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## Fission Product Release from Highly Irradiated LWR Fuel Heated to 1300-1600° C in Steam

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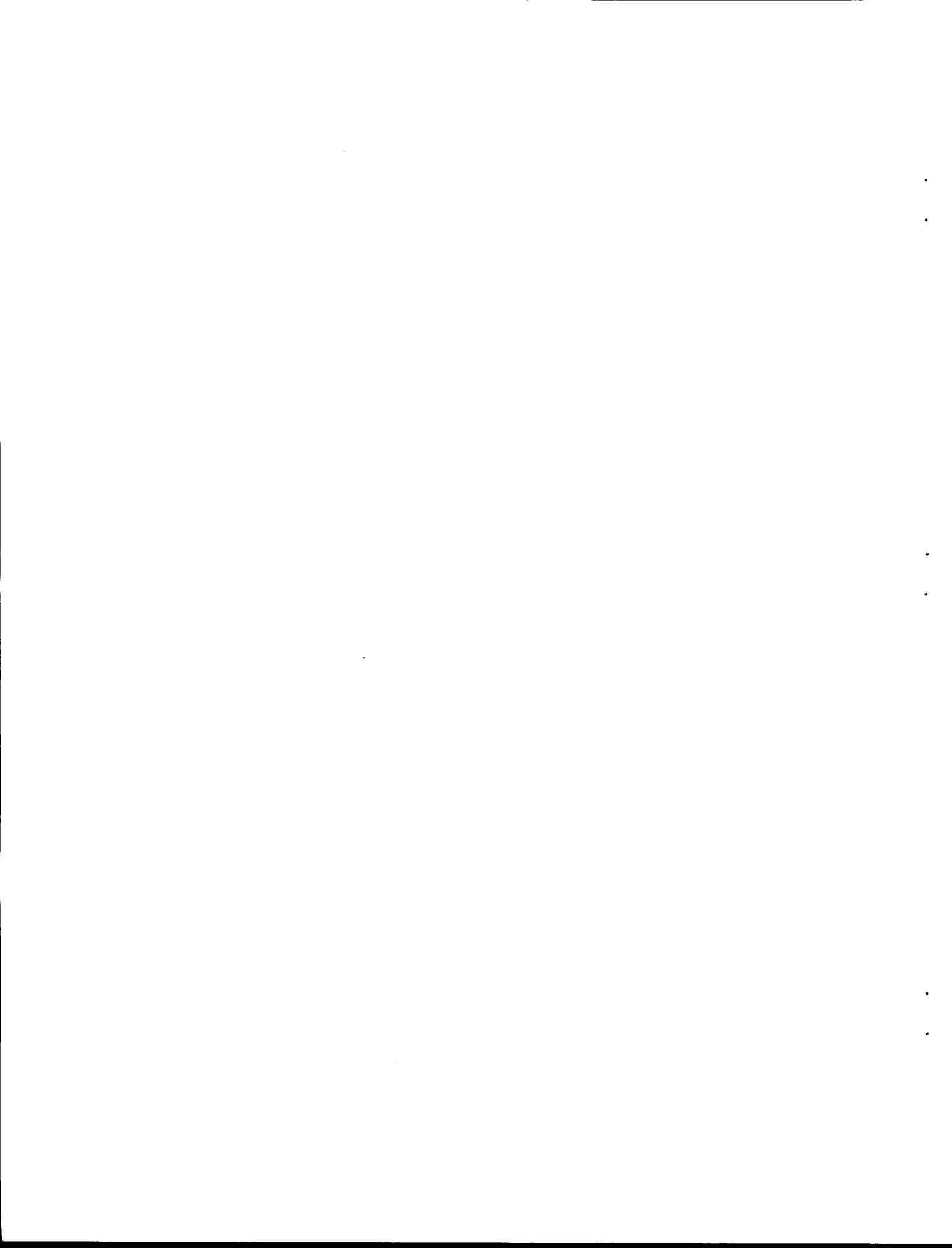
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HEATED TO 1300-1600°C IN STEAM

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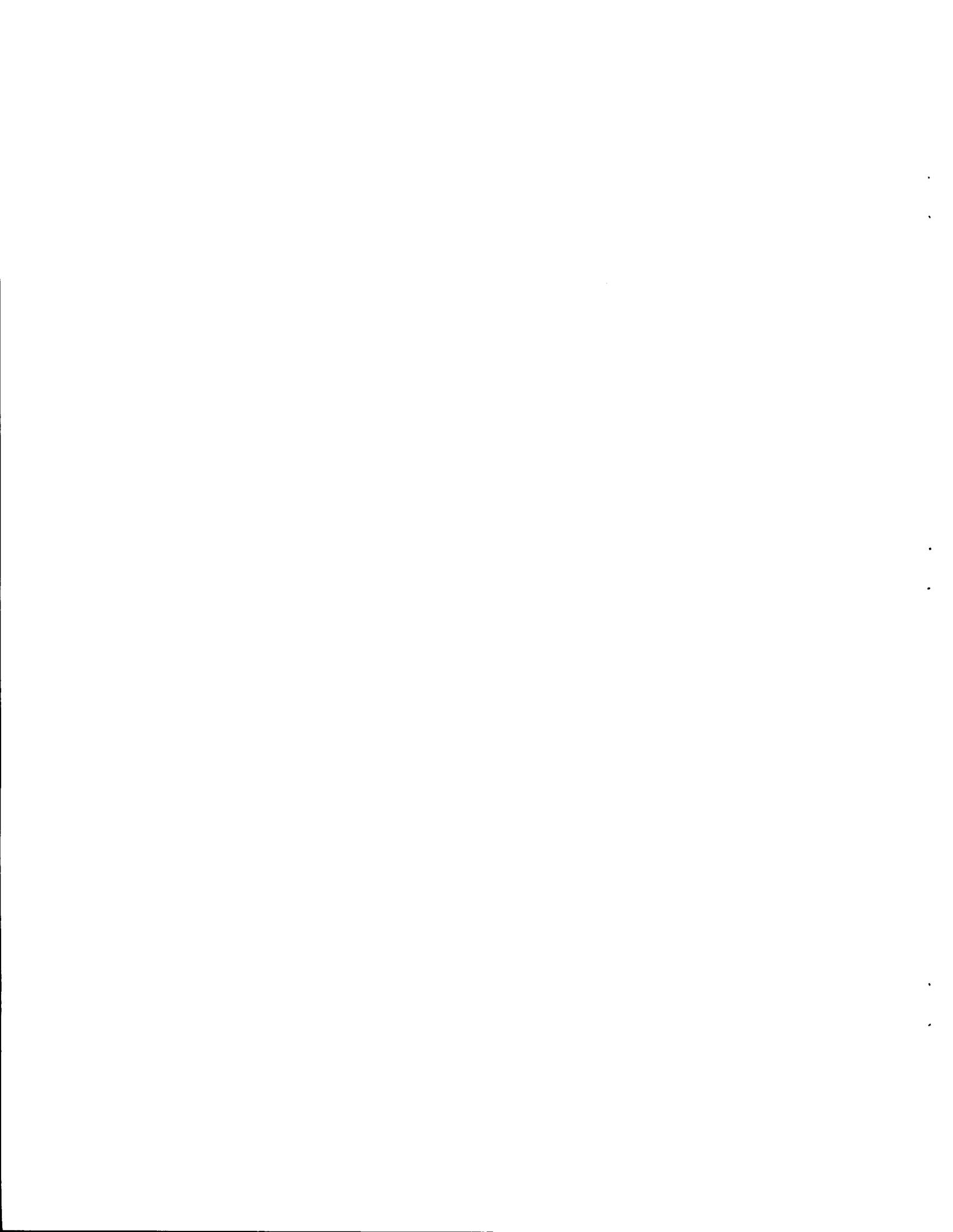
## FOREWORD

This document is one of a series of reports on studies of fission product release from LWR fuel. Other reports in this series are identified below.

1. Quarterly Progress Report on Reactor Safety Programs Sponsored by the Division of Reactor Safety Research for July-September 1974, ORNL/TM-4729, Vol. 1 (December 1974).
2. Quarterly Progress Report on Reactor Safety Programs Sponsored by the NRC Division of Reactor Safety Research for October-December 1974, ORNL/TM-4805, Vol. 1 (April 1975).
3. Quarterly Progress Report on Reactor Safety Programs Sponsored by the NRC Division of Reactor Safety Research for January-March 1975, ORNL/TM-4912, Vol. 1 (July 1975).
4. Quarterly Progress Report on Reactor Safety Programs Sponsored by the NRC Division of Reactor Safety Research for April-June 1975, ORNL/TM-5021 (September 1975).
5. A. P. Malinauskas, R. A. Lorenz, M. F. Osborne, J. L. Collins, and S. R. Manning, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period July-September 1975, ORNL/TM-5143 (November 1975).
6. R. A. Lorenz, J. L. Collins, and S. R. Manning, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period October-December 1975, ORNL/TM-5290 (March 1976).
7. J. L. Collins, M. F. Osborne, A. P. Malinauskas, R. A. Lorenz, and S. R. Manning, Knudsen Cell-Mass Spectrometer Studies of Cesium-Urania Interactions, ORNL/NUREG/TM-24 (June 1976).
8. R. A. Lorenz, M. F. Osborne, J. L. Collins, S. R. Manning, and A. P. Malinauskas, Behavior of Iodine, Methyl Iodide, Cesium Oxide, and Cesium Iodide in Steam and Argon, ORNL/NUREG/TM-25 (July 1976).
9. R. A. Lorenz, J. L. Collins, S. R. Manning, and A. P. Malinauskas, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period January-March 1976, ORNL/NUREG/TM-30 (July 1976).
10. R. A. Lorenz, J. L. Collins, S. R. Manning, O. L. Kirkland, and A. P. Malinauskas, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period April-June 1976, ORNL/NUREG/TM-44 (August 1976).
11. R. A. Lorenz, J. L. Collins, and O. L. Kirkland, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period July-September 1976, ORNL/NUREG/TM-73 (December 1976).

12. R. A. Lorenz, J. L. Collins, and O. L. Kirkland, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period October-December 1976, ORNL/NUREG/TM-88 (March 1977).
13. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1977, ORNL/NUREG/TM-122 (June 1977).
14. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April-June 1977, ORNL/NUREG/TM-139 (September 1977).
15. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period July-September 1977, ORNL/NUREG/TM-170 (January 1978).
16. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period October-December 1977, ORNL/NUREG/TM-186 (March 1978).
17. R. A. Lorenz, J. L. Collins, and A. P. Malinauskas, Fission Product Source Terms for the LWR Loss-of-Coolant Accident: Summary Report, NUREG/CR-0091 (ORNL/NUREG/TM-206) (June 1978).
18. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1978, NUREG/CR-0116 (ORNL/NUREG/TM-208) (June 1978).
19. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April-June 1978, NUREG/CR-0370 (ORNL/NUREG/TM-242) (September 1978).
20. R. A. Lorenz, J. L. Collins, and S. R. Manning, Fission Product Release from Simulated LWR Fuel, NUREG/CR-0274 (ORNL/NUREG/TM-154) (October 1978).
21. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period July-September 1978, NUREG/CR-0493 (ORNL/NUREG/TM-280) (December 1978).
22. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period October-December 1978, NUREG/CR-0682 (ORNL/NUREG/TM-308) (April 1979).
23. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1979, NUREG/CR-0917 (ORNL/NUREG/TM-332) (August 1979).
24. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April-June 1979, NUREG/CR-1061 (ORNL/NUREG/TM-348) (October 1979).

25. R. A. Lorenz, J. L. Collins, A. P. Malinauskas, O. L. Kirkland, R. L. Towns, Fission Product Release from Highly Irradiated LWR Fuel, NUREG/CR-0722 (ORNL/NUREG/TM-287R2) (February 1980).
26. R. A. Lorenz, J. L. Collins, and A. P. Malinauskas, Fission Product Source Terms for the LWR Loss-of-Coolant Accident, NUREG/CR/1288 (ORNL/NUREG/TM-321) (August 1980).



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ABSTRACT

Four tests were performed with high-burnup light water reactor (LWR) fuel to explore the amount and characteristics of fission product release at short heating times (0.4 to 10 min) in steam atmosphere in the temperature range 1300 to 1600°C. The test fuel rod segments were cut from full-length fuel rods irradiated at low heat rating to 30,000 MWd/MT in the H. B. Robinson-2 reactor.

The releases of cesium and iodine increased tenfold ( $\sim 0.3$  to  $>4\%$ ) with temperature from 1350 to 1400°C from fuel with defects that simulate ruptured cladding. Krypton release rose from  $\sim 2$  to  $\sim 11\%$  of total inventory in this temperature range. This sudden increase in release of krypton, cesium, and iodine is believed to result from prior accumulations of these species at or very near the grain boundaries. At 1600°C, the releases of krypton, cesium, and iodine were in the range 17 to 25% of total fuel inventory.

---

1. INTRODUCTION

Several series of tests have been performed to characterize the fission product release that could occur in the steam atmosphere of a controlled loss-of-coolant accident (LOCA) in a light water reactor (LWR). The controlled LOCA is one in which the emergency core-cooling devices operate sufficiently well so that the maximum cladding temperature is restricted to  $\sim 1200^\circ\text{C}$ . The High Burnup Test Series<sup>1</sup> was performed in the temperature range 500 to 1200°C with fuel from the H. B. Robinson-2 pressurized water reactor (PWR) irradiated to  $\sim 30,000$  MWd/MT. These tests were preceded by the Implant Test Series,<sup>2</sup> which used Zircaloy-clad

fuel rod segments containing unirradiated  $UO_2$  pellets coated with simulated fission products tested in the same temperature range. Both of these series included two experiments performed in dry air. A Control Test Series<sup>3</sup> provided experience with individual species introduced directly into the test apparatus. Studies of cesium-urania reactions were conducted separately.<sup>4</sup> A short series of tests with boiling water reactor (BWR) fuel is in progress<sup>5</sup> which will complete our scheduled experimental work in the controlled LOCA temperature range. A preliminary model for the controlled LOCA fission product release source term has been published.<sup>6</sup>

The High-Temperature Test Series described in this report covers the release of fission products from LWR fuel heated for short times in the temperature range 1300 to 1610°C. This range could be reached if emergency core cooling failed to properly control the LOCA yet prevented meltdown of the core.

The low-frequency (3-kHz) induction heating system used in the High Burnup Test Series was also used in this series of tests. Since low-frequency induction heating will not couple with either oxidized cladding (primarily  $ZrO_2$ ) or  $UO_2$ , the experiments were terminated when sufficient cladding oxidation occurred by the steam-Zircaloy reaction that the test temperature could not be maintained.

## 2. FUEL FOR THE HIGH-TEMPERATURE TEST SERIES

### 2.1 Description of the H. B. Robinson-2 Fuel

As with the High Burnup Test Series, fuel rod segments for the high-temperature fission product release experiments were cut from rods D-12 and H-15 of bundle B05, which operated in the Carolina Power and Light Company's H. B. Robinson-2 power plant from October 1971 to May 6, 1974; the irradiation history has been described by MacDonald.<sup>7</sup> The peak linear heat rating, 32.6 kW/m (9.95 kW/ft), occurred in December 1971 when the burnup reached 5872 MWd/MT and the average heat rating was 23.3 kW/m (7.10 kW/ft). By May 1974 the peak heat rating had decreased to 21.2 kW/m (6.45 kW/ft) and the average to 17.5 kW/m (5.34 kW/ft).

Descriptions of the fuel rods and their arrangement in the bundle have been reported by Perkins.<sup>8</sup> The cladding is Zircaloy-4, 1.072 cm (0.422 in.) diam, with 0.0617-cm (0.0243-in.) wall thickness. The original diametral pellet-to-clad gap was 0.0165 cm (0.0065 in.), the plenum length was 17.35 cm (6.83 in.), and the overall rod length was 387.0 cm (152.36 in.). We calculate from the total core loading that each fuel rod originally contained 2495.4 g of  $UO_2$  or 2199.6 g of uranium if it is assumed that the molecular weight of the enriched uranium was 237.92 g-mole.<sup>1</sup>

The two fuel rods utilized in this experimental series were sectioned by Battelle Columbus Laboratories (BCL) as depicted in Fig. 1 and described in detail elsewhere.<sup>1</sup> Segments identified with the prefix B were cut from rod H-15. The long segments 3 and 8 were recut at Oak Ridge National Laboratory to form additional 30.5-cm (12-in.) segments; segment 3a was the bottom half and 3b the top half, etc.

A scan of gamma activity in rod F-7, which was obtained by Argonne National Laboratory,<sup>9</sup> is also displayed in Fig. 1. Gamma activity distributions of other rods from bundle B05 were determined by BCL; these indicated similar profiles.<sup>10</sup> Both laboratories found fairly regular activity depressions between pellets, thus indicating that the pellets were generally intact and that no significant concentration of fission product cesium or ruthenium occurred during the in-reactor irradiation. The observed axial gamma distributions were very similar to the burnup distribution obtained from in-core flux measurements as reported by MacDonald.<sup>7</sup> Calculated fission product inventories have been published.<sup>1</sup>

## 2.2 Description of Fuel Rod Test Segments

The 30.5-cm (12-in.)-long segments were capped with stainless-steel ferrule fittings; the fitting on the bottom end of the segment contained a 5.1-cm (2-in.)-long tube for pressurization. Details of the cutting procedure and a sketch of the segments have been published.<sup>1</sup>

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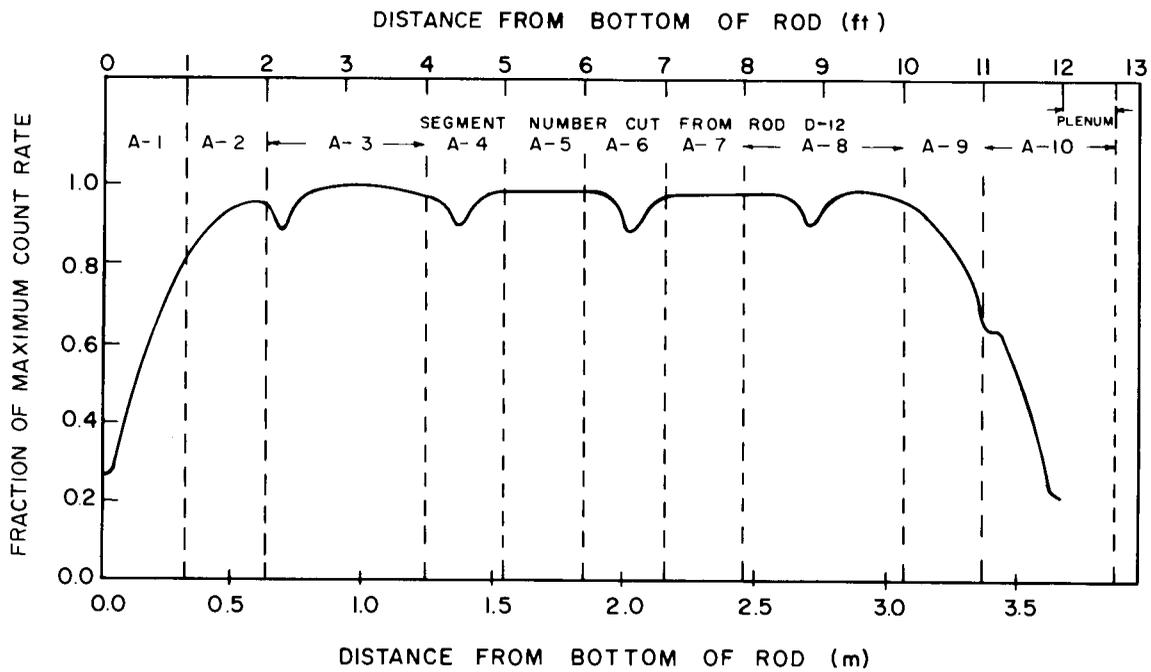


Fig. 1. Identification of fuel rod segments and gamma scan of rod F-7.

### 2.3 Fission Gas Release and Gap Inventory of Cesium and Iodine

Fission gas releases to the plenums and void spaces of rods D-12 and H-15 while operating in the H. B. Robinson-2 reactor were determined to be the following.<sup>1</sup> For rod D-12, the krypton release was 0.29% and the xenon release 0.22%; the rod H-15 results were 0.35% krypton release and 0.25% xenon release. The low fuel temperatures,<sup>1</sup> particularly toward end of life, are believed to be responsible for the relatively low fission gas release values.

Only small amounts, ~0.3% of the total fuel inventory, of cesium and iodine were found in the pellet-to-clad gap space of this fuel.<sup>1</sup> The gap space of segment B-7 was purged with purified helium in test HBU-12 while heated in the temperature range 700 to 1200°C in order to measure the readily vaporized content of cesium and iodine.

## 3. EXPERIMENTAL APPARATUS AND EQUIPMENT

### 3.1 Description of Test Apparatus

The apparatus is the same as that used in the High Burnup Test Series;<sup>1</sup> a schematic diagram is shown in Fig. 2. In each test a high-burnup fuel segment was placed on a quartz\* holder and inserted into a quartz liner that was prepositioned inside a quartz furnace tube. The fuel rod segment being tested was heated to the desired temperature level by a low-frequency induction heater.

Tests with pressure-ruptured Zircaloy cladding revealed that the expanded and thinned cladding oxidized rapidly in steam at high temperatures, causing loss of coupling in the fully oxidized locations. It was therefore decided to use test fuel rod segments with cladding expanded only sufficiently to widen the pellet-to-clad gap space enough to provide a realistic escape path for the fission products without significantly reducing the cladding wall thickness. Expansion to form a 380- $\mu$ m radial gap width combined with a 1.59-mm (1/16-in.)-diam hole drilled through the cladding

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\*Quartz as used in this report means fused silica.

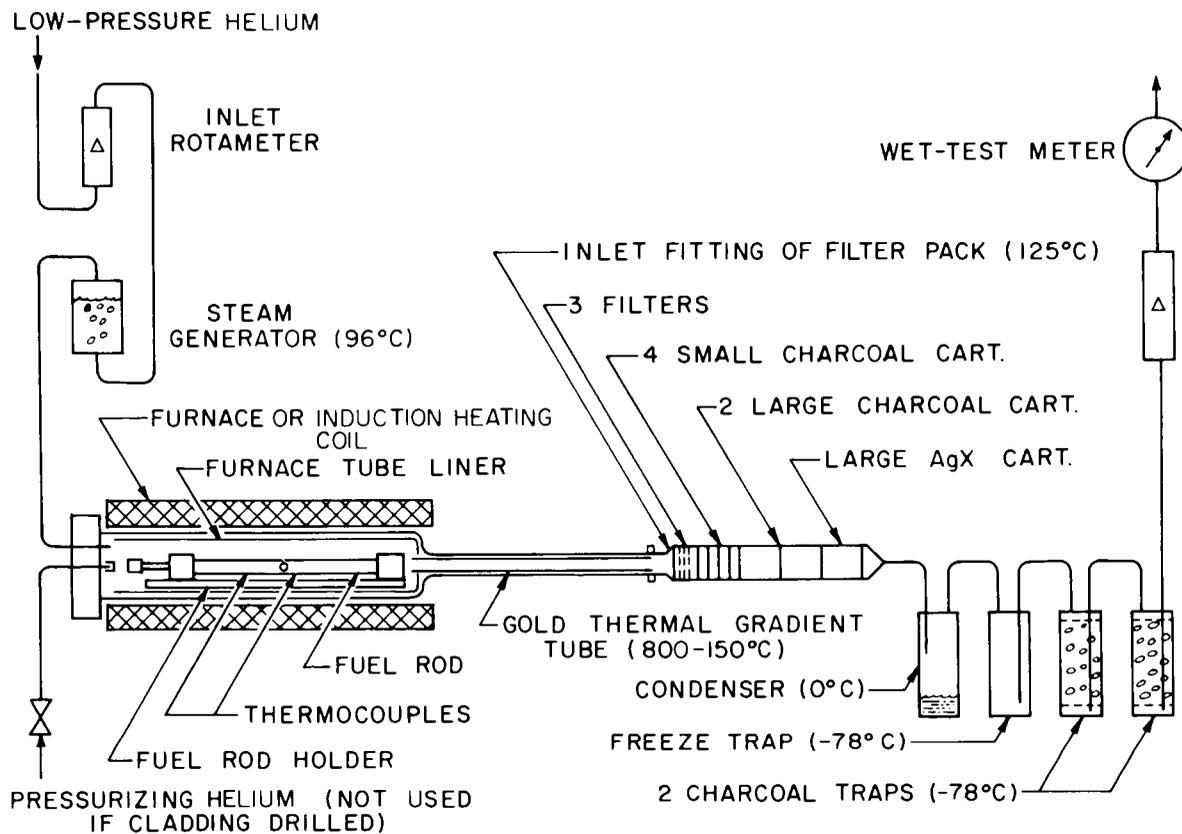


Fig. 2. Fission product release apparatus.

was believed to give the release restriction equivalent to a pressure-rupture failure. Tests with unirradiated fuel rod segments demonstrated that satisfactory heating times at temperatures up to 1600°C in steam could be obtained before loss of inductive coupling.

Material released from the fuel rod that did not deposit on the furnace tube components was carried by a flowing gas mixture of steam and helium through a gold foil-lined thermal gradient tube, where volatile species that are condensible above 150°C deposited. A filter pack containing three high-efficiency particulate aerosol (HEPA) filter papers (Reeve-Angel pure borosilicate fiberglass) was used to collect the particulate material. Elemental iodine and organic iodides were trapped by impregnated charcoal and silver-exchanged zeolite adsorbers. Each of the first four charcoal compartments (1a, 1b, 1c, and 2a) contained a volume of charcoal that would fit into a 0.8-cm<sup>3</sup> "rabbit" used in activation analysis. The other cartridges contained greater amounts. Cartridges 1, 2, and 3 contained 12- to 16-mesh type G-618 charcoal impregnated with triethylenediamine, whereas cartridge 4 contained 12- to 16-mesh 98% silver-exchanged 13X zeolite. An ice-bath condenser, which collected the steam, was followed by a dry ice-cooled freeze trap (-78°C) which reduced the partial pressure of water vapor to  $\sim 10^{-7}$  MPa ( $\sim 10^{-6}$  atm). Further downstream, fission product gases were adsorbed in two charcoal traps also at dry-ice temperature. A wet-test meter was used to measure the effluent gas flow which, in addition to helium, also contained hydrogen that was produced by the reaction of steam with the Zircaloy cladding. A shielded NaI crystal was used to monitor the <sup>134</sup>Cs and <sup>137</sup>Cs deposition in the thermal gradient tube and filter pack. The <sup>85</sup>Kr was similarly monitored as it was collected in the charcoal traps.

Temperature measurements were made with the automatic scanning optical pyrometer that was carefully calibrated as described in Sect. 3.3. This instrument was positioned in the cell on a trolley located outside the apparatus containment box. The trolley movement allowed the axial temperature distribution to be measured whenever the temperature was stabilized, and it allowed the pyrometer to be moved to the side when the center area was needed for fuel handling operations.

### 3.2 Cladding Expansion Procedure

The apparatus for controlled expansion of the cladding is shown in Fig. 3. A split Invar mold was clamped outside the fuel rod. To minimize oxidation of the Invar and the Zircaloy cladding, argon purges were used both inside and outside of the mold. The back-pressure and flow rate measurements of the argon supplied to the cavity between the Zircaloy and mold provided detection of cladding expansion at these locations.

Each rod segment was heated to  $\sim 800^{\circ}\text{C}$  while pressurized internally with purified helium to  $\sim 6.89$  MPa ( $\sim 1000$  psi) to effect the expansion. Pressurizing and venting the pellet-to-clad gap space several times before heating removed most of the trapped air. The expansion profile measured on an unirradiated fuel rod is shown in Fig. 3. Detailed measurements were not performed on the irradiated rods, but a gauge block was used to verify the expansion indicated by back-pressure and flow measurements of the two inner argon purge lines. After cooling, the fuel rod cladding was drilled to form a 0.159-cm (1/16-in.)-diam hole in the manner described in a previous test series.<sup>1,2</sup>

Based on previous experience with this fuel,<sup>1</sup> loss of iodine or cesium during the expansion operation should have been prevented by the leakproof system. Loss of some fission gas was expected because previous experience showed relatively rapid release of surface-embedded gas when this type of fuel was heated to  $800^{\circ}\text{C}$ . Because the maximum temperature and duration of heating differed for each fuel rod, the estimated release of  $^{85}\text{Kr}$  and other fission gases ranged from 0.5 to 1.5% of the original inventory. These estimates are based on experience in the High Burnup Fuel Test Series.<sup>1</sup>

### 3.3 Calibration of the Photoelectric and Optical Pyrometers

The in-cell photoelectric pyrometer (Barnes Engineering Company, Model EP-1) viewed the fuel rod through a Vycor glass window in the containment box and through the quartz furnace tube and furnace tube liner. Calibrations were made with Zircaloy cladding exposed to flowing steam since the emissivity of oxidized Zircaloy is known to be affected by temperature.<sup>11</sup> Seven calibration tests were made using thermocouples inside the cladding

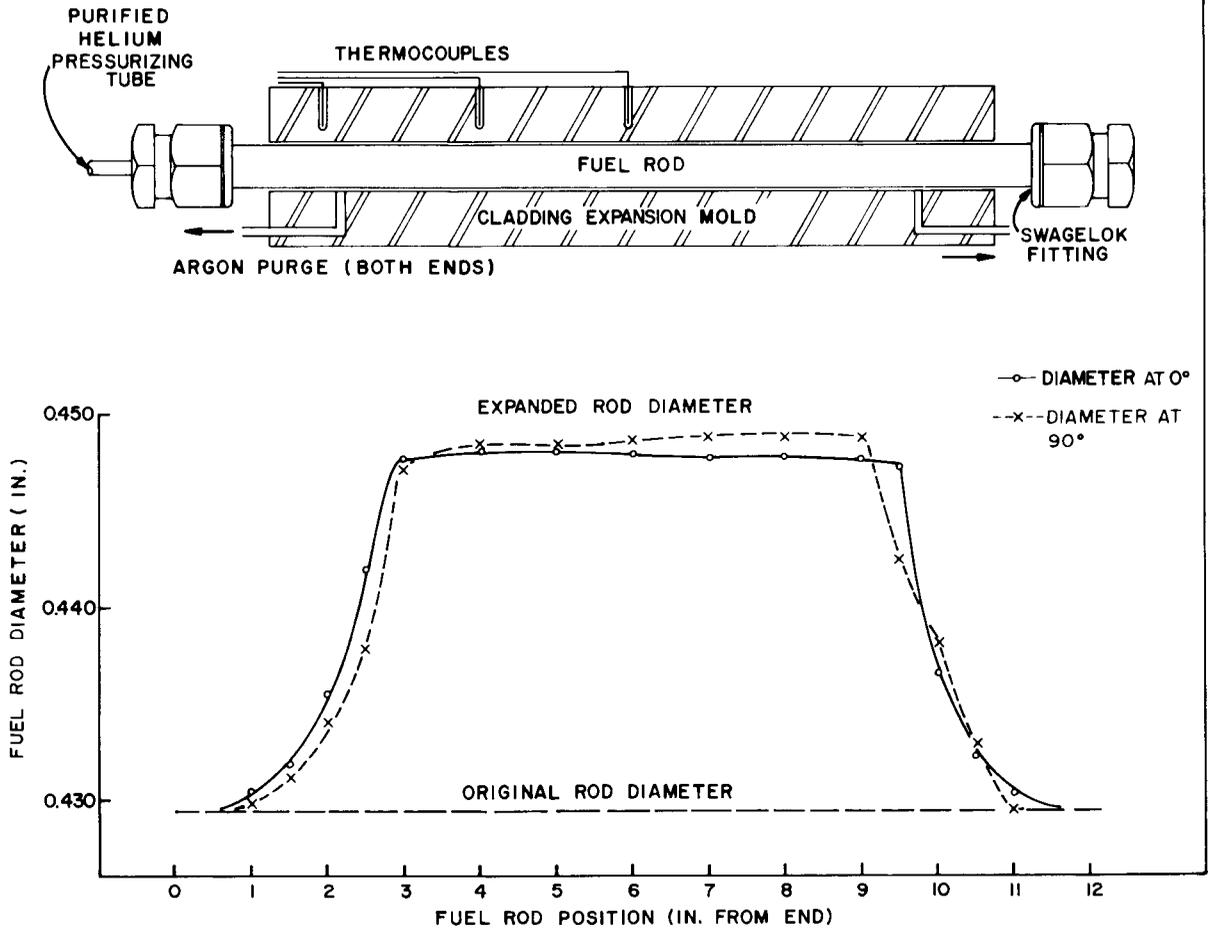


Fig. 3. Dimensional profiles of test fuel rod segment after expansion to simulate fuel-cladding gap in pressure-ruptured fuel rod.

or spot-welded to the outside as temperature references. Chromel-alumel thermocouples sheathed in stainless steel were used at lower temperatures. At higher temperatures platinum, platinum-10% rhodium thermocouples were used, usually spot-welded to thin tantalum or iridium pads to prevent platinum-zirconium eutectic formation. The short lifetime of the cladding and thermocouple installations in the high-temperature steam made it necessary to make many test runs, each with new cladding and thermocouples.

A manually operated Leeds & Northrup disappearing-filament optical pyrometer was calibrated at the same time from outside the hot cell; it viewed through ~91 cm (3 ft) of lead glass. The resulting calibrations for both pyrometers are given in Table 1. The accuracy of the calibrations is uncertain. Judging by the scatter in calibration results, we believe that the reported measurements at 1300 to 1600°C are within 40°C. Axial temperature variations resulting from the induction heating characteristics and nonuniform oxidation add to the difficulty in pinpointing test temperatures.

The thin layer of crud on the H. B. Robinson fuel rods may have had a small effect on the temperature readings since our calibrations were made with unused cladding. Burgoyne and Garlick<sup>12</sup> report that cladding with thick crud has a total emissivity of ~0.9 compared with ~0.8 for steam-oxidized unused Zircaloy.

#### 3.4 Radiochemical Procedures

The apparatus was disassembled, and individual components were counted for gamma emissions using a calibrated Ge(Li) detector. Corrections were made for adsorption in component containers. The most radioactive components were held in a specially constructed shield with calibrated removable flat lead plates [1.27 cm (0.5 in.) thick] for attenuation while counting. The principal gamma emitters were <sup>137</sup>Cs and <sup>134</sup>Cs; all cesium counting was performed in this manner. The thermal gradient tube and the furnace tube liner were scanned for the axial distribution of gamma emitters after the total count was obtained.

Following gamma counting, most components were leached with a mixture of ammonium hydroxide and hydrogen peroxide to obtain solutions for activation analysis of <sup>129</sup>I. The gold foil liner in the thermal gradient tube was cut into four or five pieces for <sup>129</sup>I analysis.

Table 1. Optical pyrometer calibration data

Thermocouple temperature (°C)	Temperature deviation (°C) <sup>a</sup>		
	In-cell automatic pyrometer <sup>b</sup>		Out-of-cell disappearing filament pyrometer <sup>c</sup>
	Low range	High range	
800	-30		-80
900	-30		-91
1000	-35		-106
1100	-45		-125
1200	-57		-149
1300	-70		-173
1400	-85		-196
1500	-102	-42	-220
1600		-60	-243

<sup>a</sup>Deviation = pyrometer reading - thermocouple reading.

<sup>b</sup>Photoelectric pyrometer Model EP-1, Barnes Engineering Co.,  
Stanford, Conn.; emissivity setting 0.5.

<sup>c</sup>Serial No. 1506172, catalog No. 8622-C; Leeds and Northrup Co.,  
Philadelphia, Pa.

The first four small charcoal samples were each placed in an irradiation rabbit for direct neutron activation without chemical treatment.

#### 4. RESULTS OF THE HIGH-TEMPERATURE TESTS

A detailed description of each of the high-temperature tests is presented in this section.

##### 4.1 High-Temperature Test 1 ( $\sim 1300^\circ\text{C}$ )

The first test in the high-temperature series, HT-1, was conducted with segment A-3b at about  $1300^\circ\text{C}$  for 10 min in a flowing steam-helium atmosphere. Before testing, the cladding had been expanded to provide a uniform fuel-cladding gap of about  $380\ \mu\text{m}$  (0.015 in.), as discussed previously, and drilled to produce the standard cladding defect — a 0.159-cm (0.0625-in.) hole. Thus the radial gap width and the hole size were similar to those resulting from the pressure-rupturing of cladding in our apparatus.<sup>1</sup>

The operating temperatures and the collections of  $^{85}\text{Kr}$  and  $^{134}\text{Cs}$  during the tests are summarized in Fig. 4. Details of the  $^{134}\text{Cs}$  and  $^{129}\text{I}$  distributions throughout the experimental apparatus are given in Fig. 5 and Tables 2 and 3. Approximately 0.112% (524  $\mu\text{g}$ ) of the total cesium inventory in the 30.5-cm (12-in.)-long rod segment was released; as shown in Fig. 5, the major portion (i.e., 87%) deposited on the quartz surfaces of the furnace tube near the drilled hole, adjacent to the rod segment. Approximately 4.6% was transported to the thermal gradient tube, where the peak concentration of  $^{134}\text{Cs}$  occurred at  $350^\circ\text{C}$ , which is in agreement with previous experimental results at lower fuel temperature.<sup>1</sup>

The amount of iodine released was 0.165% (71.3  $\mu\text{g}$ ) of the total rod inventory. Approximately 70% of it was collected in the thermal gradient tube. The concentration peak was at the same location as the cesium peak.

About 1.07% (37.1  $\mu\text{g}$ ) of the original  $^{85}\text{Kr}$  inventory was collected during the test, and some additional  $^{85}\text{Kr}$  (probably about 0.5%) was released and vented during the cladding expansion operation prior to the high-temperature test.

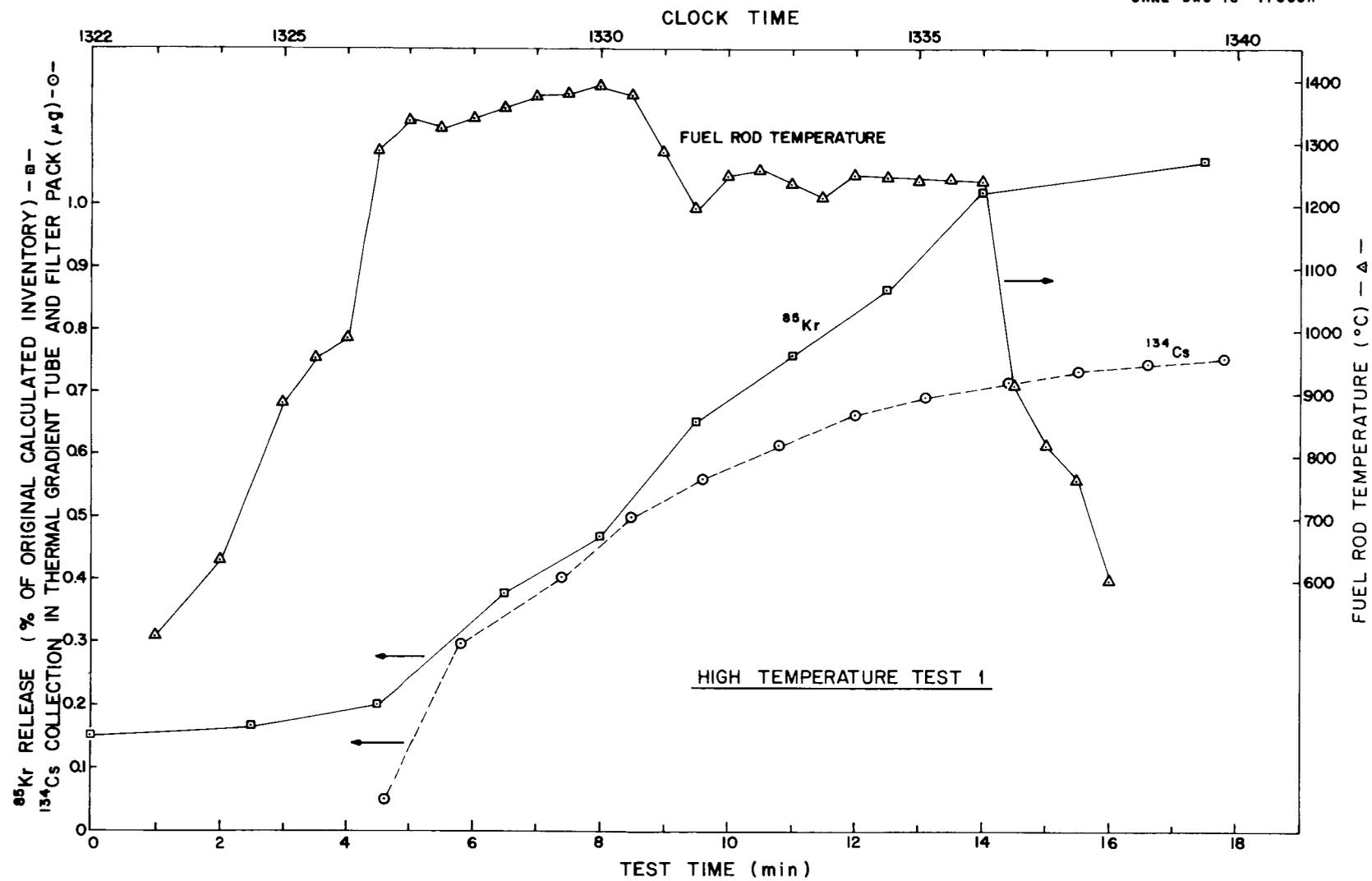


Fig. 4. Collection of <sup>134</sup>Cs in the thermal gradient tube and filter pack, and of <sup>85</sup>Kr in the charcoal traps during test HT-1.

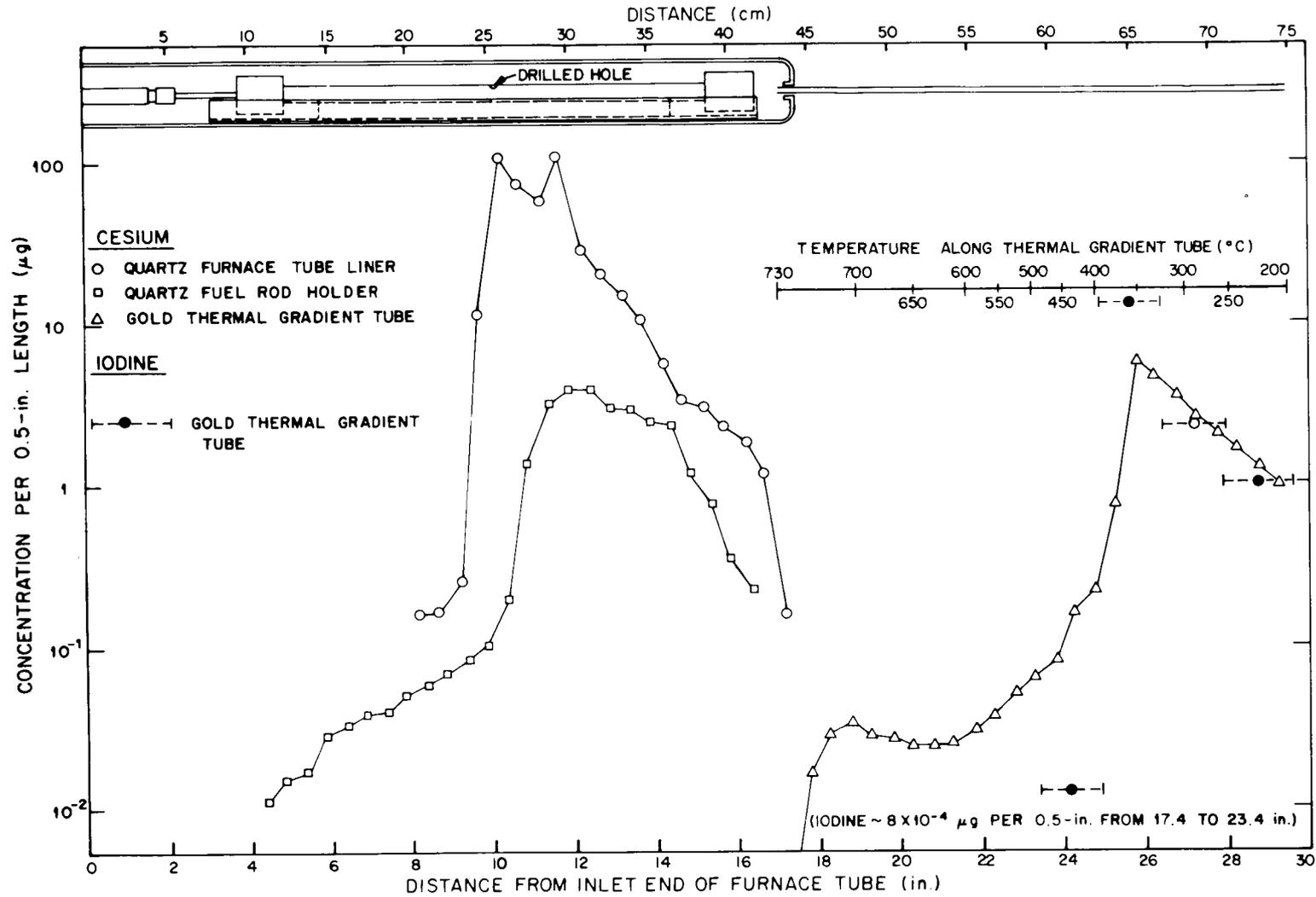


Fig. 5. Concentration profile of cesium and iodine in apparatus components in test HT-1.

Table 2. Distribution of  $^{134}\text{Cs}$  in high-temperature test 1<sup>a</sup>

Location	Temperature (°C)	Amount of $^{134}\text{Cs}$ found in each location			Total cesium found ( $\mu\text{g}$ )
		$\mu\text{g}$ <sup>b</sup>	Percent of total <sup>c</sup>	Percent of released	
Fuel rod segment, initially	~1300	8908 <sup>d</sup>			4.69 x 10 <sup>5d</sup>
Furnace tube					
Quartz liner	e	8.654	9.71 x 10 <sup>-2</sup>	86.96	455.5
Quartz fuel rod holder		0.510	5.73 x 10 <sup>-3</sup>	5.12	26.85
Thermal gradient tube	730-200	0.457	5.13 x 10 <sup>-3</sup>	4.59	24.06
Filter pack components	134				
Stainless steel inlet fitting		0.019	2.13 x 10 <sup>-4</sup>	0.19	1.00
First filter paper		0.312	3.50 x 10 <sup>-3</sup>	3.14	16.42
Second filter paper		5.50 x 10 <sup>-6</sup>	6.17 x 10 <sup>-8</sup>	5.5 x 10 <sup>-5</sup>	2.9 x 10 <sup>-4</sup>
Third filter paper		1.39 x 10 <sup>-6</sup>	1.56 x 10 <sup>-8</sup>	1.4 x 10 <sup>-5</sup>	7.3 x 10 <sup>-5</sup>
Charcoal No. 1a		0.0	0.0	0.0	0.0
Charcoal No. 1b		0.0	0.0	0.0	0.0
Charcoal No. 1c		0.0	0.0	0.0	0.0
Charcoal No. 2a		0.0	0.0	0.0	0.0
Charcoal No. 2b		0.0	0.0	0.0	0.0
AgX (silver-exchanged Zeolite)		0.0	0.0	0.0	0.0
Condenser	0	0.0	0.0	0.0	0.0
Freeze trap		0.0	0.0	0.0	0.0
Cold charcoal traps (two)	-78	0.0	0.0	0.0	0.0
Total	-78	9.952	0.112	100.00	523.9

<sup>a</sup> Steam flow rate, 715 cm<sup>3</sup>/min (STP); helium flow rate, 870 cm<sup>3</sup>/min (STP); pressure, 760 torr. Decay time, 911 days (to Nov. 2, 1976).

<sup>b</sup> Amounts less than 1.0 x 10<sup>-6</sup>  $\mu\text{g}$  are given as 0.0.

<sup>c</sup> Percent of radioactive nuclide in fuel rod segment.

<sup>d</sup> Calculated for burnup of 31,360 MWd/MT of original uranium, 183.3 g of uranium originally in 30.5-cm (12-in.) segment, and 911 days decay.

<sup>e</sup> Approximately 900°C max at center, ~600°C at outlet. Inlet end, 140°C.

Table 3. Distribution of iodine in high-temperature test 1<sup>a</sup>

Location	Temperature (°C)	Amount of <sup>129</sup> I found in each location			Total iodine found (μg)
		μg <sup>b</sup>	Percent of total <sup>c</sup>	Percent of released	
Fuel rod segment, initially	~1300	3.40 x 10 <sup>4d</sup>			4.315 x 10 <sup>4d</sup>
Furnace tube					
Quartz liner	e	<0.03	8.8 x 10 <sup>-5</sup>	0.05	0.04
Quartz fuel rod holder		<0.03	8.8 x 10 <sup>-5</sup>	0.05	0.04
Thermal gradient tube	730-200	39.37 ± 0.36	0.116	70.04	49.97
Filter pack components	134				
Stainless steel inlet fitting		3.58 ± 0.01	1.05 x 10 <sup>-2</sup>	6.37	4.54
First filter paper		9.38 ± 0.08	2.76 x 10 <sup>-2</sup>	16.69	11.90
Second and third filter papers		1.32 ± 0.02	3.88 x 10 <sup>-3</sup>	2.35	1.68
Charcoal No. 1a		2.50 ± 0.10	9.32 x 10 <sup>-3</sup>	4.45	3.17
Charcoal No. 1b		0.0	0.0	0.0	0.0
Charcoal No. 1c		0.0	0.0	0.0	0.0
Charcoal No. 2a		0.0	0.0	0.0	0.0
Charcoal No. 2b <sup>f</sup>		0.0	0.0	0.0	0.0
Charcoal No. 3 <sup>f</sup>		0.0	0.0	0.0	0.0
AgX (silver-exchanged Zeolite) <sup>f</sup>		0.0	0.0	0.0	0.0
Condenser <sup>f</sup>	0	0.0	0.0	0.0	0.0
Freeze trap <sup>f</sup>	-78	0.0	0.0	0.0	0.0
Cold charcoal traps (two) <sup>f</sup>	-78	0.0	0.0	0.0	0.0
Total		56.21 ± 0.56	0.165	100.00	71.32 ± 0.7

<sup>a</sup> Steam flow rate, 715 cm<sup>3</sup>/min (STP); helium flow rate, 870 cm<sup>3</sup>/min (STP); pressure, 760 torr. Decay time, 199 days (to Nov. 2, 1976).

<sup>b</sup> Amounts less than 0.02 μg are given as 0.0.

<sup>c</sup> Percent of radioactive nuclide in fuel rod segment.

<sup>d</sup> Calculated for burnup of 31,360 MWd/MT of original uranium, 183.3 g of uranium originally in 30.5-cm (12-in.) segment, and 911 days decay. Error estimate based solely on spread between duplicate activation samples. Total iodine mass = 1.27 x <sup>129</sup>I mass.

<sup>e</sup> Approximately 900°C max at center, ~600°C at outlet end. Inlet end, 140°C.

<sup>f</sup> Not analyzed for <sup>129</sup>I. Amount assumed to be 0.0.

#### 4.2 High-Temperature Test 2 (1445°C)

The fuel rod specimen (B-3a) in this test (HT-2) was heated by induction at 1445°C for 7 min in a flowing steam-helium atmosphere. Posttest distributions of released  $^{134}\text{Cs}$  and  $^{129}\text{I}$ , as well as total cesium and iodine, are summarized in Tables 4 and 5. As indicated, 4.82% (22.04 mg) of the total cesium and 2.35% (0.99 mg) of the total iodine inventories were released in this test. As was noted previously in test HT-1, most of the released cesium (94.6%) remained in the furnace tube, while the major fraction of the iodine released (87%) was transported downstream and condensed in the thermal gradient tube.

Figure 6 is a graphical display of the axial distribution of cesium and iodine in the thermal gradient tube. Interestingly, almost equal masses of each element were found there. Furthermore, concentration peaks for both elements occurred at about 475°C. These data, in addition to the observation that only 0.06% of the released iodine was found on the charcoal, infer that CsI was the primary iodine species released.

On the other hand, since the mass of cesium released was about 22 times greater than that of iodine, it is clear that most of the cesium was released in a form other than CsI. As mentioned previously, the cesium (other than CsI) was retained by the quartz surfaces of the furnace tube liner and fuel rod holder. (Depending on the oxygen potential, a fraction of the gaseous atoms of cesium exposed to the steam atmosphere of the furnace tube would be expected to form cesium hydroxide,<sup>13</sup> which in turn reacts rapidly with quartz to form less volatile silicates<sup>2,14</sup> at temperatures below 1000°C.)

During the test, collection of  $^{85}\text{Kr}$  in the cooled charcoal traps (-78°C) was continuously monitored; the resulting data are depicted graphically in Fig. 7. [The temperature history of the central 16.5-cm (6.5-in.)-long region of the test segment is also presented in this figure.] Approximately 5% (66 mCi) of the original  $^{85}\text{Kr}$  in the segment was released, an amount about five times larger than was released in test HT-1. However, analysis of the  $^{85}\text{Kr}$  release/collection pattern is complicated by a 1- to 2-min delay (depending on the amount of hydrogen formed by the steam-

Table 4. Distribution of cesium in high-temperature test 2<sup>a</sup>

Location	Temperature (°C)	Amount of <sup>134</sup> Cs found in each location			Total cesium found (µg)
		µg <sup>b</sup>	Percent of total <sup>c</sup>	Percent of released	
Fuel rod segment, initially	1445	8685 <sup>d</sup>			4.57 x 10 <sup>5d</sup>
Furnace tube					
Quartz liner	e	319.69	3.681	76.35	16,828
Quartz fuel rod holder		76.24	0.878	18.21	4,013
Thermal gradient tube	750-225	17.57	0.202	4.20	924.9
Filter pack components	128				
Stainless steel inlet fitting		0.60	0.007	0.14	31.58
First filter paper		4.59	0.053	1.10	241.6
Second filter paper		0.001	1.15 x 10 <sup>-5</sup>	2.39 x 10 <sup>-4</sup>	0.05
Third filter paper		2.69 x 10 <sup>-6</sup>	3.10 x 10 <sup>-8</sup>	6.42 x 10 <sup>-7</sup>	1.4 x 10 <sup>-4</sup>
Charcoal No. 1a		0.0	0.0	0.0	0.0
Charcoal No. 1b		0.0	0.0	0.0	0.0
Charcoal No. 1c		0.0	0.0	0.0	0.0
Charcoal No. 2a		0.0	0.0	0.0	0.0
Charcoal No. 2b		0.0	0.0	0.0	0.0
Charcoal No. 3		0.0	0.0	0.0	0.0
AgX (silver-exchanged Zeolite)		0.0	0.0	0.0	0.0
Condenser	0	0.0	0.0	0.0	0.0
Freeze trap	-78	0.0	0.0	0.0	0.0
Cold charcoal traps (two)	-78	0.0	0.0	0.0	0.0
Total		418.69	4.821	100.00	22,040

<sup>a</sup> Steam flow rate, 935 cm<sup>3</sup>/min (STP); helium flow rate, 870 cm<sup>3</sup>/min (STP); pressure, 760 torr.  
Decay time, 911 days (to Nov. 2, 1976).

<sup>b</sup> Amounts less than 1.0 x 10<sup>-6</sup> µg are given as 0.0.

<sup>c</sup> Percent of radioactive nuclide in fuel rod segment.

<sup>d</sup> Calculated for burnup of 30,570 MWd/MT of original uranium, 183.3 g of uranium originally in 30.5-cm (12-in.) segment, and 911 days decay. Total mass of all cesium isotopes = 52.6 x <sup>134</sup>Cs mass.

<sup>e</sup> Approximately 1000°C max at center, ~615 C at outlet end. Inlet end, 125°C.

Table 5. Distribution of iodine in high-temperature test 2<sup>a</sup>

Location	Temperature (°C)	Amount of <sup>129</sup> I found in each location			Total iodine found (µg)
		µg <sup>b</sup>	Percent of total <sup>c</sup>	Percent of released	
Fuel rod segment, initially	1445	3.32 x 10 <sup>4</sup> <sup>d</sup>			4.21 x 10 <sup>4</sup> <sup>d</sup>
Furnace tube					
Quartz liner	e	25.84 ± 3.6	7.78 x 10 <sup>-2</sup>	3.31	32.79
Quartz fuel rod holder		0.749 ± 0.3	2.26 x 10 <sup>-3</sup>	0.10	0.95
Thermal gradient tube	750-225	675.6 ± 11.0	2.035	86.55	857.5
Filter pack components	128				
Stainless steel inlet fitting		19.99 ± 0.1	6.02 x 10 <sup>-2</sup>	2.56	25.37
First filter paper		57.88 ± 4.0	0.174	7.41	73.45
Second and third filter papers		0.107 ± 0.1	3.22 x 10 <sup>-4</sup>	0.01	0.14
Charcoal No. 1a		0.473 ± 0.03	1.42 x 10 <sup>-3</sup>	0.06	0.60
Charcoal No. 1b		0.0	0.0	0.0	0.0
Charcoal No. 1c		0.0	0.0	0.0	0.0
Charcoal No. 2a		0.0	0.0	0.0	0.0
Charcoal No. 2b <sup>f</sup>		0.0	0.0	0.0	0.0
Charcoal No. 3 <sup>f</sup>		0.0	0.0	0.0	0.0
AgX (silver-exchanged Zeolite) <sup>f</sup>		0.0	0.0	0.0	0.0
Condenser <sup>f</sup>	0	0.0	0.0	0.0	0.0
Freeze trap <sup>f</sup>	-78	0.0	0.0	0.0	0.0
Cold charcoal (two) <sup>f</sup>	-78	0.0	0.0	0.0	0.0
Total		780.7 ± 19	2.351	100.00	990.8 ± 24

<sup>a</sup>Steam flow rate, 935 cm<sup>3</sup>/min (STP); helium flow rate, 870 cm<sup>3</sup>/min (STP); pressure, 760 torr. Decay time, 911 days (to Nov. 2, 1976).

<sup>b</sup>Amounts less than 0.03 µg are given as 0.0.

<sup>c</sup>Percent of radioactive nuclide in fuel rod segment.

<sup>d</sup>Calculated for burnup of 30,570 MWd/MT of original uranium, 183.3 g of uranium originally in 30.5-cm (12-in.) segment, and 911 days decay. Error estimate based solely on spread between duplicate activation samples. Total iodine mass = 1.27 x <sup>129</sup>I mass.

<sup>e</sup>Approximately 1000°C max at center, ~615°C at outlet end. Inlet end, 125°C.

<sup>f</sup>Not analyzed for <sup>129</sup>I. Amount assumed to be 0.0.

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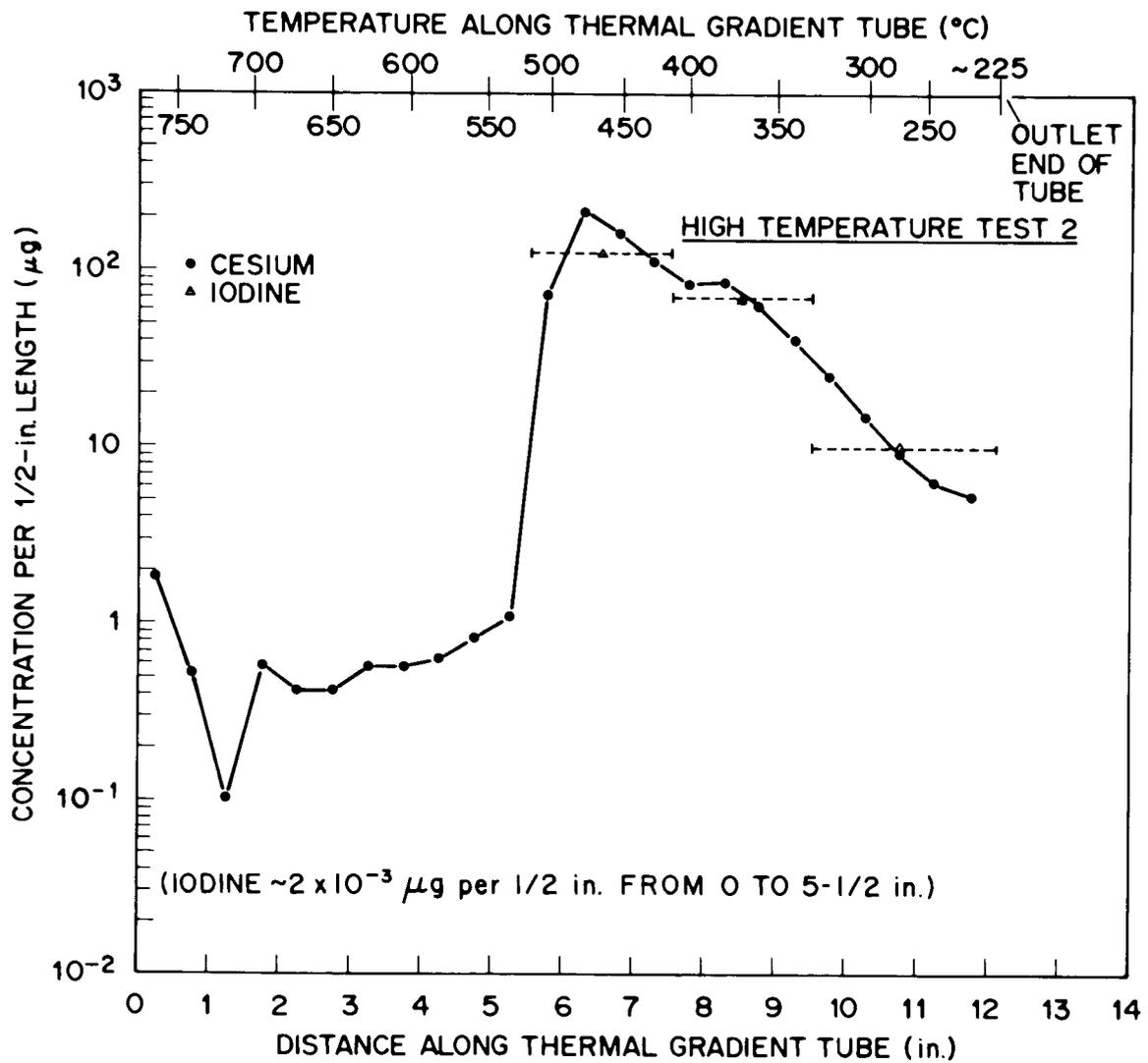


Fig. 6. Distributions of cesium and iodine collected in the thermal gradient tube during test HT-2.

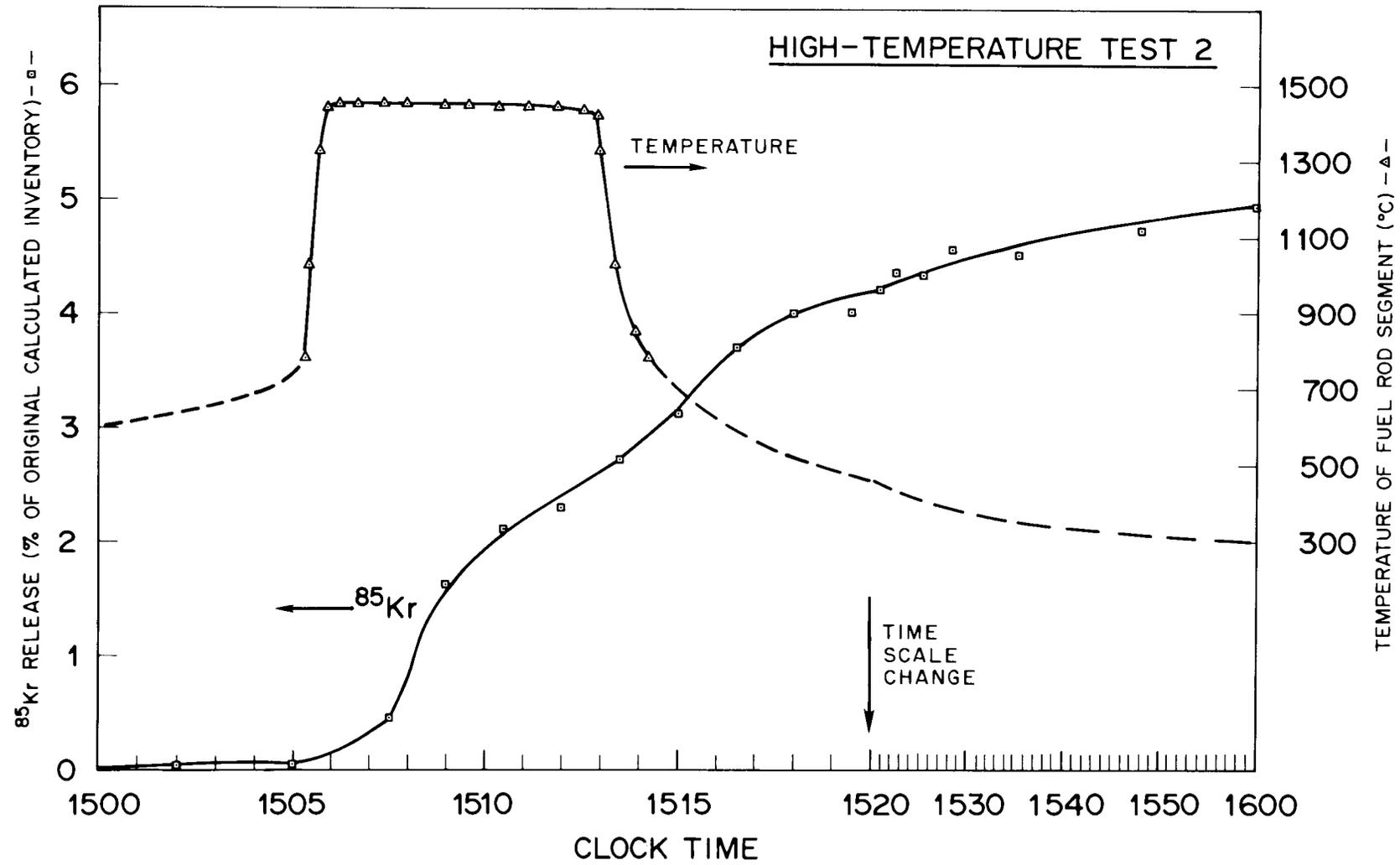


Fig. 7. Collection of <sup>85</sup>Kr in the charcoal traps during test HT-2.

zirconium reaction) between release and collection in the cold charcoal traps. This delay, as well as the concomitant smearing of the data, is caused by holdup and mixing in two 1-liter tanks, the condenser, and the freeze trap. Nonetheless, the data displayed in Fig. 7 suggest that maximum  $^{85}\text{Kr}$  release rates may have occurred at two stages of the test — at initial heating and at the time of initial cool-off.

#### 4.3 High-Temperature Test 3 ( $\sim 1610^\circ\text{C}$ )

This test was conducted at about  $1610^\circ\text{C}$  for 3 min in a flowing steam-helium atmosphere. Segment B-3b of fuel rod H-15 was used. The cladding was expanded and drilled as previously described.

The distribution of cesium in the apparatus at the end of the test, tabulated in Table 6, indicates that 10.95% (51.36 mg) of the total cesium inventory was released. Approximately 96.8% of this amount deposited in the furnace tube, primarily on the surface of the quartz liner.

The distributions of cesium and iodine deposited in the thermal gradient tube are illustrated in Fig. 8. The maximum concentrations occurred at about  $475^\circ\text{C}$ , as was the case in test HT-2 (Fig. 6). The rate of deposition of cesium in the thermal gradient tube and filter pack during the test is shown in Fig. 9 (segment temperature as a function of time is also presented in Fig. 9). Approximately 2.2% of the released cesium deposited in the thermal gradient tube. During the 1.5-min heating period ( $800$  to  $1600^\circ\text{C}$ ), no cesium activity was monitored in the thermal gradient tube, whereas the maximum rate of deposition was observed about 1 min after the test segment had attained the target temperature.

As shown in Table 7, 12.6% (5.43 mg) of the total iodine inventory in the test segment was released. At some time, the HEPA filter papers were ruptured, and some of the cesium and iodine that would normally have been retained by these papers was carried as far as the first charcoal trap. (In all previous tests with irradiated fuel, little or no cesium was found beyond the first HEPA filter paper.)

Figure 9 is also a presentation of data obtained for the collection of  $^{85}\text{Kr}$  in the cold charcoal traps. Analysis of these results indicates

Table 6. Distribution of cesium in high-temperature test 3<sup>a</sup>

Location	Temperature (°C)	Amount of <sup>134</sup> Cs found in each location			Total cesium found (μg)
		μg	Percent of total <sup>b</sup>	Percent of released	
Fuel rod segment, initially	1610	8908 <sup>c</sup>			4.69 x 10 <sup>5c</sup>
Furnace tube					
Quartz liner	d	746.0	8.37	76.48	39,278
Quartz fuel rod holder		197.9	2.22	20.29	10,419.30
Thermal gradient tube	750-185	20.93	0.235	2.15	1,102
Filter pack components	130				
Stainless steel inlet fitting		0.25	0.003	0.026	13.2
First filter paper		7.26	0.081	0.744	382.2
Second and third filter papers		0.57	0.006	0.058	30.0
Charcoal No. 1a		0.35	0.004	0.036	18.4
Charcoal No. 1b		0.34	0.004	0.035	17.9
Charcoal No. 1c		0.21	0.002	0.022	11.1
Charcoal No. 2a		0.23	0.003	0.024	12.1
Charcoal No. 2b		0.14	0.002	0.014	7.4
Charcoal No. 3		0.15	0.002	0.015	7.9
AgX (silver-exchanged Zeolite)		0.12	0.001	0.012	6.3
Cartridges and		0.72	0.008	0.074	37.9
Condenser	0	0.04	0.001	0.004	2.1
Freeze trap	-78	0.06	0.001	0.006	3.2
First charcoal trap	-78	0.10	0.001	0.010	5.3
Second charcoal trap		0.0	0.0	0.0	0.0
Tubing from filter pack to					
first charcoal trap	120-25	0.12	0.001	0.012	6.3
Total		975.5	10.95	100.00	51,360

<sup>a</sup>Steam flow rate, 1020 cm<sup>3</sup>/min (STP); helium flow rate, 870 cm<sup>3</sup>/min (STP); pressure, 760 torr. Decay time, 911 days (to Nov. 2, 1976).

<sup>b</sup>Percent of radioactive nuclide in fuel rod segment.

<sup>c</sup>Calculated for burnup of 31,360 MWd/MT of original uranium, 183.3 g of uranium originally in 30.5-cm (12-in.) segment, and 911 days decay.

<sup>d</sup>Approximately 1100°C max at center, ~590°C at outlet end. Inlet end, 120°C.

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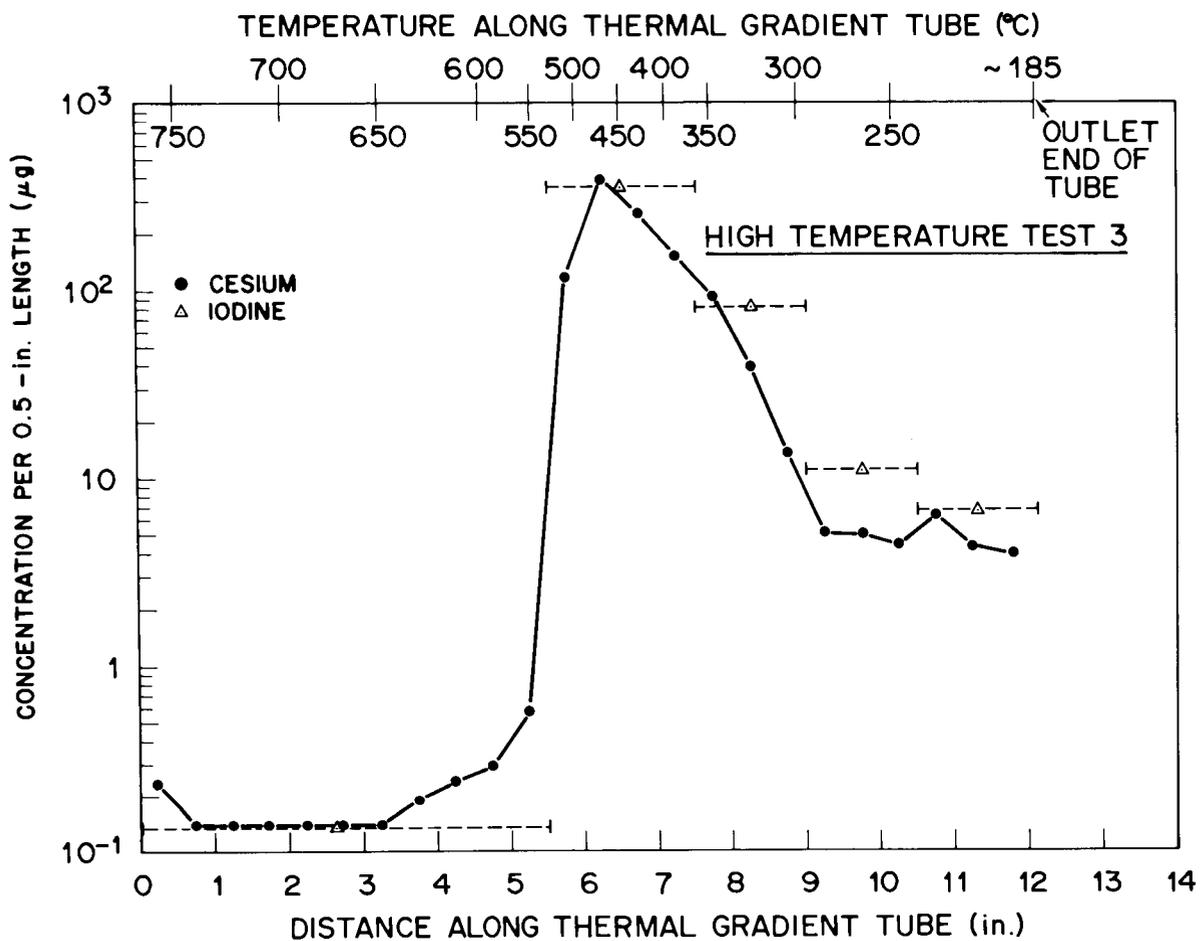


Fig. 8. Distributions of cesium and iodine collected in the thermal gradient tube during test HT-3.

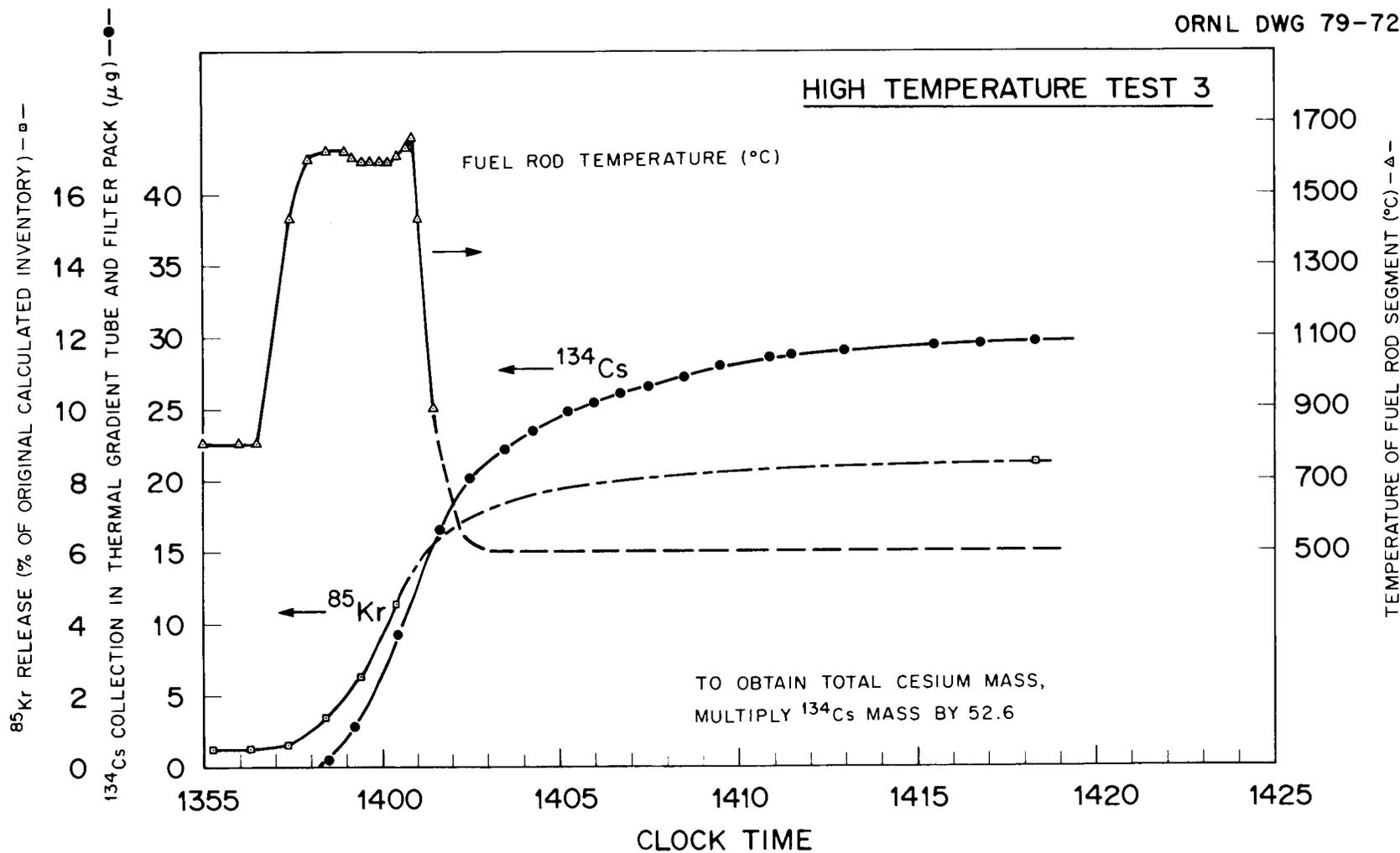


Fig. 9. Collection of <sup>134</sup>Cs in the thermal gradient tube and filter pack and of <sup>85</sup>Kr in the charcoal traps during test HT-3.

Table 7. Distribution of iodine in high-temperature test 3<sup>a</sup>

Location	Temperature (°C)	Amount of <sup>129</sup> I found in each location			Total iodine found (μg)
		μg	Percent of total <sup>b</sup>	Percent of released	
Fuel rod segment, initially	1610	3.4 x 10 <sup>4c</sup>			4.32 x 10 <sup>4c</sup>
Furnace tube					
Quartz liner	d	2091 ± 14.0	6.15	48.83	2653
Quartz fuel rod holder		259.15 ± 13.0	7.62 x 10 <sup>-1</sup>	6.05	328.9
Thermal gradient tube	750-185	1352 ± 4.0	3.98	31.58	1716
Filter pack components	130				
Stainless steel inlet fittings		16.49 ± 0.1	4.85 x 10 <sup>-2</sup>	0.39	20.93
First filter paper		440.7 ± 10.0	1.30	10.29	559.3
Second and third filter papers		34.42 ± 1.0	1.01 x 10 <sup>-1</sup>	0.80	43.68
Charcoal No. 1a		18.02 ± 0.4	5.30 x 10 <sup>-2</sup>	0.42	22.87
Charcoal No. 1b		13.82 ± 0.1	4.06 x 10 <sup>-2</sup>	0.32	17.54
Charcoal No. 1c		8.84 ± 0.2	2.60 x 10 <sup>-2</sup>	0.21	11.22
Charcoal No. 2a		9.32 ± 0.2	2.74 x 10 <sup>-2</sup>	0.22	11.83
Charcoal No. 2b		6.03 ± 0.2	1.77 x 10 <sup>-2</sup>	0.14	7.65
Charcoal No. 3		6.14 ± 0.2	1.81 x 10 <sup>-2</sup>	0.14	7.79
AgX (silver-exchanged Zeolite)		5.0 <sup>e</sup>	1.47 x 10 <sup>-2</sup>	0.12	6.3
Cartridges and screens		3.18 ± 0.2	9.35 x 10 <sup>-3</sup>	0.07	4.04
Condenser	0	4.02 ± 0.2	1.18 x 10 <sup>-2</sup>	0.09	5.10
Freeze trap	-78				
First charcoal trap	-78	4.2 <sup>e</sup>	1.24 x 10 <sup>-2</sup>	0.10	5.3
Second charcoal trap	-78	0.0	0.0	0.0	
Tubing from filter pack to					
first charcoal trap	120-25	9.33 ± 0.2	2.74 x 10 <sup>-2</sup>	0.22	11.84
Total		4281 ± 44	12.59	100.00	5433

<sup>a</sup>Steam flow rate, 1020 cm<sup>3</sup>/min (STP); helium flow rate, 870 cm<sup>3</sup>/min (STP); pressure, 760 torr. Decay time, 911 days (to Nov. 2, 1976).

<sup>b</sup>Percent of radioactive nuclide in fuel rod segment.

<sup>c</sup>Calculated for burnup of 31,360 MWd/MT of original uranium, 183.3 g of uranium originally in 30.5-cm (12-in.) segment, and 911 days decay. Error estimate based solely on spread between duplicate activation samples.

<sup>d</sup>Approximately 1100°C max at center, ~590°C at outlet end. Inlet end, 120°C.

<sup>e</sup>Estimated values. It was assumed that cesium found was present as CsI.

that 8.25% (116.4 mCi) of the total  $^{85}\text{Kr}$  inventory of the test specimen was released in the experiment.

#### 4.4 High-Temperature Test 4 (1400°C)

Prior to conducting HT-4, the cladding of the test specimen (B-8b of fuel rod H-15) was expanded, as were other fuel segments in this experimental series. The segment was heated by induction at 1400°C for only 20 sec in a flowing steam-helium atmosphere. The test was terminated because an axial crack ~10 cm long developed in the cladding that caused rapid loss of induction coupling.

The distributions of cesium and iodine in the apparatus are listed in Tables 8 and 9. These data show that 3.1% (14.2 mg) of the total cesium and 1.8% (0.75 mg) of the total iodine inventories were released.

Table 8 shows that 98.4% of the released cesium remained in the quartz furnace tube, which is typical. It was further determined by making an axial scan of the gamma activity that the released cesium was deposited primarily within a 5-cm (2-in.) region downstream from the defect opening; this suggests that most of the release occurred prior to the axial cracking of the cladding. In Table 9, it is seen that no iodine reached the impregnated charcoal; most of the iodine was found in the furnace tube (59%), the thermal gradient tube (19%), and on the first filter paper (22%). These data suggest that the released iodine was in a form other than elemental iodine, probably CsI.

Approximately 1% of the released cesium deposited in the thermal gradient tube in a manner displayed in Fig. 10, where the peak concentration occurred at 425°C. The iodine that deposited in the thermal gradient tube was also found in the region of primary cesium deposition. Additionally, Tables 8 and 9 show that similar masses of cesium (129  $\mu\text{g}$ ) and iodine (143  $\mu\text{g}$ ) deposited in the thermal gradient tube. These data suggest that iodine was primarily present as CsI.

Figure 11 gives the temperature chronology of the test and the collections of  $^{134}\text{Cs}$  in the thermal gradient tube and filter pack and of  $^{85}\text{Kr}$  in the cold charcoal traps. A total of 38.4 mCi of  $^{85}\text{Kr}$  was collected,

Table 8. Distribution of cesium in high-temperature test 4<sup>a</sup>

Location	Temperature (°C)	Amount of <sup>134</sup> Cs found in each location			Total cesium found (μg)
		μg <sup>b</sup>	Percent of total <sup>c</sup>	Percent of release	
Fuel rod segment, initially	~1400	8837 <sup>d</sup>			4.65 x 10 <sup>5d</sup>
Furnace tube					
Quartz liner	e	178.49	2.020	66.15	9395
Quartz fuel rod holder		86.49	0.984	32.22	4576
Thermal gradient tube	750-235	2.45	0.028	0.91	129.0
Filter pack components	120				
Stainless steel inlet fitting		0.09	0.001	0.03	4.74
First filter paper		1.86	0.021	0.69	97.91
Second and third filter papers		8.1 x 10 <sup>-7</sup>	9.1 x 10 <sup>-9</sup>	3.0 x 10 <sup>-7</sup>	4.3 x 10 <sup>-5</sup>
Charcoal No. 1a		3.8 x 10 <sup>-7</sup>	4.3 x 10 <sup>-9</sup>	1.4 x 10 <sup>-7</sup>	2.0 x 10 <sup>-5</sup>
Charcoal No. 1b		2.7 x 10 <sup>-7</sup>	3.1 x 10 <sup>-9</sup>	1.0 x 10 <sup>-7</sup>	1.4 x 10 <sup>-5</sup>
Charcoal No. 1c		7.6 x 10 <sup>-7</sup>	8.6 x 10 <sup>-9</sup>	2.8 x 10 <sup>-7</sup>	4.0 x 10 <sup>-5</sup>
Charcoal No. 2a		2.7 x 10 <sup>-7</sup>	3.1 x 10 <sup>-9</sup>	1.0 x 10 <sup>-7</sup>	1.4 x 10 <sup>-5</sup>
Charcoal No. 2b		2.5 x 10 <sup>-7</sup>	2.8 x 10 <sup>-9</sup>	9.2 x 10 <sup>-8</sup>	1.3 x 10 <sup>-5</sup>
Charcoal No. 3		0.0	0.0	0.0	0.0
AgX (silver-exchanged Zeolite)		1.5 x 10 <sup>-4</sup>	1.7 x 10 <sup>-6</sup>	5.6 x 10 <sup>-5</sup>	7.9 x 10 <sup>-5</sup>
Condenser	0	0.0	0.0	0.0	0.0
Freeze trap	-78	0.0	0.0	0.0	0.0
Cold charcoal traps (two)	-78	0.0	0.0	0.0	0.0
Total		269.8	3.054	100.00	14,203

<sup>a</sup>Steam flow rate, 925 cm<sup>3</sup>/min (STP); helium flow rate, 785 cm<sup>3</sup>/min (STP); pressure, 760 torr. Decay time, 911 days (to Nov. 2, 1976)

<sup>b</sup>Amounts less than 1.0 x 10<sup>-7</sup> μg are given as 0.0.

<sup>c</sup>Percent of radioactive nuclide in fuel rod segment.

<sup>d</sup>Calculated for burnup of 31,110 MWd/MT of original uranium, 183.3 g of uranium originally in 50.5-cm (12-in.) segment, and 911 days decay.

<sup>e</sup>Approximately 950°C maximum at center and ~600°C at outlet end. Inlet end, 125°C.

Table 9. Distribution of iodine in high-temperature test 4<sup>a</sup>

Location	Temperature (°C)	Amount of <sup>129</sup> I found in each location			Total iodine found (µg)
		µg <sup>b</sup>	Percent of total <sup>c</sup>	Percent of release	
Fuel rod segment, initially	1400	3.37 x 10 <sup>4</sup> <sup>d</sup>			4.28 x 10 <sup>4</sup> <sup>d</sup>
Furnace tube	e	345.7 ± 3.1	1.03	58.59	438.8
Thermal gradient tube	750-235	112.8 ± 2.6	3.35 x 10 <sup>-1</sup>	19.12	143.2
Filter pack components	120				
Stainless-steel inlet fitting		3.39 ± 0.06	1.01 x 10 <sup>-2</sup>	0.58	4.30
First filter paper		126.9 ± 3.5	3.77 x 10 <sup>-1</sup>	21.51	161.1
Second and third filter papers		1.17 ± 0.01	3.47 x 10 <sup>-3</sup>	0.20	1.48
Charcoal No. 1a		0.0	0.0	0.0	0.0
Charcoal No. 1b		0.0	0.0	0.0	0.0
Charcoal No. 1c <sup>f</sup>		0.0	0.0	0.0	0.0
Charcoal No. 2a <sup>f</sup>		0.0	0.0	0.0	0.0
Charcoal No. 2b <sup>f</sup>		0.0	0.0	0.0	0.0
Charcoal No. 3 <sup>f</sup>		0.0	0.0	0.0	0.0
AgX (silver-exchanged Zeolite) <sup>f</sup>		0.0	0.0	0.0	0.0
Condenser <sup>f</sup>	0	0.0	0.0	0.0	0.0
Freeze trap <sup>f</sup>	-78	0.0	0.0	0.0	0.0
Cold charcoal traps (two) <sup>f</sup>	-78	0.0	0.0	0.0	0.0
Total		590.0 ± 9.3	1.75	100.00	748.8 ± 11.7

<sup>a</sup>Steam flow rate, 925 cm<sup>3</sup>/min (STP); helium flow rate, 785 cm<sup>3</sup>/min (STP); pressure, 760 torr. Decay time, 911 days (to Nov. 2, 1976).

<sup>b</sup>Amounts less than 0.02 µg are given as 0.0.

<sup>c</sup>Percent of radioactive nuclide in fuel rod segment.

<sup>d</sup>Calculated for burnup of 31,110 MWd/MT of original uranium, 183.3 g of uranium originally in 30.5-cm (12-in.) segment, and 911 days decay.

<sup>e</sup>Approximately 950°C max at center and ~600°C at outlet end. Inlet end, 125°C.

<sup>f</sup>Not analyzed for <sup>129</sup>I. Amount assumed to be 0.0.

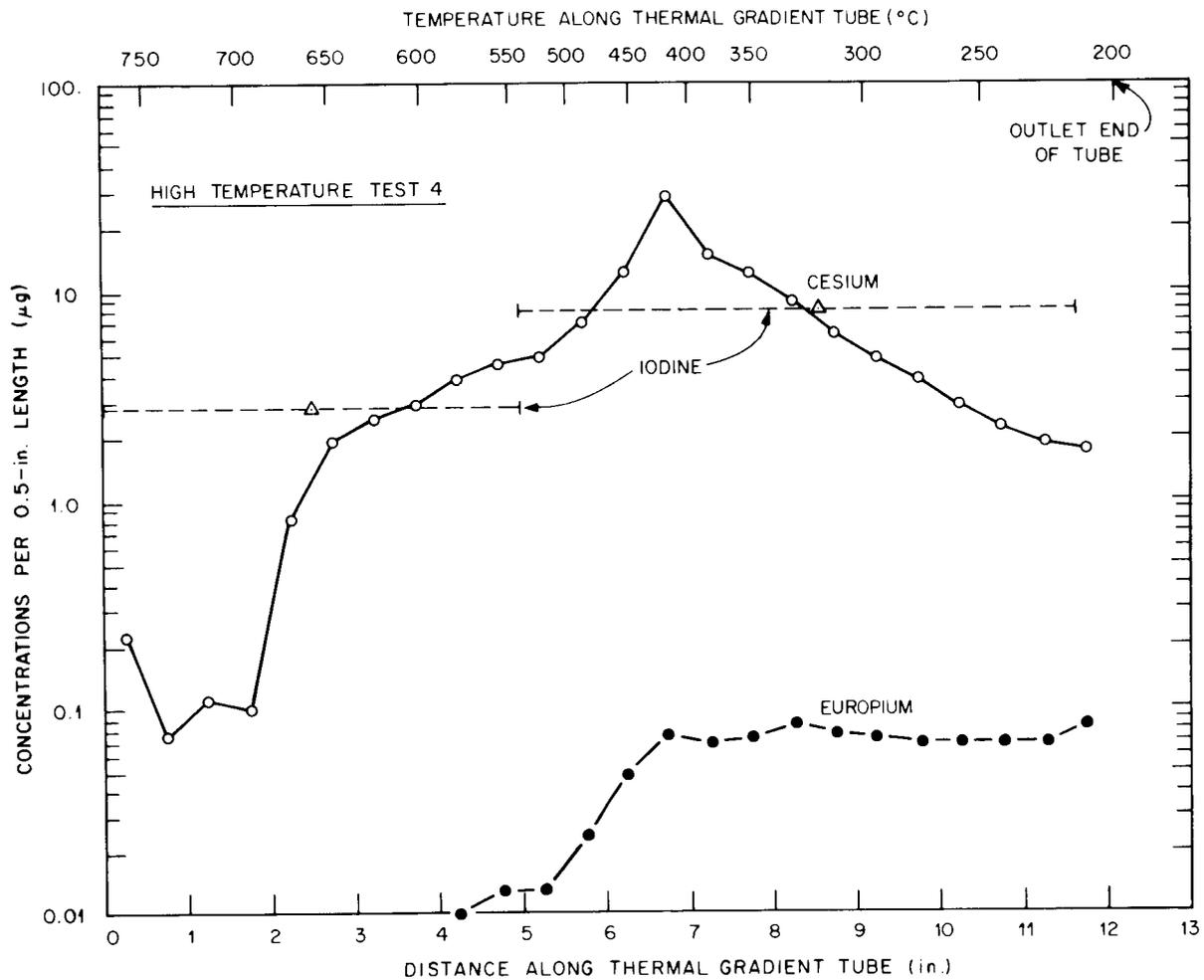


Fig. 10. Distributions of cesium, iodine, and europium collected in the thermal gradient tube during test HT-4.

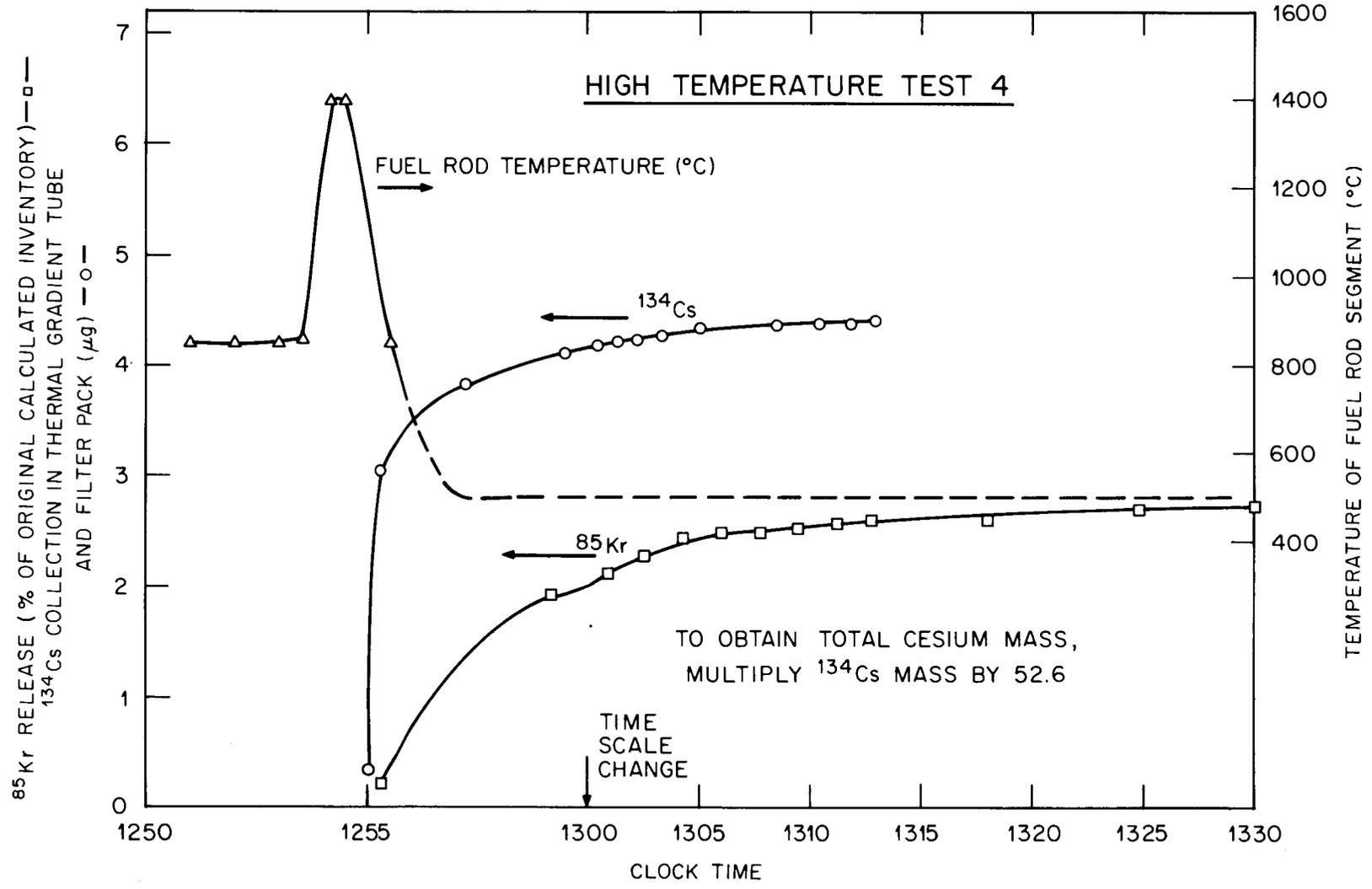


Fig. 11. Collection of  $^{134}\text{Cs}$  in the thermal gradient tube and filter pack, and of  $^{85}\text{Kr}$  in the charcoal traps during test HT-4.

which is 2.8% of the total inventory in the 30.5-cm (12-in.)-long segment. It is estimated that an additional 1.5% was lost during the pretest cladding expansion step.

The deposition of europium in the thermal gradient tube (Fig. 10) has not been observed in previous tests. The distribution is unusual in that there was no deposits in the high-temperature region, but uniform concentration occurred below 450°C. Near-uniform concentration could occur by the deposition of particles; thermophoresis would inhibit deposition in the high-temperature region only as long as the surface remained hotter than the flowing gas.

Normally in experiments of this type europium does not separate from the fuel. Europium-154 can be used as a tracer for fuel particles and dust, which are usually released only as ejected material from pressure-ruptured fuel rods. (See Sect. 7 for typical isotopic ratios in fuel particles.) In test HT-4,  $^{154}\text{Eu}$  was distributed as follows: 4.32  $\mu\text{g}$  on the furnace tube liner, 2.19  $\mu\text{g}$  on the fuel rod holder, 0.23  $\mu\text{g}$  in the thermal gradient tube, 0.08  $\mu\text{g}$  on the stainless steel inlet fitting, and 1.41  $\mu\text{g}$  on the first filter paper. The amount of  $^{154}\text{Eu}$  on the filter paper was much more than could be present in fuel particles based on a single uranium analysis. The chemical/physical form of the released europium is therefore uncertain. It is possible that reaction of zirconium with  $\text{UO}_2$ <sup>15,16</sup> might have affected the chemical form of the europium.

## 5. DISCUSSION

### 5.1 Physicochemical Form of Cesium and Iodine Collected in the Apparatus

The physicochemical forms of cesium and iodine released in the High-Temperature Test Series are summarized in Table 10. Similar data from the High Burnup Test Series is given for comparison;<sup>1</sup> only high-burnup fuel rod segments from the H. B. Robinson-2 reactor were used. The released cesium and iodine were divided into three forms according to the location of collection in the apparatus. Fuel dust ejected from pressure-ruptured test rods was not included. The ejected fuel, most of which could be

Table 10. Physicochemical form of cesium and iodine collected in fission product release tests

Test No.	Temperature (°C)	Test duration (min)		Amount of collected material in designated form						
				Condensed or reacted >200°C		Particulate		Reactive iodine		Total
				µg	%	µg	%	µg	%	µg
HT-3	1600	3	Cesium	48,648	98.86	561.3	1.14	10.0	0.18	49,210
			Iodine	4,698	86.47	725.4	13.85			
HT-2	1445	7	Cesium	21,766	98.76	273.2	1.24	1.09	0.11	22,039
			Iodine	891.2	89.95	98.47	9.94			
HT-4	1400	0.4	Cesium	14,101	99.277	102.7	0.723	2.28	0.30	14,203
			Iodine	582.0	77.72	164.6	21.98			
HT-1	1300	10	Cesium	506.4	96.67	17.42	3.33	7.23	10.13	523.9
			Iodine	50.05	70.16	14.06	19.71			
HBU-12 <sup>a</sup>	700-1200	480	Cesium	1896.5	95.02	99.38	4.98	0.50	0.265	1995.9
			Iodine	171.9	91.19	16.13	8.56			
HBU-11 <sup>b</sup>	1200	27	Cesium	99.97	68.98	44.06	31.0	1.69	8.37	142.0
			Iodine	6.82	33.8	11.67	57.8			
HBU-2	900	120	Cesium	2.817	99.997	7.75 x 10 <sup>-5</sup>	0.0028	1.288	73.40	2.818
			Iodine	0.245	13.95	0.222	12.69			
HBU-1	700	300	Cesium	0.1229	99.996	5.26 x 10 <sup>-6</sup>	0.0043	0.669	72.39	0.1230
			Iodine	0.1623	17.56	0.0930	10.06			
HBU-4	500	1200	Cesium	0.01696	99.919	1.38 x 10 <sup>-5</sup>	0.0813	0.00457	43.52	0.0170
			Iodine	0.043	40.95	0.0163	15.52			
HBU-10 <sup>c</sup>	900-1200	11	Cesium	263.9	94.43	15.53	5.56	7.33	52.7	279.5
			Iodine	3.67	26.4	2.81	20.2			
HBU-9 <sup>c</sup>	900-1100	10	Cesium	110.6	98.48	1.74	1.55	14.93	88.2	112.3
			Iodine	1.00	5.91	0.99	5.85			
HBU-8 <sup>c</sup>	900	61	Cesium	37.13	93.76	2.47	6.24	11.85	87.6	39.60
			Iodine	0.50	3.70	1.18	8.72			
HBU-7 <sup>c</sup>	900	1	Cesium	122.3	93.64	8.32	6.37	0.47	4.22	130.6
			Iodine	7.87	70.7	2.79	25.1			

<sup>a</sup>Fission products purged from gap space with purified helium.

<sup>b</sup>High temperature of quartz may have resulted in high particulate levels.

<sup>c</sup>Fuel rod segment pressure-ruptured at 900°C and heat continued for time shown.

shaken from apparatus components, was identified by its content of gamma-emitting isotopes such as  $^{106}\text{Ru}$ ,  $^{144}\text{Ce}$ , and  $^{154}\text{Eu}$ .<sup>1</sup>

The first form, the amount condensed or reacted at temperatures above 200°C, consists of all of the cesium and iodine collected on the furnace tube, fuel rod holder, and thermal gradient tube. In the case of iodine, most of this form is believed to be CsI. Except for tests HT-3 and HT-4, most of the iodine in this category was found in the thermal gradient tube. Some cesium deposited as CsI, but most apparently formed CsOH by reaction with steam; the CsOH in turn reacted with the quartz furnace tube and fuel rod holder. According to the calculations of Besmann and Lindemer,<sup>13</sup> vaporized cesium in equilibrium with  $\text{UO}_2$ , zirconium, and steam would be predominantly in the form Cs (gas) rather than CsOH (gas) or  $(\text{CsOH})_2$  (gas) as long as the oxygen potential remained below approximately -450 kJ/mol. In our apparatus, Cs (gas) would be carried through the thermal gradient tube and condense in the filter pack ( $\sim 125^\circ\text{C}$ ). Since we find only small amounts of cesium in this part of the apparatus, it appears that any Cs (gas) released from the fuel rod reacts outside of the fuel rod with steam or possibly with oxidizing contaminants in the flowing steam-helium mixture to form species condensible at 300 to 500°C. In all tests except test HBU-11 (and test HBU-12 which bypassed the furnace tube), more cesium was collected in the furnace tube than in the thermal gradient tube.

The second physicochemical category is particulate. For cesium, all of the material collected on the stainless steel inlet to the filter pack and on the filter papers was counted as particulate. In all but test HT-3, there was no real penetration of the filter papers; in this test the papers were damaged, and considerable penetration of cesium and iodine occurred. Because essentially identical masses of cesium and iodine were found at most locations, it is highly probable that the penetrating particulate material was CsI (or some other solid material containing condensed CsI). Material in fuel dust was not included in this category.

Determination of the amount of iodine in the particulate form was more difficult because some reactive iodine ( $\text{I}_2$  or HI) deposited on the stainless steel filter pack inlet fitting. Iodine collected on this fitting was therefore apportioned between particulate and reactive in

direct proportion to particulate iodine and reactive iodine passing the inlet fitting. Particulate iodine passing the fitting was defined as total iodine on the first filter, less the average iodine content of the second and third filter papers. Experience has shown that the filter papers adsorb small but significant amounts of reactive iodine and that iodine concentrations on the second and third filter papers are essentially the same.<sup>3</sup>

The third physicochemical category is reactive iodine, which includes species such as  $I_2$ , HI,  $CH_3I$ , and HOI. The amount of reactive iodine is given by the sum of that found on the charcoal, on the second and third filters, a portion of that on the first filter paper equal to the average of the second and third filter papers, and the nonparticulate amount calculated to be on the inlet fitting.

For all of the tests detailed in Table 10, the percentage of released iodine in the condensed or reacted  $>200^\circ C$  form ranged from 3.7 to 91.2%, that in the particulate form ranged from 5.8 to 57.8%, and that in the reactive form ranged from 0.11 to 88.2%. Iodine collected in the high-temperature tests tended to have large amounts in the first category and small amounts in the third. The large mass of iodine released may have contributed to this distribution. The reverse situation, small fraction collected  $>200^\circ C$  and large fraction in the reactive form, occurred mainly with lower temperature tests conducted for longer time spans. The long exposure would permit slow oxidation of released CsI by contaminants in the steam and helium to form  $I_2$ .<sup>13</sup> In no case did the mass of reactive iodine exceed 15  $\mu g$ .

## 5.2 Volume of Fission Products Escaping in Gaseous Form

The volume of fission gas and volatile fission products released during the high-temperature tests is listed in Table 11. The volume of released material is considerably greater than the volume of the expanded gap space, 1.91  $cm^3$ ; thus the gap probably remained purged free of steam for most of each test by the action of released fission products. The velocity through the hole, averaged for the entire run, is also given in Table 11. It appears that the steam-hydrogen-helium atmosphere outside the cladding could not have had much influence on releases that occurred inside the cladding.

Table 11. Amount of fission products escaping in gaseous form

Test No.	Test Temperature (°C)	Volume of gas at test temperature (cm <sup>3</sup> ) <sup>a</sup>				Average velocity of escaping gas in hole (cm/sec)
		Xe + Kr <sup>b</sup>	Cs + Rb <sup>c</sup>	I + Br <sup>d</sup>	Total	
HT-1	1300	10.7	0.59	0.82	11.4	0.93
HT-2	1445	53.1	27.4	1.24	81.7	9.82
HT-3	1610	102.6	52.8	7.46	162.9	45.7
HT-4	1410	30.1	17.2	0.91	48.2	101.3

<sup>a</sup>Assumes all of each element in gaseous form.

<sup>b</sup>91.1% xenon.

<sup>c</sup>84.1% cesium.

<sup>d</sup>87.6% iodine.

### 5.3 Time and Temperature Release Dependence

The release data for krypton, cesium, and iodine are restated in Table 12 with release calculated on the basis of the 15.24-cm heated length rather than the 30.48-cm total segment length. The percentage values are therefore double those given in Tables 2 through 9. The estimated amount of krypton released during the clad expansion procedure was not changed.

Table 12. Release of krypton, cesium, and iodine from heated length of H. B. Robinson-2 fuel

Test No.	Temperature (°C)	Amount of total inventory in heated length released (%)		
		Krypton	Cesium	Iodine
HT-1	~1300	2.65	0.224	0.330
HT-2	1445	11.0	9.64	4.70
HT-3	1610	17.5	21.9	25.2
HT-4	~1400	7.1	6.11	3.50

Figure 12 displays cesium and iodine release as a function of time and temperature. Except for the 1400°C curve for which two points are available, the curves are speculative in nature. Guidance for the slope is obtained from the 1400°C data and from fission gas release at 900°C (Fig. 13) where several data points are available. According to this interpretation, most of the cesium and iodine were released quickly, especially at the higher temperatures.

Krypton release percentages are plotted in Fig. 13. Additional release values from the High Burnup Test Series are shown, as well as fission gas release calculated using the ANS 5.4 proposed standard model.<sup>17</sup> The ANS equations model in-reactor fission gas release for temperatures ranging from 1000 to 2500°C. They apply to steady power conditions and therefore should not be expected to predict the transient gas release that occurred in our tests.

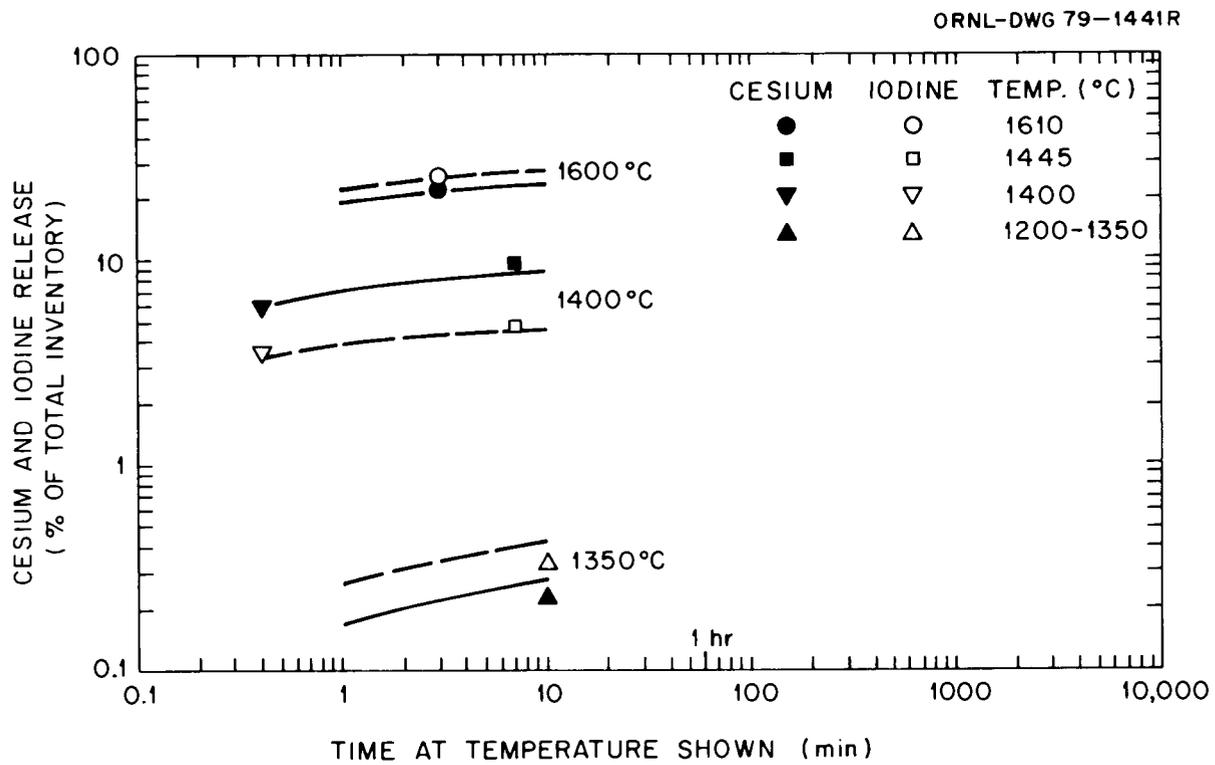


Fig. 12. Release of cesium and iodine from 15.2 cm of H. B. Robinson-2 fuel in steam.

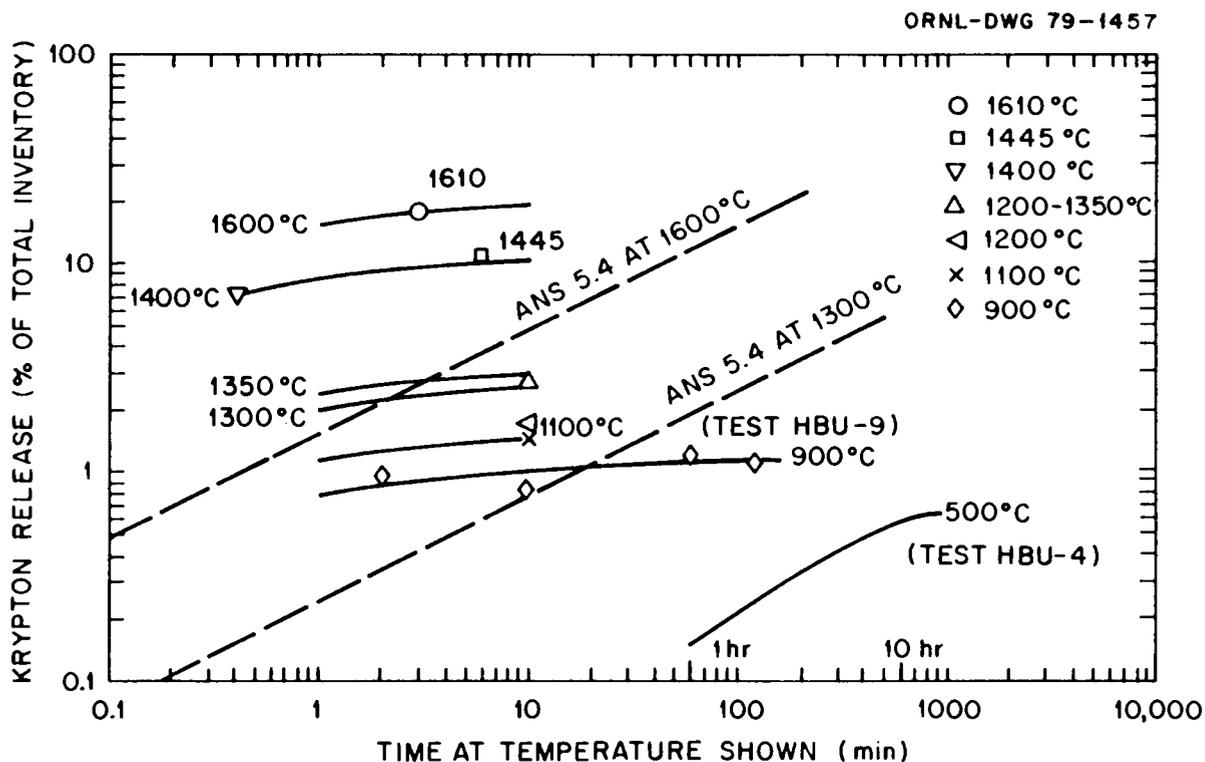


Fig. 13. Release of <sup>85</sup>Kr from H. B. Robinson-2 fuel in steam.

Using the data from these figures and similar data from the High Burnup Test Series, Fig. 14 was constructed showing fission product release from H. B. Robinson-2 fuel heated for 10 min using extrapolated data from Figs. 12 and 13 and additional data from the source term model.<sup>6</sup> Below 1300°C, most of the released gas is believed to have been shallowly embedded in the surfaces of the fuel and cladding.<sup>1</sup> The sharp increase in release between 1350 and 1400°C is believed to be a result of the release between 1350 and 1400°C is believed to be a result of the release of fission products previously accumulated at the grain boundaries. Details of this mechanism are discussed in Sect. 5.5.

#### 5.4 Application of the Data to Transient Release Calculations

Because our high-temperature tests were of relatively short duration, the data are most suitable for use with transient heating scenarios. The data from Figs. 12 and 13 and from the source term model<sup>6</sup> were used to prepare Table 13 and Fig. 15, which show fission product release occurring in 2 min. Figure 15 includes cesium and iodine release from the initial gap inventory using our LOCA source term model for gap release at  $\leq 1200^\circ\text{C}$ .<sup>6</sup> For H. B. Robinson fuel, grain boundary release exceeds the gap release above  $\sim 1350^\circ\text{C}$ . It should be emphasized that Table 13 and Figs. 14 and 15 apply directly only to LWR fuel with an initial gap inventory of 0.3% for cesium and iodine, and burnup of  $\sim 30,000$  MWd/MT.

Most rapid transients in reactor fuels would be caused by internal heating, either fission or decay. If the existence of temperature gradients does not significantly affect fission product release, Table 13 and Fig. 15 can be used to estimate fission gas release by dividing the fuel into volume increments according to the temperature distribution. Figure 15 can be used to estimate cesium and iodine release in the same manner.

For the special case of uniform internal heat generation and constant thermal conductivity, a cylindrical fuel rod at heat flow equilibrium will have equal volumes of fuel exposed to identical temperature differentials. For example, if the fuel centerline temperature is 1250°C and the surface 850°C, 25% of the fuel will reside within the temperature range 850 to 950°C, 25% within 950 to 1050°C, etc. It was then assumed that the fuel

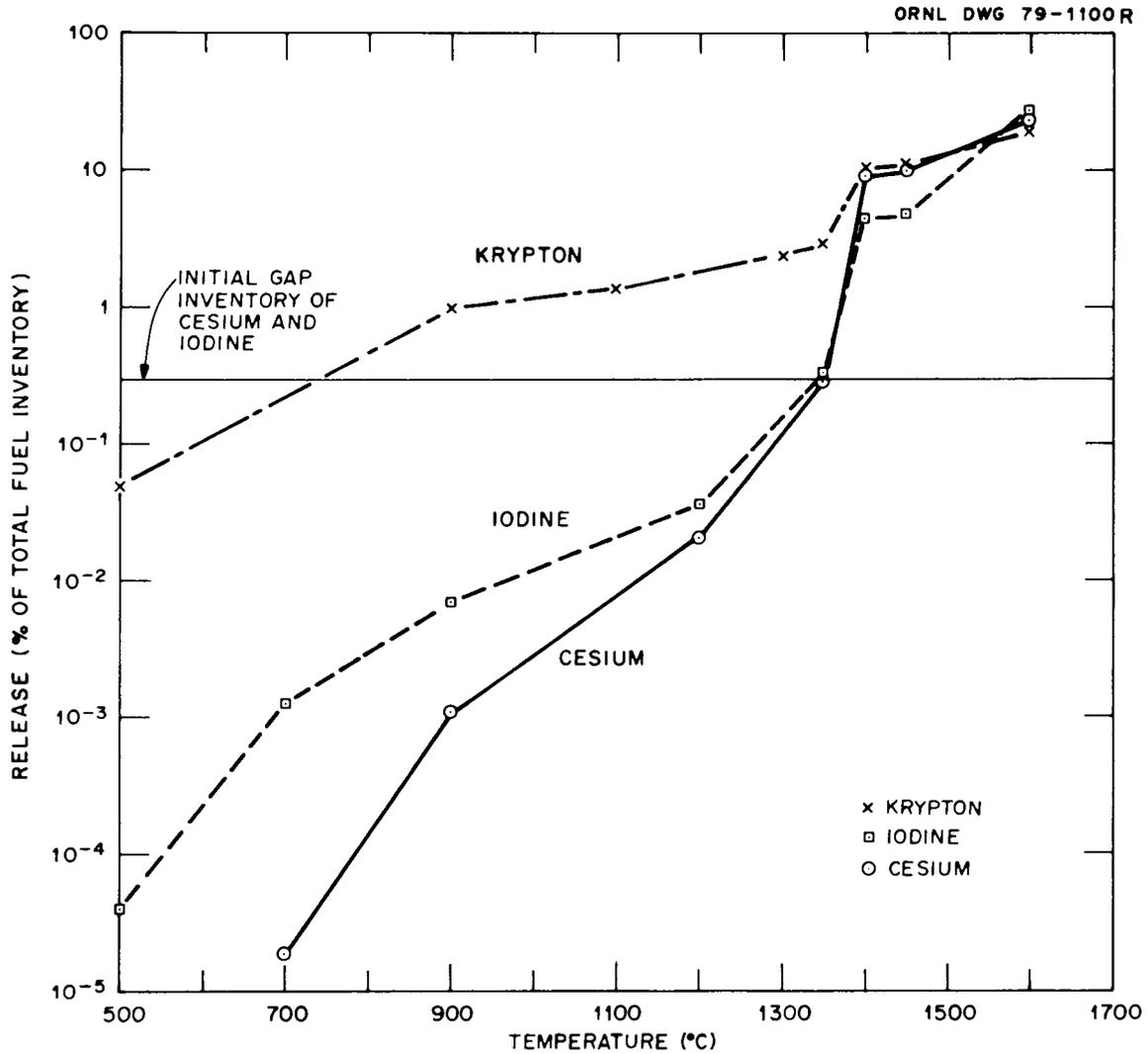


Fig. 14. Release of krypton, iodine, and cesium from H. B. Robinson-2 fuel heated for 10 min in steam (based on extrapolations from other heating times).

Table 13. Percent of krypton, cesium, and iodine released in 2 min from H. B. Robinson-2 fuel

Temperature (°C)	Krypton	Cesium	Iodine
900	0.9	$3.08 \times 10^{-4}$	$2.26 \times 10^{-3}$
1000	1.1	$1.16 \times 10^{-3}$	$6.10 \times 10^{-3}$
1100	1.3	$3.60 \times 10^{-3}$	0.014
1200	1.7	$9.58 \times 10^{-3}$	0.030
1300	2.2	0.06	0.08
1400	9.4	7.0	4.3
1500	14.3	14.6	13.1
1600	16.7	21.0	24.5

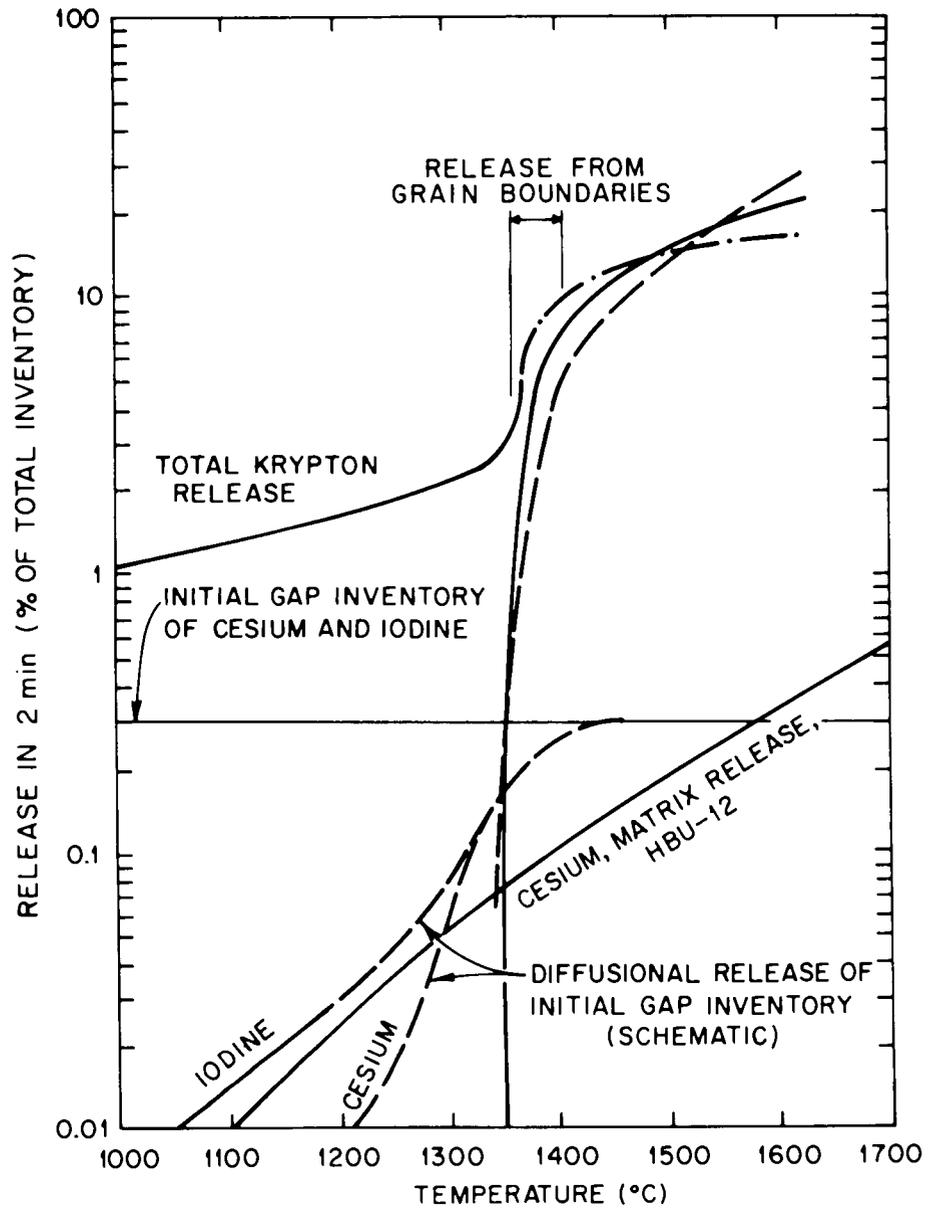


Fig. 15. Release of krypton, iodine, and cesium from H. B. Robinson-2 fuel heated for 2 min in steam (based on extrapolations from other heating times).

within each temperature zone would release the fraction of krypton shown in Table 13 and Fig. 15. Table 14 was prepared for this special case and covers fission gas release for transients in which the fuel is held for 2 min at a selection of surface and centerline temperatures. For the above conditions of 2 min with surface at 850°C and centerline 1250°C, the estimated fission gas release is 1.3%. Fission gas already in the plenum must be added to this amount, of course.

Table 14. Predicted release of krypton from H. B. Robinson fuel heated for 2 min

Surface temperature (°C)	Amount of krypton released (%) for centerline temperature shown <sup>a</sup>				
	850°C	1050°C	1250°C	1450°C	1650°C
850	0.8	1.0	1.3	2.8	6.0
1050		1.2	1.5	3.7	7.6
1250			1.9	5.8	10.7
1450				12.2	15.5
1650					17.0

<sup>a</sup>Assumes uniform volumetric heating of fuel, uniform thermal conductivity, and no temperature gradient effects.

This method is not directly applicable to the release of cesium and iodine for surface temperatures below 1300°C because of retention in the gap space. Above 1300°C the released cesium and iodine species seem to behave more like inert gases, so the method is probably applicable. Estimates of fission gas, cesium, and iodine release obtained by this method are only applicable to low gap inventory, high-burnup fuel similar to the H. B. Robinson-2 fuel tested. If the curves shown in Figs. 12 and 13 are correct, releases at 1 min heating time would be only ~80% of those at 2 min, and releases at 10 min would be only ~1.2 times greater.

### 5.5 High-Temperature Release Mechanisms

The rapid release between 1350 and 1400°C is believed to have occurred from fission products previously accumulated in or very close to the grain boundaries.

Many investigators have discussed the role of grain boundaries in fission gas release during in-reactor operation.<sup>18-23</sup> Evidence indicates that fission gas slowly diffuses from the UO<sub>2</sub> grains to the grain boundaries and thence to the grain edges.<sup>20,24</sup> As fission gas accumulates, bubbles link, especially at the grain edges, to form tunnels to connect with open porosity, thus releasing the fission gas. High temperature accelerates all stages of fission gas release. Temperature gradients have the greatest effect on fission gas release at high temperatures when columnar grain growth occurs.<sup>19</sup>

Argonne National Laboratory has conducted tests with H. B. Robinson fuel from the same bundle as our test rods using direct resistive electrical heating of a short column of unclad fuel pellets.<sup>25</sup> The direct electric heating (DEH) of the pellets results in temperature gradients similar to those caused by fission or decay heat in the UO<sub>2</sub>. Examination of heated specimens using the scanning electron microscope (SEM) shows that fission gas is released from the grain boundaries by two processes. One is the linkage of bubbles to form tunnels, and the other is a more mechanical separation of grain boundaries, probably a result of stresses in the heated pellets combined with the pressure exerted by fission gas bubbles.

One would expect somewhat lower stresses in our nearly uniformly heated fuel. How much difference in release this should make from DEH fuel is not clear. Most of the DEH tests have been transients of shorter duration than ours, yet their SEM examinations show that the time was sufficient for tunnels to form or for grains to separate.

Examinations of the fuel rod segments from tests HBU-10 and -11, and from tests HT-1, -2, -3, and -4 are being conducted at Argonne National Laboratory.<sup>26</sup> The results of these examinations should be valuable in establishing the mechanism of release at the higher temperatures.

Normal operation for the H. B. Robinson fuel was apparently at low enough temperature<sup>1</sup> that neither grain boundary separations nor gas bubble linkage could occur.<sup>25,27</sup> (The fission gas release in H. B. Robinson-2 bundle B0-5 rods was only ~0.25% at ~28,000 MWd/MT average burnup.) One would expect that either continued burnup at the same temperature or a moderate temperature increase from higher linear power would serve to initiate bubble coalescence, linkage, and more significant in-reactor fission gas release.

It must be emphasized that the results presented in this report are for a specific type of fuel: high burnup attained at low fuel temperature (low fission gas release). Fuel irradiated at higher temperatures to high burnup may not have the same quantity of fission gas and other fission products in the grain boundaries ready for release. Also, the grain boundary structure may be different.

Fuel irradiated to lower burnup at comparable temperatures would not have as much gas in the grain boundaries. Zimmerman<sup>21</sup> found that the concentration of gas in pores and bubbles increased with burnup.

## 6. CONCLUSIONS

The data presented in this report demonstrate that for LWR fuel irradiated to high burnup with low fission gas release, a large increase in release of fission gas, cesium, and iodine occurs when the fuel is heated uniformly to a minimum of 1350 to 1400°C. Within 2 min at 1400°C, ~4 to 9% of the fission gas, cesium, and iodine in the fuel rod is released. Releases at 1-min and 10-min heating times are estimated to be only factors of 0.8 or 1.2 different, respectively, from those at 2 min. At 1600°C the releases in 2 min are about 17 to 25% of the total inventory.

Because only four tests with a single type of LWR fuel were performed, it is probably appropriate to consider this series of tests as scoping in character. LWR fuel with different irradiation histories can be expected to give different release results in the temperature range 1300 to 1600°C.

7. SUPPLEMENTARY INFORMATION ON AMOUNTS OF FISSION PRODUCTS IN H. B. ROBINSON-2 FUEL

Both  $^{144}\text{Ce}$  and  $^{154}\text{Eu}$  have been used as tracers for fuel particles either released or shaken loose from the test fuel segments in our fission product release studies. A summary of the amounts of these and several other gamma emitters relative to the amount of  $^{137}\text{Cs}$  measured in a number of fuel particle or fuel dust samples is presented in Table 15. Although cesium is one of the more volatile fission products, the H. B. Robinson fuel used in our high-burnup and high-temperature tests was irradiated at sufficiently low heat ratings that very little cesium migration occurred. Also, the  $^{137}\text{Cs}$  isotope yield from fissioning is relatively uncomplicated by neutron capture effects and is not greatly different for uranium and plutonium fissions. The long half-life gives essentially equal weight to fissions that occurred early and late in the 2.4-year irradiation period in the Robinson core.

The columns in Table 15 are arranged according to fuel rod number and distance from the bottom of the rod. There appears to be no correlation with rod number, location in rod (the  $^{144}\text{Ce}/^{154}\text{Eu}$  ratio might be an exception), or half-life. Occasional large deviations from the mean might be the result of local contamination. The Zircaloy cladding contains significant amounts of  $^{125}\text{Sb}$ , for example.

A fission product inventory calculation performed with ORIGEN in 1976<sup>1</sup> was used as a basis for calculating the fractional release from H. B. Robinson-2 fuel for all pertinent isotopes except  $^{134}\text{Cs}$ . Table 16 summarizes isotopic ratios from this ORIGEN run and includes two recent calculations by ORIGEN using more recent values for yields, cross sections, etc.<sup>28</sup>

Table 15. Ratios of gamma emitters in H. B. Robinson fuel particles

Segment number <sup>a</sup>	Normalized isotope mass ratios <sup>b</sup>					Source of particle
	$26.2 \left( \frac{^{106}\text{Ru}}{^{137}\text{Cs}} \right)$	$278 \left( \frac{^{125}\text{Sb}}{^{137}\text{Cs}} \right)$	$24.5 \left( \frac{^{134}\text{Cs}}{^{137}\text{Cs}} \right)$	$41.5 \left( \frac{^{144}\text{Ce}}{^{137}\text{Cs}} \right)$	$61.0 \left( \frac{^{154}\text{Eu}}{^{137}\text{Cs}} \right)$	
A2	0.94	2.48	0.98	1.06	0.92	HBU-6; drill bit
A2	0.55	0.73	1.00	0.98	0.76	HBU-6; dust shaken from fuel rod holder (air, 700°C)
A3b	1.70	77.10	1.14	1.14	0.82	HT-1; drill bit
A3b	0.99	1.28	0.97	1.16	1.00	HT-1; dust from drill stand
A5	0.82	1.00	1.07	1.01	0.98	HBU-5; drill bit
A6	1.05	0.98	0.98	0.98	1.12	HBU-10; dust shaken from fuel rod holder (900°C rupture)
B3a	0.67	0.76	1.03	0.98	0.89	HT-2; particles from work table
B3a	0.77	1.01	1.02	1.23	1.09	HT-2; particles from roughing filter
B4	1.00	1.03	1.00	1.00	1.09	HBU-8; dust shaken from furnace tube liner (900°C rupture)
B5	1.02	1.06	1.01	1.03	1.14	HBU-9; dust shaken from furnace tube liner (900°C rupture)
B6	1.02	1.04	1.00	0.98	1.17	HBU-7; dust shaken from fuel rod holder (900°C rupture)
B7	0.64	0.76	1.05	0.97	0.97	Fuel chip from broken segment
B7	0.65	0.79	1.07	0.93	1.01	Fuel chip from broken segment
B8b	1.26	1.06	1.04	1.91	1.32	HT-4; drill bit

<sup>a</sup>The "A" segments were cut from rod D-12 and the "B" segments from rod H-15. The numbers relate to distance from the bottom of the rods as detailed in ref. 1.

<sup>b</sup>Amounts present Nov. 2, 1976. (911 days decay).

Table 16. Mass ratios of selected isotopes for H. B. Robinson-2 fuel

Source of ratio	Mass ratio <sup>a</sup>					
	$\frac{^{137}\text{Cs}}{^{106}\text{Ru}}$	$\frac{^{137}\text{Cs}}{^{125}\text{Sb}}$	$\frac{^{137}\text{Cs}}{^{134}\text{Cs}}$	$\frac{^{137}\text{Cs}}{^{144}\text{Ce}}$	$\frac{^{137}\text{Cs}}{^{154}\text{Eu}}$	$\frac{^{137}\text{Cs}}{^{129}\text{I}}$
Table 15	26.2	278	24.5	41.5	61.0	b
1979 ORIGEN (31,360 MWd/MT)	34.1	138	21.6	28.7	33.0	6.10
1979 ORIGEN (30,000 MWd/MT)	34.1	137	22.3	28.7	33.7	6.08
1976 ORIGEN (31,360 MWd/MT)	35.2 <sup>c</sup>	211 <sup>c</sup>	14.3 <sup>d</sup>	29.7	23.6	6.13 <sup>c</sup>
Dissolved fuel sample 1 BC <sup>e</sup>	43.3	391	25.3	35.1	b	b

<sup>a</sup>Masses corrected to 911 days decay (Nov. 2, 1976).

<sup>b</sup>Not determined.

<sup>c</sup>These values were used in determining the total inventory in H. B. Robinson-2 fuel.

<sup>d</sup>An experimentally measured ratio of 23.4 was used in determining the total inventory of  $^{134}\text{Cs}$  in H. B. Robinson-2 fuel for fission product release calculations.

<sup>e</sup>J. H. Goode and R. G. Stacy, Head-End Reprocessing Studies with H. B. Robinson-2 Fuel, ORNL/TM-6037, pp. 58-65 (June 1978).

Isotopic ratios for dissolved H. B. Robinson-2 fuel are also listed in Table 16. Sample 1 BC is typical of the results obtained from several different samples of fuel from bundle B05.<sup>29</sup> These analyses have shown that the ORIGEN calculations for  $^{137}\text{Cs}$  inventory are accurate in absolute terms (g  $^{137}\text{Cs}$ /g initial U). The data in Table 16 indicate that our basis for total inventory and fractional release of  $^{106}\text{Ru}$ ,  $^{125}\text{Sb}$ , and  $^{129}\text{I}$  as determined by the 1976 ORIGEN calculation and an experimental value for  $^{134}\text{Cs}$  need not be corrected. All ORIGEN calculations for  $^{154}\text{Eu}$ , and the 1979 ORIGEN calculations for  $^{125}\text{Sb}$ , appear to be significantly low.

The ORIGEN calculation believed to be most representative of the H. B. Robinson-2 fuel used in our high-temperature tests is the 1979 ORIGEN

calculation for a burnup of 30,000 MWd/MT.<sup>28</sup> The masses of important elements so calculated are given in Table 17, whereas Table 18 is a listing of the masses and radioactivities of the principal radionuclides.

For each of our fission product release tests we determined the release of  $^{134}\text{Cs}$ ,  $^{129}\text{I}$ , and  $^{85}\text{Kr}$  both as mass released and percentage of total inventory. In Table 19 are listed the total inventories used (reference inventory) along with inventories calculated by two recent ORIGEN runs. The releases of  $^{106}\text{Ru}$  and  $^{125}\text{Sb}$  were reported only when detected; in many of the tests the high gamma activity of the released cesium may have prevented their detection. The fractional release of  $^{134}\text{Cs}$  was occasionally checked using  $^{137}\text{Cs}$  to verify that  $^{134}\text{Cs}$  was representative of other cesium isotopes.

The reference inventory is the inventory of the highest burnup portions of the fuel rod. For test segments cut from regions containing lower burnup, the reference inventory was reduced according to the gamma scan shown in Fig. 1.

If the 1979 ORIGEN calculations are used to determine an "updated inventory," we believe that the appropriate inventory is midway between 30,000 and 31,360 MWd/MT. The last column in Table 19 shows the ratio of the reference inventory actually used in our calculations to the 1979 ORIGEN average (30,680 MWd/MT). This is the correction factor that should be applied to percentage release values (not mass releases) if the updated inventory is preferred. We believe that such an adjustment is warranted only for the determination of conservative (high) percentage releases.

## 8. ACKNOWLEDGMENTS

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Table 17. Amounts of principal fission product elements in  
H. B. Robinson fuel after 911 days decay<sup>a</sup>

Element	Grams per metric ton	Milligrams per 30.5 cm (12 in.)	Element	Grams per metric ton	Milligrams per 30.5 cm (12 in.)
Br	19.29	3.536	Te	447.6	82.05
Kr	317.2	58.14	I	223.5	40.97
Rb	297.3	54.50	Xe	4,884.0	895.2
Sr	733.7	134.5	Cs	2,352.0	431.1
Y	391.6	71.78	Ba	1,348.0	247.1
Zr <sup>b</sup>	3,113.0	570.6	La	1,094.0	200.5
Mo	3,010.0	551.7	Ce	2,178.0	399.2
Tc	701.5	128.6	Pr	1,004.0	184.0
Ru	2,066.0	378.7	Nd	3,577.0	655.7
Rh	433.3	81.26	Sm	766.1	140.4
Pd	1,348.0	247.1	Eu	144.5	26.49
Sn <sup>b</sup>	88.53	16.23	U	959,400.0	175,860.0 <sup>c</sup>
Sb	25.55	4.683	Pu	9,061.0	1,661.0

<sup>a</sup>Calculated by ORIGEN computer program on Oct. 16, 1979, for a burnup of 30,000 MWd/MT and 911 days of decay (to Nov. 2, 1976).

<sup>b</sup>Calculated amounts in the cladding are  $2.086 \times 10^5$  g of zirconium per metric ton and 2.480 g of tin per metric ton of initial uranium.

<sup>c</sup>Initial uranium content of 30.5 cm (12 in.) of fuel rod was calculated to be 183.3 g. Initial enrichment was 2.561%.

Table 18. Principal radioactive components of H. B. Robinson fuel after 911 days decay<sup>a</sup>

Isotope	Amount per metric ton of initial uranium (g)	Amount per 30.5-cm (12-in.) length of fuel rod (mg)
<sup>83</sup> Kr	35.93	6.586
<sup>84</sup> Kr	98.97	18.14
<sup>85</sup> Kr	17.69	3.243
<sup>86</sup> Kr	163.7	30.01
<sup>90</sup> Kr	432.7	79.13
<sup>95</sup> Zr <sup>b</sup>	$3.227 \times 10^{-3}$	$5.915 \times 10^{-4}$
<sup>106</sup> Ru	30.15	5.526
<sup>110m</sup> Ag	$7.329 \times 10^{-2}$	$1.343 \times 10^{-2}$
<sup>125</sup> Sb <sup>b</sup>	7.478	$1.371 \times 10^{-2}$
<sup>129</sup> I	169.0	30.98
<sup>128</sup> Xe	2.88	$5.28 \times 10^{-1}$
<sup>130</sup> Xe	11.46	2.101
<sup>131</sup> Xe	394.5	72.31
<sup>132</sup> Xe	987.9	181.1
<sup>134</sup> Xe	1330.0	243.8
<sup>136</sup> Xe	2157.0	395.4
<sup>134</sup> Cs	46.12	8.454
<sup>137</sup> Cs	1027.0	188.2
<sup>144</sup> Ce	35.73	6.549
<sup>147</sup> Pm	68.74	12.60
<sup>154</sup> Eu	30.52	5.594

<sup>a</sup>Calculated by ORIGEN computer program on Oct. 17, 1979, for a burnup of 30,000 MWd/MT and 911 days of decay (to Nov. 2, 1976).

<sup>b</sup>Fission product activity only. Activation of cladding components produced significant amounts of <sup>95</sup>Zr, <sup>95</sup>Nb, <sup>119m</sup>Sn, <sup>123</sup>Sn, <sup>125</sup>Sb, <sup>125m</sup>Te, and <sup>60</sup>Co.

Table 19. Comparison of H. B. Robinson-2 fuel inventory calculations<sup>a</sup>

Isotope	Reference inventory (g/MT)	October 1979 ORIGIN inventory; 30,000 MWd/MT (g/MT)	July 1979 ORIGIN inventory; 31,360 MWd/MT (g/MT)	Reference inventory 1979 ORIGIN average
<sup>134</sup> Cs	48.60 <sup>b</sup>	46.12	49.57	1.016
<sup>129</sup> I	185.5 <sup>c</sup>	169.0	175.9	1.076
<sup>85</sup> Kr	18.97 <sup>c</sup>	17.69	18.52	1.048
<sup>106</sup> Ru	32.34 <sup>c</sup>	30.15	31.50	1.049
<sup>125</sup> Sb	5.40	7.478	7.768	0.708 <sup>d</sup>
<sup>137</sup> Cs	1138.0 <sup>c,e</sup>	1027.0	1073.0	1.084 <sup>e</sup>

<sup>a</sup>Inventory calculated for 911 days decay (to Nov. 2, 1976).

<sup>b</sup>Measured in dissolved H. B. Robinson-2 fuel.

<sup>c</sup>Calculated by ORIGIN in November 1976 for 31,360 MWd/MT burnup.

<sup>d</sup>If the gamma-measured values for <sup>125</sup>Sb listed in Table 16 are used instead of the 1979 ORIGIN average, the ratio (reference inventory/inventory by gamma counting) is 1.428.

<sup>e</sup>Cesium-137 was not used in making the fission product release calculations.

## 9. REFERENCES

1. R. A. Lorenz et al., Fission Product Release from Highly Irradiated Fuel, NUREG/CR-0722 (ORNL/NUREG/TM-287R2), February 1980.
2. R. A. Lorenz, J. L. Collins, and S. R. Manning, Fission Product Release from Simulated LWR Fuel, NUREG/CR-0274 (ORNL/NUREG/TM-154), October 1978.
3. R. A. Lorenz et al., Behavior of Iodine, Methyl Iodide, Cesium Oxide, and Cesium Iodine in Steam and Argon, ORNL/NUREG/TM-25, July 1976.
4. J. L. Collins et al., Knudsen Cell-Mass Spectrometer Studies of Cesium-Urania Interaction, ORNL/NUREG/TM-24, June 1976.
5. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1979, NUREG/CR-0917 (ORNL/NUREG/TM-332), August 1979.
6. R. A. Lorenz, J. L. Collins, and A. P. Malinauskas, "Fission Product Source Terms for the Light Water Reactor Loss-of-Coolant Accident," Nucl. Technol. 46(3): 404-410 (1979).
7. P. E. MacDonald to A. P. Malinauskas, Letter Report, "Transmittal of CPL Assembly B05 Axial Flux Measurements," MacD-67-75, Sept. 11, 1975.
8. K. R. Perkins to L. B. Thompson, "Thermal Transients of the Carolina Power and Light Fuel Bundle in Air," KRP-475, July 14, 1975.
9. S. M. Gehl, L. R. Kelmen, et al., Light-Water-Reactor Safety Research Program: Quarterly Progress Report July-September 1975, ANL-75-72, pp. 42-44.
10. A. A. Bauer, L. M. Lowry, and J. S. Perrin, Progress in Evaluating Strength and Ductility of Irradiated Zircaloy During July Through September 1975, BMI-1938, September 1975.
11. D. L. Hagrman in Quarterly Technical Progress Report on Water Reactor Safety Programs Sponsored by the Nuclear Regulatory Commission's Division of Reactor Safety Research, January-March 1977, TREE-NUREG-1128, June 1977, pp. III-65 to III-70.
12. T. B. Burgoyne and A. Garlick, "The Influence of Corrosion on the Emissivity of Zircaloy Cladding," Proceedings of Specialist Meeting on the Behavior of Water Reactor Fuel Elements Under Accident Conditions, Session II, Paper No. 21, CSNI Report No. 13, March 1977.
13. T. M. Besmann and T. B. Lindemer, "Chemical Thermodynamics of the System Cs-U-Zr-H-I-O in the Light Water Reactor Fuel-Cladding Gap," Nucl. Technol. 40: 297-305 (1978).

14. A. D. Aleckseeva, "The  $\text{Cs}_2\text{O-SiO}_2$  System," Russian J. Inorg. Chem. 2(5): 626-29 (1966).
15. P. Hofmann and C. Politis, Chemical Interaction between  $\text{UO}_2$  and Zry-4 in the Temperature Range between 900 and 1500°C, paper prepared for the 4th International Conference on Zirconium in the Nuclear Industry, Stratford-upon-Avon, England, June 1978.
16. S. L. Seiffert and R. R. Hobbins, Oxidation and Embrittlement of Zircaloy-4 Cladding from High Temperature Film Boiling Operation, NUREG KR-0517 (TREE-1327), April 1979, p. 30.
17. L. D. Noble, "An Exposure Dependent Diffusion Model for Fission Gas Release - A Progress Report from the ANS 5.4 Working Group," Proceedings of the ANS Topical Meeting on Light Water Reactor Fuel Performance, April 1979, UC-78, August 1979.
18. B. R. T. Frost, "Studies of Irradiation Effects," pp. 225-43 in Proceedings of the International Symposium on Ceramic Nuclear Fuels, May 3-8, 1969, Washington, D. C., Nuclear Division of the American Ceramic Society, 1969.
19. W. Chubb, V. W. Storhok, and D. L. Keller, "Observations Relating the Mechanisms of Swelling and Gas Release in Uranium Dioxide at High Temperatures," J. Nucl. Mater. 44: 136-52 (1972).
20. J. A. Turnbull and C. A. Friskney, "The Relation Between Micro-structure and the Release of Unstable Fission Products During High Temperature Irradiation of Uranium Dioxide," J. Nucl. Mater. 71: 238-48 (1978).
21. H. Zimmerman, "Investigations on Swelling and Fission Gas Behavior in Uranium Dioxide," J. Nucl. Mater. 75: 154-61 (1978).
22. H. Zimmerman, Untersuchungen zum Schwellen und Spaltgasverhalten in Oxidischem Kernbrennstoff Unter Neutronenbestrahlung, KFK 2467, June 1977.
23. J. Rest, GRASS-SST: A Comprehensive, Mechanistic Model for the Prediction of Fission-gas Behavior in  $\text{UO}_2$  Base Fuels During Steady-State and Transient Conditions, NUREG/CR-0202 (ANL-7853), June 1978.
24. M. O. Tucker, "Relative Growth Rates of Fission-Gas Bubbles on Grain Faces," J. Nucl. Mater. 78: 17-23 (1978).
25. S. M. Gehl, M. G. Seitz, and J. Rest, Fission-Gas Release from Irradiated PWR Fuel During Simulated PCM-Type Accidents: Progress Report, NUREG/CR-0088 (ANL-77-80), May 1978.
26. S. M. Gehl, Argonne National Laboratory, personal communication, July 1980.

27. R. A. Lorenz, "ANS-5.4 Fission Gas Release Model. III. Low Temperature Release," Proceedings of the ANS Topical Meeting on Light Water Reactor Fuel Performance, April 1979, August 1979.
28. C. Alexander, Oak Ridge National Laboratory, private communication to R. A. Lorenz (April 1979).
29. J. H. Goode and R. G. Stacy, Head-End Reprocessing Studies with H. B. Robinson-2 Fuel, ORNL/TM-6037, June 1978, pp. 58-65.

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