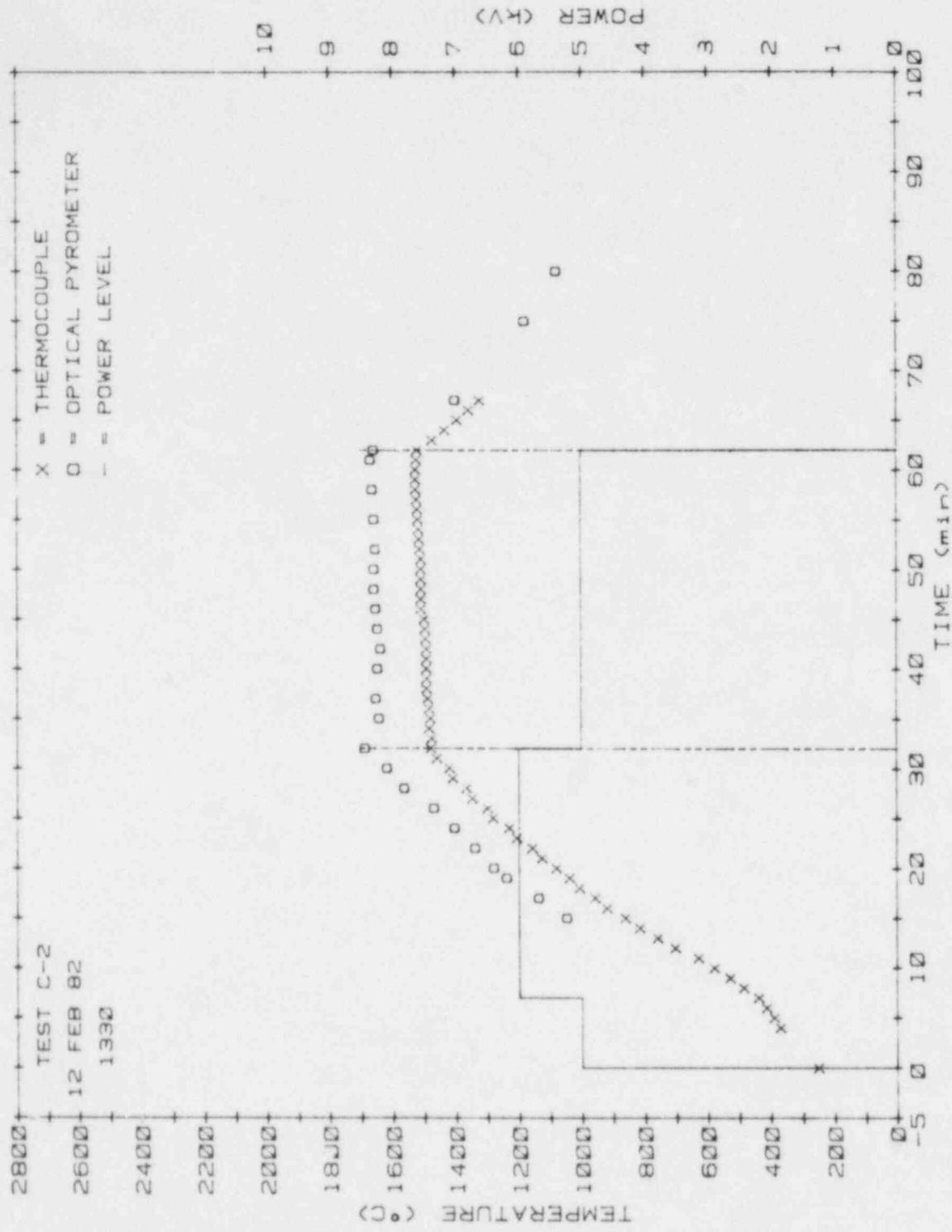


Fig. 19. Temperature and power histories of test C-2.

I<sub>2</sub> in the apparatus, especially in the TGT. Test materials, temperatures, and gas flow rates were very similar to those in test C-2 (see Table 2 and Fig. 20). Following the test, the test C-3 specimen was compared with an identical untested specimen in Fig. 21. The Zircaloy cladding appeared to be completely oxidized to ZrO<sub>2</sub> and was quite brittle. In addition, diameter measurements indicated oxidation induced increases of up to 11.5% for the cladding and 8.6% for the end caps; the length increase was about 9%.

Utilizing a technique developed in earlier studies,<sup>14</sup> the iodine tracer was prepared by combining <sup>129</sup>I with palladium to form PdI<sub>2</sub>. This material, in powder form, was sealed in a quartz (fused silica) ampoule for irradiation in order to generate the desired amount of 12.4-h <sup>130</sup>I. Following the irradiation in the ORNL Bulk Shielding Reactor, the ampoule was analyzed by gamma spectrometry, then broken and immediately loaded into a platinum tube in the inlet end of the furnace at room temperature. Because PdI<sub>2</sub> begins to dissociate at <400°C,<sup>15</sup> the ampoule was positioned in the inlet end plug where the temperature would not reach ~400°C until the specimen was at least 1300°C. The PdI<sub>2</sub> should have been completely dissociated, releasing the <sup>130</sup>I vapor into the flowing steam-argon atmosphere, during the first 10 min of the 20-min test.

After the test the apparatus was disassembled and inspected, and the amount of <sup>130</sup>I on each component was determined by gamma spectrometry, using a calibrated Ge(Li) detector and an MCA. The distribution of <sup>130</sup>I in the test apparatus is summarized in Table 4; only 4.7% of the total iodine injected into the system was found on the TGT whereas 9.61% was found on the filter media, and 49.1% passed through the filters and was adsorbed on the heated charcoal. Only 85.5% of the original <sup>130</sup>I was found at the end of the test. As shown in Fig. 22, no <sup>130</sup>I deposited on the platinum TGT until the temperature had dropped to ~450°C; the maximum concentration occurred at ~410°C, then declined rapidly. This distribution of <sup>130</sup>I in the furnace, TGT, and filter package is consistent with previous experience in collecting elemental iodine under these conditions.<sup>14</sup> The iodine deposit in the TGT probably results from an iodine formed from impurities in the system. Relatively small amounts of iodine deposited on Teflon surfaces compared with stainless steel surfaces. More iodine was found on the stainless steel components of the filter holders than on the filter media, which is not unusual behavior for molecular iodine (I<sub>2</sub>).

### 5.3 TEST C-4

Two forms of radioactive tracer material, elemental <sup>130</sup>I (as in test C-3) and irradiated Zircaloy containing <sup>51</sup>Cr, <sup>60</sup>Co, <sup>95</sup>Zr, <sup>113</sup>Sn, and <sup>125</sup>Sb, were used in test C-4. In this way, indicators were included to monitor the behavior of both fission products and structural materials in our apparatus. The irradiated Zircaloy was in the form of two thin strips, ~1 mm × 0.4 mm × 15 cm long, weighing 3.16 and 3.05 g; in conducting this tests, one strip was placed on each side of the fuel specimen in the furnace.

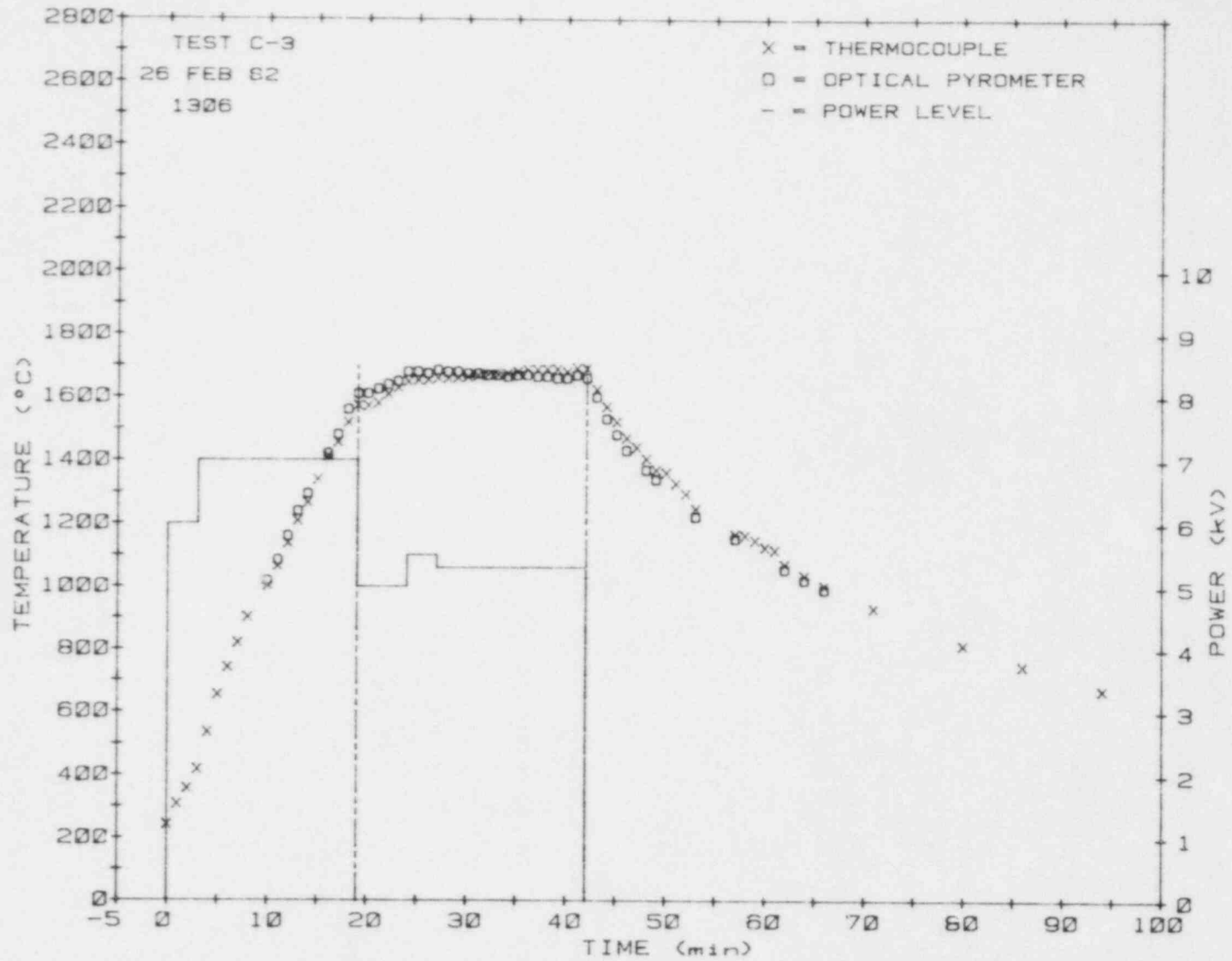


Fig. 20. Temperature and power histories of test C-3.



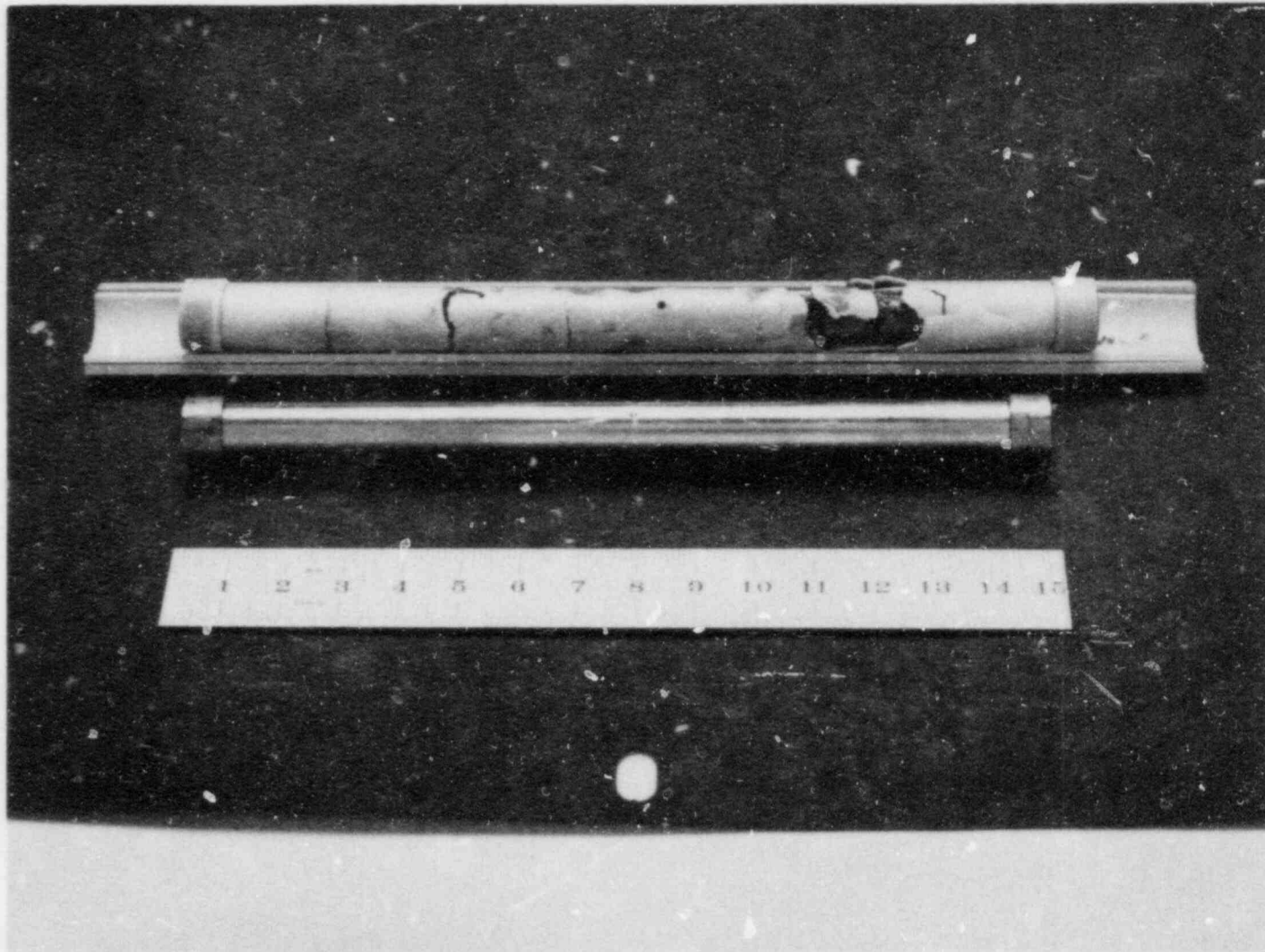


Fig. 21. Comparison of the oxidized fuel specimen (Zircaloy-clad  $UO_2$ ) from test C-3 with an untested specimen.

Table 4. Distribution of  $^{130}\text{I}$  in test C-3

Test component/collector	Approximate temperature (°C)	Amount of $^{130}\text{I}$	
		( $\mu\text{Ci}$ )	% of original total
<b>Pretest:</b>			
$^{130}\text{I}$ source ampoule		398.2	100
<b>Posttest:</b>			
PdI <sub>2</sub> ampoule	~700	0.01	0.003
Furnace and specimen	1000-1800	0.08	0.02
Thermal gradient tube	150-850	18.67	4.69
O-ring and Teflon connector to filter pack	150	8.14	2.04
Teflon entrance cone to filters	150	1.60	0.40
Glass wool filter	150	38.28	9.61
25- $\mu\text{m}$ filter	150	7.36	1.85
11- $\mu\text{m}$ filter	150	7.37	1.85
Teflon holder and SS screen for glass wool	150	28.09	7.05
Teflon holder and SS screen for filters	150	28.49	7.15
Other filter pack body parts	150	5.55	1.39
Charcoal 1	150	161.1	40.46
Charcoal 2	150	22.81	5.73
Charcoal 3	150	5.50	1.38
Charcoal 4	150	1.33	0.33
Empty charcoal holders (plastic)	150	4.90	1.23
Line to condenser and condensate	150	1.27	0.32
Total recovered		340.55	85.5
Unaccounted for		57.65	14.5

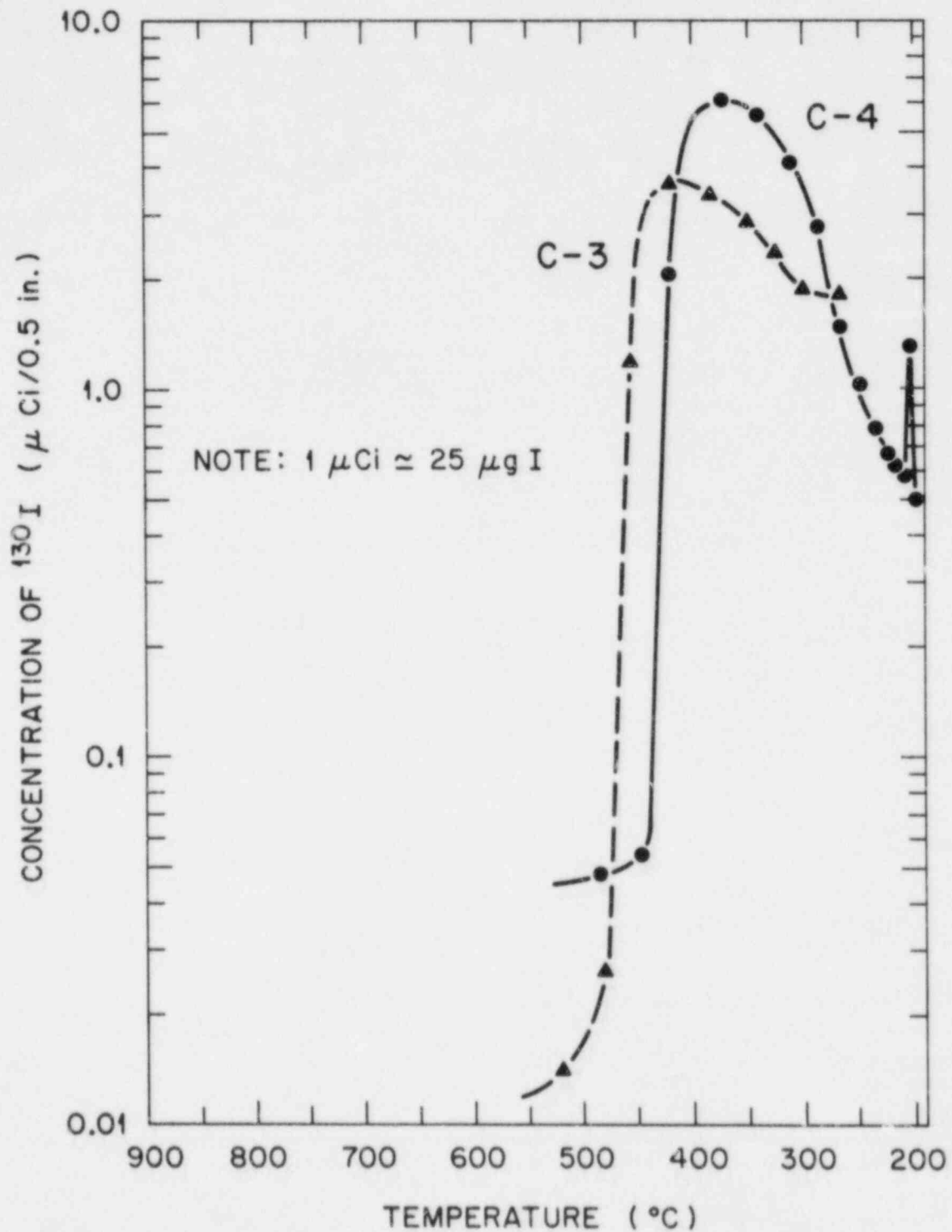


Fig. 22. Distribution of  $^{130}\text{I}$  along thermal gradient tubes in tests C-3 and C-4.

The construction details and operating conditions for this test are summarized in Table 2, while the temperature and flow histories are illustrated in Fig. 23. Temperature control of the specimen was quite good, and the hydrogen generation peak resulting from the steam-Zircaloy reaction was clearly defined, as shown in this figure.

Posttest examination revealed that the Zircaloy cladding was heavily oxidized; the appearance was very similar to that of the fuel specimen in test C-3 (Fig. 21). In addition, the irradiated Zircaloy strips had oxidized and disintegrated into powder and short pieces <1 cm long. The mass of water collected in the condenser and the combined flow data indicated the flow rates shown in Table 2.

The test apparatus was completely disassembled and each component was analyzed by gamma spectrometry. The distribution of  $^{130}\text{I}$ , similar to that in test C-3, is summarized in Table 5. Unlike test C-3, no  $^{130}\text{I}$  was detected in the water from the condenser. A somewhat larger fraction of the  $^{130}\text{I}$  was collected on the filters, indicating more particulate material was present in this test. The deposition profile for  $^{130}\text{I}$  in test C-4 is compared with that of test C-3 in Fig. 22.

The distribution of the Zircaloy activation products is summarized in Table 6. As expected, the transport of  $^{95}\text{Zr}$  beyond the furnace was less than that for the more volatile components of the Zircaloy; much larger fractions of  $^{113}\text{Sn}$  and  $^{125}\text{Sb}$  (48 and 43% respectively) than of  $^{51}\text{Cr}$  and  $^{60}\text{Co}$  were found outside the furnace. Rapidly declining concentrations of tin and antimony were detected at the inlet end of the TGT and are shown graphically in Fig. 24. The deposition behavior of antimony on the platinum TGT liner appears to be similar to previously observed antimony deposition in gold TGT liners.<sup>2</sup>

#### 5.4 TEST C-6

Test C-5 was prepared, but experimental problems were encountered during flow testing in the preheat phase. Electrical interference made thermocouple readings doubtful, and abnormally high back pressures to flow were measured. The test sequence was interrupted, and after thermocouple revisions and replacement of the filters, the test was begun again denoted as C-6 (see Table 2 for construction and operating details). This test, run at a higher temperature (2100°C), included irradiated Zircaloy strips as in test C-4, but did not contain  $^{130}\text{I}$  tracer. The primary objectives were to test the performance of a graphite susceptor and to investigate temperature measurement and test effects on the fuel specimen. (Although graphite susceptors had been used previously in furnace testing and temperature calibration, this was the first test using both a graphite susceptor and a steam test atmosphere.) Temperature and flow histories are shown in Fig. 25, with Fig. 26 illustrating the posttest specimen appearance. Apparently, the Zircaloy cladding had been almost completely molten early in the test, then was thoroughly oxidized to  $\text{ZrO}_2$ ; only powder and very small chips of oxide remained under and beside the  $\text{UO}_2$

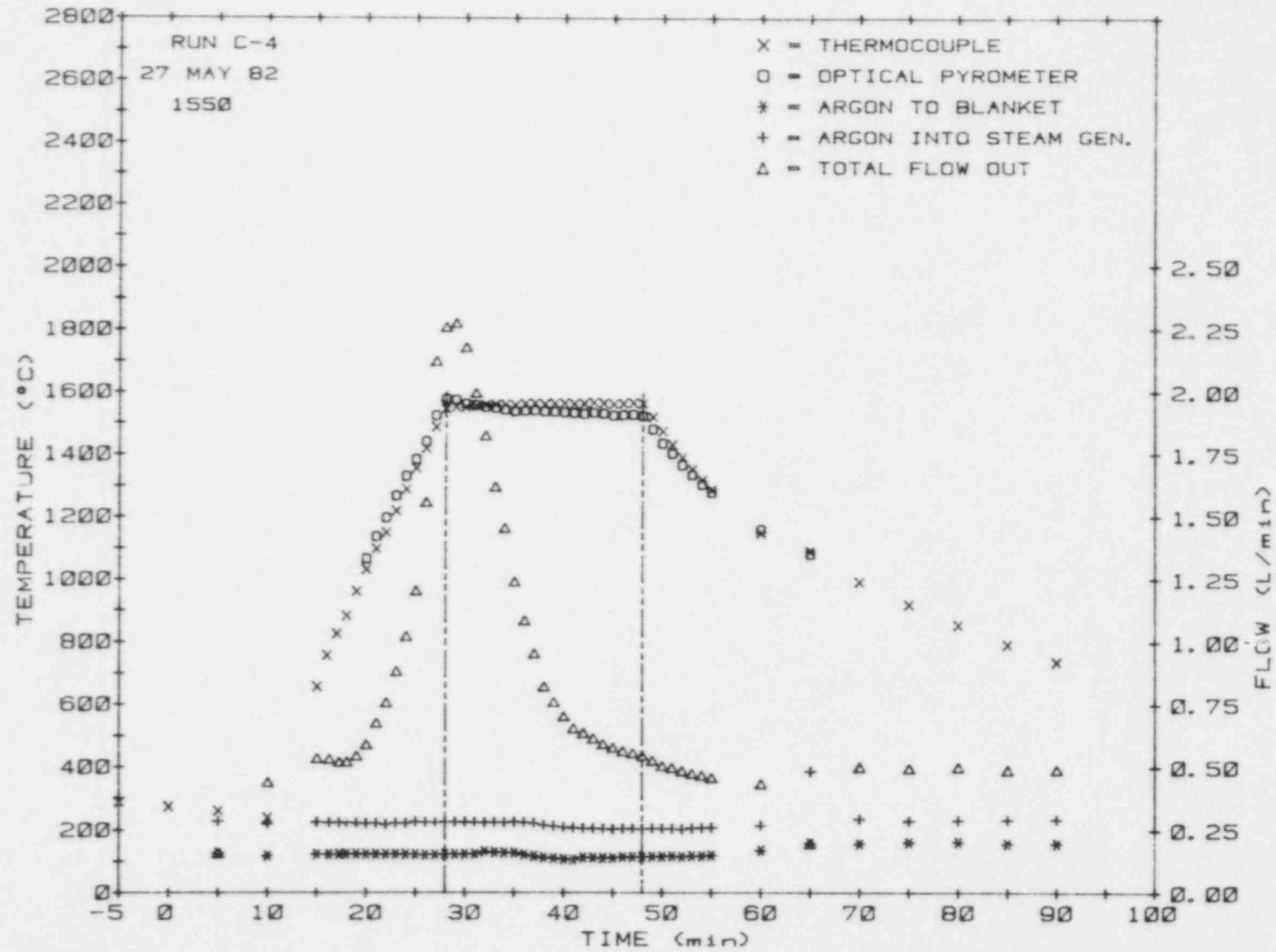


Fig. 23. Temperature and flow histories of test C-4.



Table 5. Distribution of  $^{130}\text{I}$  in test C-4

Test component/collector	Approximate temperature (°C)	Amount of $^{130}\text{I}$	
		( $\mu\text{Ci}$ )	% of original total
Source ampoule (pretest)		409.0	100
Posttest data:			
Source ampoule	~700	0.013	0.003
Fuel specimen and $\text{ZrO}_2$ boat	1700	0.0	0.0
$\text{ZrO}_2$ furnace tube	1000-1700	0.2122	0.519
$\text{ZrO}_2$ sliding plug	1700	0.0040	0.001
$\text{ZrO}_2$ plug right	1000-1400	0.0185	0.005
$\text{ZrO}_2$ insulator right	600-1200	7.256	1.774
Total in furnace		7.491	1.83
Thermal gradient tube	200-860	27.85	6.81
Filter inlet tube (Teflon)	150	8.17	2.00
Filter inlet cone (Teflon)	150	2.37	0.58
Glass wool prefilter and Teflon holder	140	121.5	29.71
HEPA filters and Teflon holder	140	62.95	16.39
Total on filters and holders		195.0	47.68
Charcoal 1	140	127.6	31.20
Charcoal 2	140	16.97	4.15
Charcoal 3	140	5.719	1.40
Charcoal 4	140	0.8896	0.22
Holder for charcoal	140	4.138	1.01
Total on charcoal and holder		155.3	37.97
Condenser	0	0.0	0.0
Total released from furnace		378.2	92.46
Percent of original recovered			94.29

IMAGE EVALUATION  
TEST TARGET (MT-3)

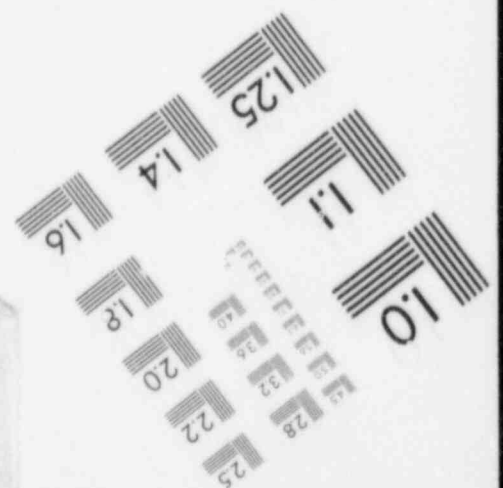
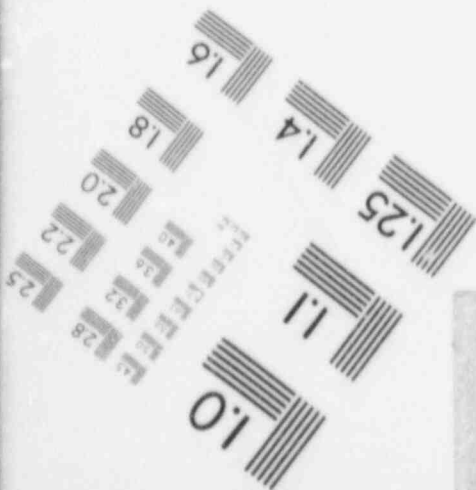
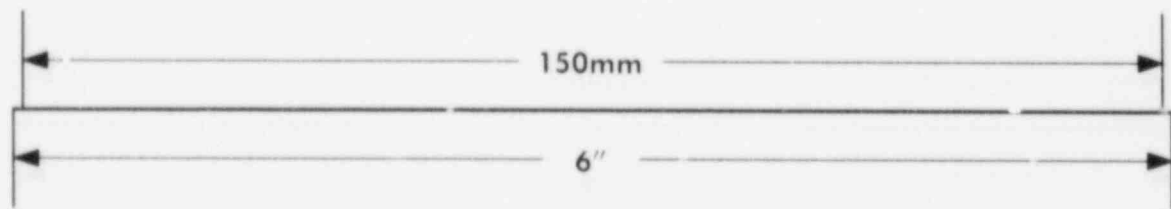
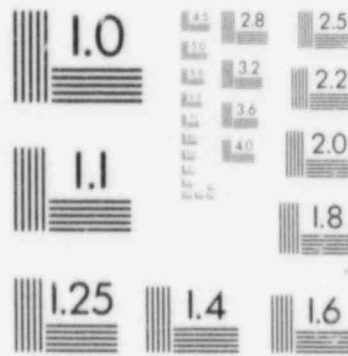
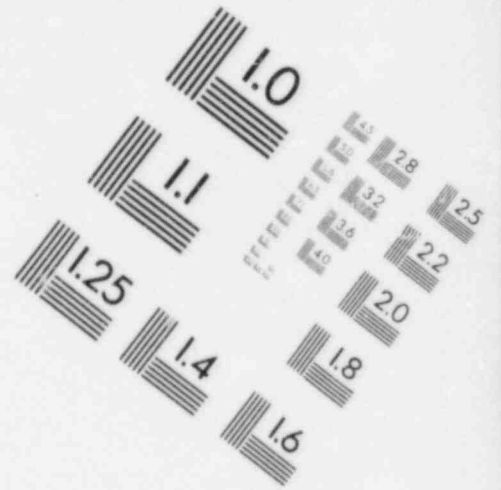
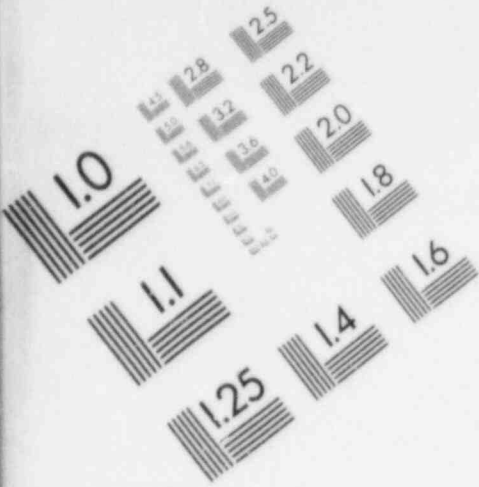


Table 6. Distribution of radioactive tracers from irradiated Zircaloy in test C-4

Test component or collector	Approximate temperature during test (°C)	Tracer (μCi)				
		<sup>51</sup> Cr	<sup>60</sup> Co	<sup>95</sup> Zr	<sup>113</sup> Sn	<sup>125</sup> Sb
Source strips (pretest)		1.253+2 <sup>a</sup>	4.86-2	4.204+2	1.378+0	2.58-1
Posttest data:						
Fuel specimen and ZrO <sub>2</sub> boat	1700	8.398+1	3.215-2	3.742+2	5.123-1	
ZrO <sub>2</sub> furnace tube	1000-1700	1.124+1	1.300-2	1.495+0	4.52-3	
ZrO <sub>2</sub> sliding plug	1700	6.46-1	1.28-3	3.69-2	3.47-3	
ZrO <sub>2</sub> plug right	1000-1400	1.801+0	2.36-3	5.30-2	1.482-2	4.09-3
ZrO <sub>2</sub> insulator right	600-1200		2.5-5	6.0-4	1.501-2	2.25-3
Total in furnace		9.767+1	4.882-2	3.758+2	5.501-1	6.34-3
Thermal gradient tube	200-860	9.87-2	2.13-3	1.20-2	5.274-1	8.86-2
Filter inlet		7.22-2	6.1-5	4.50-3	1.175-2	1.91-3
Glass wool prefilter		1.002-1	3.9-4	4.40-3	1.163-1	1.892-2
HEPA filters (2)		4.91-3		3.6-4	5.72-3	1.48-3
Total on filters		1.773-1	4.51-4	9.36-3	1.338-1	2.23-2
Total released from furnace		2.760-1	2.58-3	3.34-2	6.612-1	1.11-1
Percent of original released from furnace		0.220	5.31	0.0079	47.98	43.0
Percent of original remaining in furnace		77.95	100.5	89.39	39.92	2.46
Percent of original recovered		78.17	105.8	89.40	87.90	45.5

<sup>a</sup>Data in exponential notation: 1.253+2 = 1.253 × 10<sup>+2</sup>, 4.86-2 = 4.86 × 10<sup>-2</sup>.

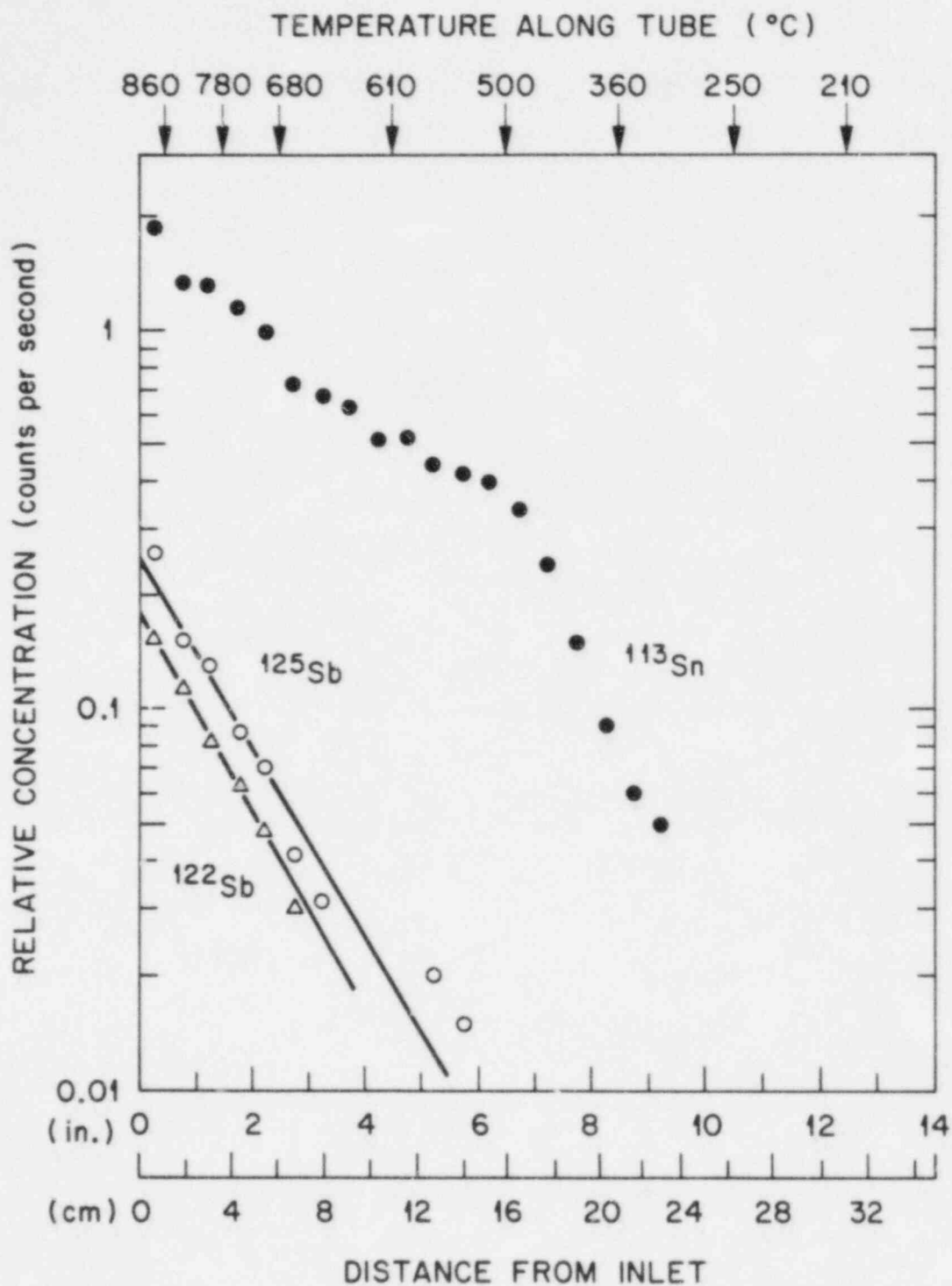


Fig. 24. Distribution of tin and antimony in test C-4 thermal gradient tube.

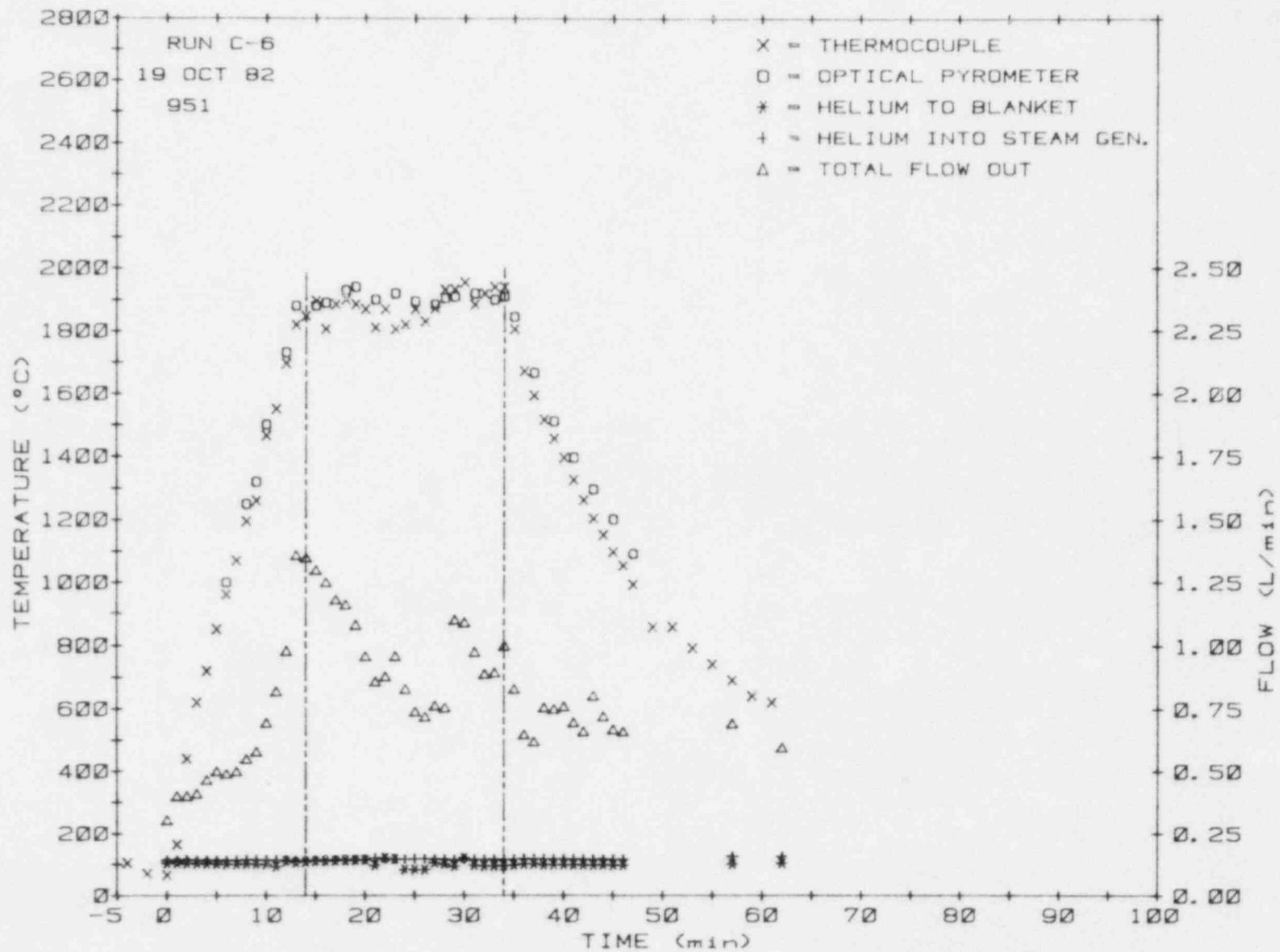


Fig. 25. Temperature and flow histories for test C-6.



ORNL-PHOTO 4379-82

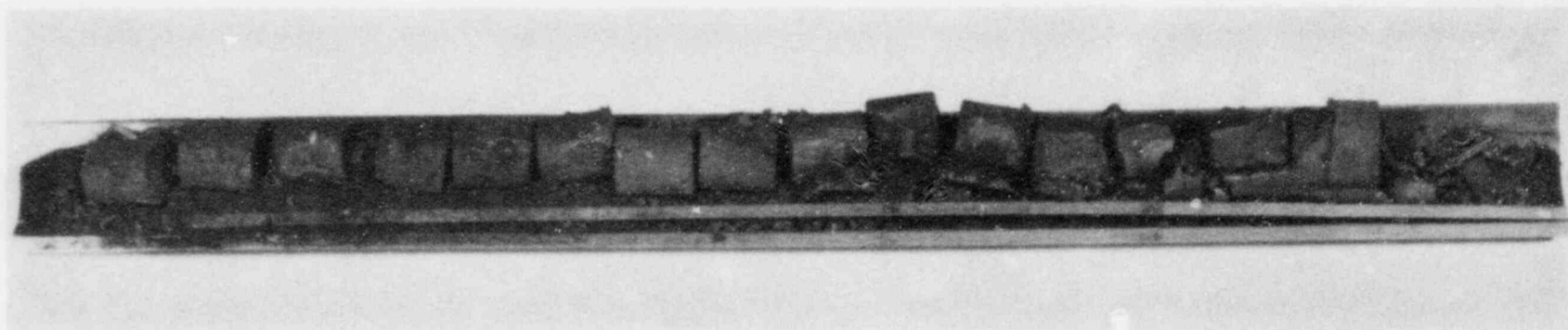


Fig. 26. Appearance of the fuel specimen from test C-6.

fuel pellets. The lower sides of the pellets had been damaged, apparently the result of attack (partial dissolution) by liquid zirconium.

Relatively low radioactivity levels of the tracers made measurement difficult. The distribution of  $^{51}\text{Cr}$ ,  $^{60}\text{Co}$ ,  $^{95}\text{Zr}$ ,  $^{113}\text{Sn}$ , and  $^{125}\text{Sb}$  in the test apparatus are summarized in Table 7. The cladding source strips used in tests C-4 and C-6 were irradiated immediately prior to test C-4. The pretest tracer concentrations shown in Tables 6 and 7 were obtained by analyses performed immediately after irradiation; all posttest analyses for both tests were decay-corrected to the initial analysis date. Because of the decay time in test C-6 (146 d), relatively poor counting precision was obtained for some of the tracer nuclides, compared with test C-4. Only  $^{113}\text{Sn}$  was released from the furnace in significant amount; as indicated in test C-4, appreciable quantities of  $^{125}\text{Sb}$  probably were released but not detected. Because of the very low levels of radioactivity, it was not possible to measure the distribution along the TGT with reasonable precision. However, most of the  $^{113}\text{Sn}$  was located near the inlet (high temperature) end, as in test C-4 (Fig. 24).

#### 5.5 TESTS C-7 AND C-8

These recently conducted tests were designed to compare the behavior of CsI, CsOH, and Te in platinum and stainless steel TGTs. A smaller, furnace, heated by electrical resistance, was substituted for the induction furnace, and the maximum furnace temperature was only 1000°C. Four tracer nuclides were used:  $^{130}\text{I}$  (as CsI),  $^{134}\text{Cs}$  (as CsI),  $^{137}\text{Cs}$  (as CsOH), and  $^{129\text{m}}\text{Te}$  (as metallic Te). Because these two tests are being reported in detail separately,<sup>16</sup> and because the furnace and test procedures were quite different from tests of fuel specimens, no further discussion will be included in this report.

#### 6. REFERENCES

1. 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.
2. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, O. L. Kirkland, and R. L. Towns, Fission Product Release from Highly Irradiated LWR Fuel, NUREG/CR-0772 (ORNL/NUREG/TM-287/R2), February 1980.
3. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, M. F. Osborne, and R. L. Towns, Fission Product Release from Highly Irradiated LWR Fuel Heated to 1300-1600°C in Steam, NUREG/CR-1386 (ORNL/NUREG/TM-346), November 1980.
4. R. A. Lorenz, J. L. Collins, M. F. Osborne, R. L. Towns, and A. P. Malinauskas, Fission Product Release from BWR Fuel Under LOCA Conditions, NUREG/CR-1773 (ORNL/NUREG/TM-338), July 1981.

Table 7. Distribution of radioactive tracers (from irradiated Zircaloy) in test C-6

Test component or collector	Approximate temperature during test (°C)	Tracer (μCi)				
		<sup>51</sup> Cr	<sup>60</sup> Co	<sup>95</sup> Zr	<sup>113</sup> Sn	<sup>125</sup> Sb
Source strips (pretest)		1.192+2 <sup>a</sup>	7.147-2	4.115+2	1.512+0	2.460-1
Posttest data:						
Fuel specimen and ZrO <sub>2</sub> boat	2100	3.428+1	2.663-2	1.751+2	5.121-1	
Debris from furnace <sup>b</sup>	2100	5.297+1	2.796-2	1.613+2	5.337-1	
Total in furnace		8.725+1	5.459-2	3.364+2	1.046-0	
Thermal gradient tube	175-760				1.357-1	
Filter entrance tube and cone	125			1.253-3	1.172-3	
Glass wool prefilter	125	3.435-2		3.399-2	9.668-3	
HEPA filters (2)	125					
Total in filter package		3.435-2		3.524-2	1.084-2	
Total released from furnace		3.435-2		3.524-2	1.465-1	
Percent of original released from furnace		0.0288	0.0	0.0086	9.69	0.0
Percent of original remaining in furnace <sup>c</sup>		73.20	76.88	81.75	69.18	0.0
Percent of original recovered		73.23	76.88	81.76	78.87	0.0

<sup>a</sup>Data in exponential notation: 1.192+2 = 1.192 × 10<sup>+2</sup>, 7.148-2 = 7.148 × 10<sup>-2</sup>.

<sup>b</sup>Debris included particles of oxidized Zircaloy cladding and source strips and UO<sub>2</sub> fuel that fell off boat.

<sup>c</sup>Since several furnace components were not analyzed, these values are known to be low.

5. M. F. Osborne and G. W. Parker, The Effect of Irradiation on the Failure of Zircaloy-Clad Fuel Rods, ORNL-TM-3626, January 1972.
6. H. Albrecht, M. F. Osborne, and H. Wild, "Experimental Determination of Fission and Activation Product Release During Core Meltdown," Proceedings of Thermal Reactor Safety Meeting, Sun Valley, Idaho, Aug. 1-4, 1977.
7. 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-1918 (ORNL/TM-8500), December 1982.
8. 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.
9. 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-3355 (ORNL/TM-8793), March 1984.
10. R. Honnell, Los Alamos National Laboratory, personal communication, to M. F. Osborne, 1983.
11. W. H. McCulla, Oak Ridge Gaseous Diffusion Plant, personal communication, to M. F. Osborne, August 1982 and July 1983.
12. W. H. McCulla, G. E. Nelson, and R. A. Lorenz, "Applications of Laser Spectroscopy for Species Determination in Fission Product Release Experiments," presented at the 26th Oak Ridge National Laboratory Conference on Analytical Chemistry in Energy Technology, Oct. 11-13, 1983, Knoxville, Tenn.
13. P. Hofmann, D. Kerwin-Peck, and P. Nikolopolous, Physical and Chemical Phenomena Associated with the Dissolution of Solid UO<sub>2</sub> by Molten Zircaloy-4, Proceedings of the Sixth International Conference on Zirconium in the Nuclear Industry, Vancouver, British Columbia, Canada, June 28-July 1, 1982.
14. R. A. Lorenz, M. F. Osborne, J. L. Collins, and S. R. Manning, Behavior of Iodine, Methyl Iodide, Cesium Oxide, and Cesium Iodide in Steam and Argon, ORNL/NUREG/TM-25, July 1976.
15. R. C. Weast, ed., Handbook of Chemistry and Physics, The Chemical Rubber Co., 1966.
16. J. L. Collins, R. A. Lorenz, K. S. Norwood, and M. F. Osborne, Behavior of the Fission Product Species CsI, CsOH, and Te in Platinum and Stainless Steel Thermal Gradient Tubes, in preparation.

## APPENDIX. COMMERCIALY SUPPLIED EQUIPMENT AND INSTRUMENTATION

1. Electrical power supply for furnace:
  - RF generator, Model No. T-50-3RS, 50 kW input, 300 kHz, Lepel Corp.
2. Temperature measurement, control, and recording:
  - a. Two-color optical pyrometer, Model No. R-99C05, 1000° to 2600°C, Ircon, Inc.
  - b. Digital indicator, Model 2168A, Omega Engineering, Inc.
  - c. Two-pen strip chart recorder, Model No. 7100B, Hewlett-Packard, Inc.
  - d. Twelve point strip chart recorder, for thermal gradient tube and filter package, Brown Electronik, 0 to 1200°C (Type K thermocouples), Minneapolis Honeywell, Inc.
  - e. Twelve-point strip-chart recorder, for steam supply system, Brown Electronik, 0 to 350°C (Type K thermocouples), Minneapolis Honeywell, Inc.
  - f. Temperature controllers, for thermal gradient tube, four each, Model No. 527Z, 0 to 999°C, Barber Coleman Co.
  - g. Temperature controller, for steam generator, Series 520 digital controller, Barber Coleman Co.
  - h. Thermocouples, for furnace, Pt vs Pt-10% Rh and/or W-5% Re vs W-26% Re, Omega Engineering, Inc.
  - i. Thermocouples, other apparatus locations, Chromel-Alumel (Type K), sheathed with stainless steel, Omega Engineering, Inc.
3. Measurement and recording of gas flow:
  - a. Mass flowmeters, for helium inlet flow, 0 to 500 cc/min, Teledyne Hastings-Raydist.
  - b. Mass flowmeter, for helium outlet flow, 0 to 5.0 L/min with Totalizer, Model ALL-5K, Teledyne Hastings-Raydist.
  - c. Four-pen strip-chart recorder, for recording signals from mass flowmeters, Model FLO4W6D, Texas Instruments, Inc.



## 4. Filtration materials:

- a. Filters, borosilicate glass fiber, MSF Nos. GA-100 and GA-200, Micro Filtration Systems, Inc.
- b. Charcoal, impregnated with triethylenediamine (TEDA), Type NAC-G-618, North American Carbon, Inc.

## 5. Radiation detection and analysis:

- a. NaI(Tl) detectors, Type 125, Harshaw Chemical Co.
- b. Ge(Li) detectors, Model 8101, EG&G ORTEC, Inc.
- c. Multichannel analyzer/computer, System 682, Nuclear Data, Inc.

## INTERNAL DISTRIBUTION

- |        |                   |        |   |
|--------|-------------------|--------|---|
| 1.     | R. E. Adams       | 27.    | M. G. Stewart   |
| 2.     | C. W. Alexander   | 28.    | O. K. Tallent   |
| 3-7.   | J. L. Collins     | 29-30. | J. R. Travis  |
| 8.     | R. W. Glass       | 31-32. | C. S. Webster   |
| 9.     | J. H. Goode       | 33.    | S. K. Whatley   |
| 10.    | J. R. Hightower   | 34.    | R. P. Wichner   |
| 11.    | E. K. Johnson     | 35.    | A. L. Wright  |
| 12.    | T. S. Kress       | 36.    | R. G. Wymer   |
| 13.    | C. E. Lamb        | 37.    | Central Research Library                                  |
| 14-17. | R. A. Lorenz      | 38.    | ORNL-Y-12 Technical Library<br>Document Reference Section |
| 18.    | A. P. Malinauskas | 39.    | Laboratory Records  |
| 19-23. | M. F. Osborne     | 40.    | Laboratory Records, ORNL-RC                               |
| 24.    | G. W. Parker      | 41.    | ORNL Patent Section                                       |
| 25.    | D. R. Reichle     |        |   |
| 26.    | R. D. Spence      |        |   |

## EXTERNAL DISTRIBUTION

42. Office of Assistant Manager for Energy Research and Development, ORO-DOE, P.O. Box E, Oak Ridge, TN 37831
- 43-44. Director, Division of Reactor Safety Research, U.S. Nuclear Regulatory Commission, Washington, DC 20555
- 45-46. Technical Information Center, DOE, Oak Ridge, TN 37831
47. Division of Technical Information and Document Control, U.S. Nuclear Regulatory Commission, Washington, DC 20555
48. L. K. Chan, Fuel Systems Research Branch, Division of Accident Evaluation, U.S. Nuclear Regulatory Commission, Washington, DC 20555
49. M. Jankowski, Fuel Systems Research Branch, Division of Accident Evaluation, U.S. Nuclear Regulatory Commission, Washington, DC 20555
50. R. V. Strain, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439
- 51-325. Given distribution as shown in Category R3 (NTIS - 10)

<b>NRC FORM 335</b> <small>(11-81)</small>		<b>U.S. NUCLEAR REGULATORY COMMISSION</b> <b>BIBLIOGRAPHIC DATA SHEET</b>		<b>1. REPORT NUMBER (Assigned by DDC)</b> NUREG/CR-3715 ORNL/TM-9135	
<b>4. TITLE AND SUBTITLE (Add Volume No., if appropriate)</b> Design, Construction, and Testing of a 2000°C Furnace and Fission Product Collection System				<b>2. (Leave blank)</b>	
<b>7. AUTHOR(S)</b> M. F. Osborne, J. L. Collins, R. A. Lorenz, J. R. Travis, and C. S. Webster				<b>3. RECIPIENT'S ACCESSION NO.</b>	
<b>9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)</b> Chemical Technology Division Oak Ridge National Laboratory P.O. Box X Oak Ridge, TN 37831				<b>5. DATE REPORT COMPLETED</b> MONTH   YEAR March   1984	
<b>12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)</b> Fuel Systems Research Branch U.S. Nuclear Regulatory Commission 7915 Eastern Avenue Mail Stop 1130 S.S. Silver Spring, MD 20910				<b>DATE REPORT ISSUED</b> MONTH   YEAR July   1984	
<b>13. TYPE OF REPORT</b> NUREG/CR and ORNL/TM				<b>PERIOD COVERED (Inclusive dates)</b> (NA)	
<b>15. SUPPLEMENTARY NOTES</b>				<b>14. (Leave blank)</b>	
<b>16. ABSTRACT (200 words or less)</b> <p>An induction furnace, capable of operation at 2000°C in steam, was developed to conduct product release tests. The test specimen and steam atmosphere are contained in a stabilized ZrO<sub>2</sub> furnace tube, which is heated by a concentric susceptor of either tungsten or graphite. A two-color optical pyrometer and high-temperature thermocouples are used for temperature measurement. The furnace has operated reliably for periods up to 30 min at test temperatures of 1400 to 2000°C with steam flowing at ~1 L/min.</p> <p>The apparatus for collecting the released fission products includes a TGT, an aerosol deposition sampler, a series of glass fiber filters, and heated charcoal. A steam condenser and cooled charcoal, for inert gas adsorption, are located further downstream.</p> <p>The principal analytical techniques used for fission product identification and measurement are (1) gamma spectrometry, for all radionuclides on all test components; (2) spark-source mass spectrometry, for all elements, primarily deposits on the thermal gradient tube and filters; and (3) neutron activation, for iodine on selected test components.</p>					
<b>17. KEY WORDS AND DOCUMENT ANALYSIS</b> Fission product release Fission product collection Gamma ray spectrometry LWR fuel damage			<b>17a. DESCRIPTORS</b>		
<b>17b. IDENTIFIERS OPEN ENDED TERMS</b>					
<b>18. AVAILABILITY STATEMENT</b>			<b>19. SECURITY CLASS (This report)</b> Unclassified		<b>21. NO. OF PAGES</b> 5
			<b>20. SECURITY CLASS (This page)</b> Unclassified		<b>22. PRICE</b> \$

120555078877 1 IANR3  
US NRC  
ADM-DIV OF TIDC  
POLICY & PUB MGT BR-PDR NUREG  
W-501  
WASHINGTON DC 20555