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Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January–March 1979

A. P. Malinauskas

Prepared for the
U.S. Nuclear Regulatory Commission
Division of Reactor Safety Research
Office of Nuclear Regulatory Research
Under Interagency Agreements DOE 40-551-75 and 40-552-75

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QUARTERLY PROGRESS REPORT ON FISSION PRODUCT BEHAVIOR IN LWRs FOR THE PERIOD JANUARY-MARCH 1979

A. P. Malinauskas, Program Manager

R. A. Lorenz
J. L. Collins
M. F. Osborne
R. L. Towns

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OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
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FOREWORD

This report documents progress made during the period January-March 1979. Previous reports in the 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).

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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).

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SUMMARY

The four scheduled tests in the High Temperature Test Series have been completed. High-burnup fuel rod segments from the H. B. Robinson-2 reactor were used in this series. The releases obtained for ^{85}Kr, ^{134}Cs, and ^{129}I from the 30.5-cm-long induction-heated segments are summarized as follows:

Test No.	Temperature (°C)	Time period (min)	Percent of total inventory released		
			^{85}Kr	^{134}Cs	^{129}I
HT-1	1300	10	1.07 + ∿0.5[a]	0.112	0.165
HT-4	1400	0.4	2.8 + ∿1.5[a]	3.05	1.75
HT-2	1445	7	5.0 + ∿1.0[a]	4.82	2.35
HT-3	1610	3	8.3 + ∿1.0[a]	8.27	—

[a]Approximate amounts released while test specimen was being heated for cladding expansion.

The large increase in release between 1300 and 1400°C suggests that a mechanism other than normal diffusion from either the gap or the UO_2 matrix has become dominant. This release mechanism is probably the linkage of bubbles at grain boundaries to form tunnels connecting to open void spaces. Since only about half of the length of each specimen was at test temperature, the release values presented above should be approximately doubled in order to obtain a more realistic estimate of the percentage release from the 16.0-cm heated section.

1. INTRODUCTION

Current experimental work in this program is concerned with the quantitative characterization of fission product release from highly irradiated light water reactor (LWR) fuel under hypothetical accident conditions. The High Temperature Test Series, completed in this reporting

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period, explored a temperature range that could be attained if the cladding were to exceed the upper temperature range for a controlled loss-of-coolant accident (LOCA) but not reach meltdown. In addition, the Boiling Water Reactor Fuel Test Series was initiated. The irradiated fuel rods examined in this series have higher concentrations of cesium and iodine in the pellet-to-clad gap space than did the previously tested PWR rods.

2. HIGH TEMPERATURE TESTS

The high temperature tests were conducted with H. B. Robinson-2 pressurized water reactor (PWR) fuel that had been irradiated to ∿30,000 MWd/MT. The tests were conducted in a flowing steam-helium mixture with the cladding expanded and drilled in the manner described in Sect. 2.1. In contrast to the previous series of experiments, in which the duration of a test was predetermined, the experiments in this series were terminated when loss of induction coupling occurred as a result of rapid oxidation of the Zircaloy cladding.

The results for ^{129}I release in test HT-3, which was conducted at 1610°C for 3 min, have indicated greater release of iodine than cesium. Since this behavior is unexpected, several of the samples which were submitted for chemical analysis are being reanalyzed in order to confirm the initial results.

2.1 High Temperature Test 4 (1400°C)

Prior to conducting test HT-4, the test specimen (B-86 of fuel rod H-15) was expanded, as were other fuel segments in this experimental series,[1] to a uniform radial gap width of 380 μm (0.015 in.) along a 15.2-cm-long section at the middle. A 0.159-cm (0.0625-in.)-diam hole was then drilled through the cladding at the center to provide a simulated defect. 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 about 10 cm in length developed in the cladding; this caused rapid loss of induction coupling.

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The distributions of cesium and iodine in the apparatus are listed in Tables 1 and 2. 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. However, it is more reasonable to express the releases as percentages of the region heated to the test temperature for two reasons:

1. Only the central 15.2 cm (6 in.) of the segment was heated to 1400°C (the temperature at the ends was probably no higher than 1000°C).
2. Most of the release is from the center, isothermally heated region.

One can therefore double the above percentage release values in order to obtain release from the heated length.

Considering the magnitude of these releases and the short testing time involved (20 sec), it becomes evident that the "breakaway" release observed during tests HT-2 (1445°C) and -3 (1610°C)[2] occurs in the temperature range 1350 to 1400°C. This breakaway release mechanism is probably due to the release of fission products already accumulated at grain boundaries via gas bubble linkage to form release paths. The temperature 1350°C is tentatively taken as the threshold for this mechanism since in test HT-1 the fuel rod segment was maintained at 1350°C for 2 min (and at somewhat lower temperatures for 8 min), yet was observed to release no more than the initial gap inventory of cesium and iodine (i.e., about 0.3% of total rod inventory). Moreover, diffusion from the UO_2 matrix could not account for the large releases, even at 1400°C.[1]

The data presented in Table 1 indicate that 98.4% of the released cesium remained in the quartz furnace tube, which is typical. In addition, an axial scan of the gamma activity along the furnace tube revealed that the cesium was deposited primarily within a 5-cm (2-in.) region downstream from the defect opening; this suggests that the bulk of the release occurred prior to the axial cracking of the cladding.

From an examination of the data listed in Table 2, it is clear that most of the iodine deposited in the furnace tube (59%), the thermal gradient tube (19%), and the first filter paper (22%); none was transported to the impregnated charcoal. These data tend to affirm the postulate

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Table 1. Distribution of cesium in High Temperature Test 4[a]

Location	Temperature (°C)	Amount of ^{134}Cs found in each location: (μg)[b]	Percent of total[c]	Percent of released	Total Cs found (μg)
Fuel rod segment	1400	(8837)[d]			(4.65×10^{5})[d]
Furnace tube					
Quartz liner	e	178.49	2.020	66.15	9395.37
Quartz fuel rod holder		86.94	0.984	32.22	4576.35
Thermal gradient tube	750-235	2.45	0.028	0.91	128.96
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		8.1×10^{-7}	9.1×10^{-9}	3.0×10^{-7}	4.3×10^{-5}
Charcoal No. 1a		3.8×10^{-7}	4.3×10^{-9}	1.4×10^{-7}	2.0×10^{-5}
Charcoal No. 1b		2.7×10^{-7}	3.1×10^{-9}	1.0×10^{-7}	1.4×10^{-5}
Charcoal No. 1c		7.6×10^{-7}	8.6×10^{-9}	2.8×10^{-7}	4.0×10^{-5}
Charcoal No. 2a		2.7×10^{-7}	3.1×10^{-9}	1.0×10^{-7}	1.4×10^{-5}
Charcoal No. 2b		2.5×10^{-7}	2.8×10^{-9}	9.2×10^{-8}	1.3×10^{-5}
Charcoal No. 3		0.0	0.0	0.0	0.0
AgX		1.5×10^{-4}	1.7×10^{-6}	5.6×10^{-5}	7.9×10^{-3}
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		269.83	3.054	100.00	14203.33

[a]Steam flow rate, 925 cm^3/min (STP); helium flow rate, 785 cm^3/min (STP); pressure, 760 torr. Decay time, 911 days (to November 2, 1976).

[b]Amounts less than 1.0×10^{-7} μg are given as 0.0.

[c]Percent of radioactive nuclide in fuel rod segment.

[d]Calculated for burnup of 30,500 MWd/MT of original uranium, 183.3 g of uranium originally in 12-in. segment, and 911 days decay.

[e]Approximately 950°C maximum at center and 600°C at outlet end.

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Table 2. Distribution of iodine in High Temperature Test 4[a]

Location	Temperature (°C)	Amount of ^{129}I found in each location: (μg)[b]	Percent of total[c]	Percent of released	Total iodine found (μg)
Fuel rod segment	1400	(3.37×10^{4})[d]			(4.28×10^{4})[d]
Furnace tube	e	345.72 ± 3.10	1.03	58.59	438.76
Thermal gradient tube	750-235	112.84 ± 2.61	3.35×10^{-1}	19.12	143.21
Filter pack components	120				
Stainless-steel inlet fitting		3.39 ± 0.06	1.01×10^{-2}	0.58	4.30
First filter paper		126.92 ± 3.47	3.77×10^{-1}	21.51	161.08
Second and third filter papers		1.17 ± 0.01	3.47×10^{-3}	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		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		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		590.04 ± 9.25	1.75	100.00	748.83 ± 11.74

[a]Steam flow rate, 925 cm^3/min (STP); helium flow rate, 785 cm^3/min (STP); pressure, 760 torr. Decay time, 911 days (to November 2, 1976).

[b]Amounts less than 0.02 µg are given as 0.0.

[c]Percent of radioactive nuclide in fuel rod segment.

[d]Calculated for burnup of 30,500 MWd/MT of original uranium, 183.3 g of uranium originally in 12-in. segment, and 911 days decay.

[e]Approximately 950°C maximum at center and 600°C at outlet end.

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that the iodine which was released from the test segment was in a form other than elemental iodine (probably CsI).

Approximately 1% (129 µg) of the cesium deposited in the thermal gradient tube in a manner displayed in Fig. 1; 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 1 and 2 indicate that similar amounts of cesium (129 µg) and iodine (143 µg) deposited in the thermal gradient tube. These data suggest that the cesium and iodine deposited in the thermal gradient tube were primarily present as CsI.

The temperature chronology of test HT-4 is presented graphically in Fig. 2, along with the deposition of ^{134}Cs in the thermal gradient tube and filter pack and the collection of ^{85}Kr in the cold charcoal traps. A total of 38.4 mCi of ^{85}Kr was collected; this amounts to 2.8% of the total inventory in the 12-in.-long segment. It is estimated that an additional 1.5% of the ^{85}Kr was released during the pretest cladding expansion procedure.

The deposition of europium in the thermal gradient tube, which is also presented in Fig. 1, had not been observed in previous tests. The deposition profile is unusual, in that it shows essentially uniform deposition at temperatures below 450°C but no deposition at all in the high temperature region. A near-uniform deposition profile could occur if the material were particulate. Moreover, thermophoresis would inhibit deposition in the high temperature region only as long as the surface remained hotter than the flowing gas.

In experiments of this type, europium is not usually observed to separate from the fuel. On the contrary, ^{154}Eu is frequently used as a tracer for fuel particles and dust, which are normally released only as ejected material from pressure-ruptured fuel rods. In test HT-4, ^{154}Eu was distributed as follows: 4.32 µg on the furnace tube liner, 2.19 µg on the fuel rod holder, 0.23 µg in the thermal gradient tube, 0.08 µg on the stainless-steel inlet fitting, and 1.41 µg on the first filter paper. The amount of ^{154}Eu found on the filter paper was significantly greater

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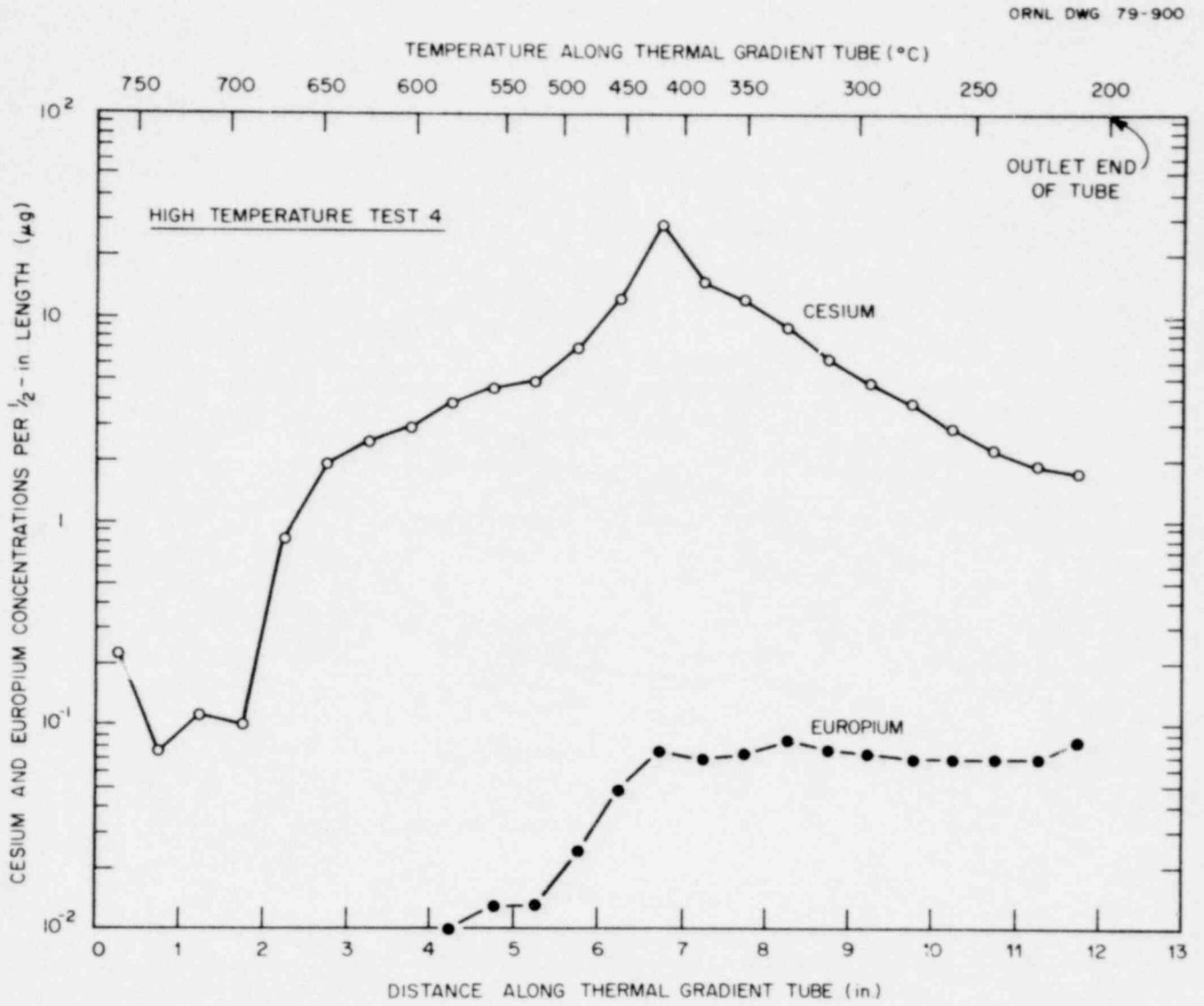

Fig. 1. Distribution of cesium and europium collected in the thermal gradient tube during test HT-4.

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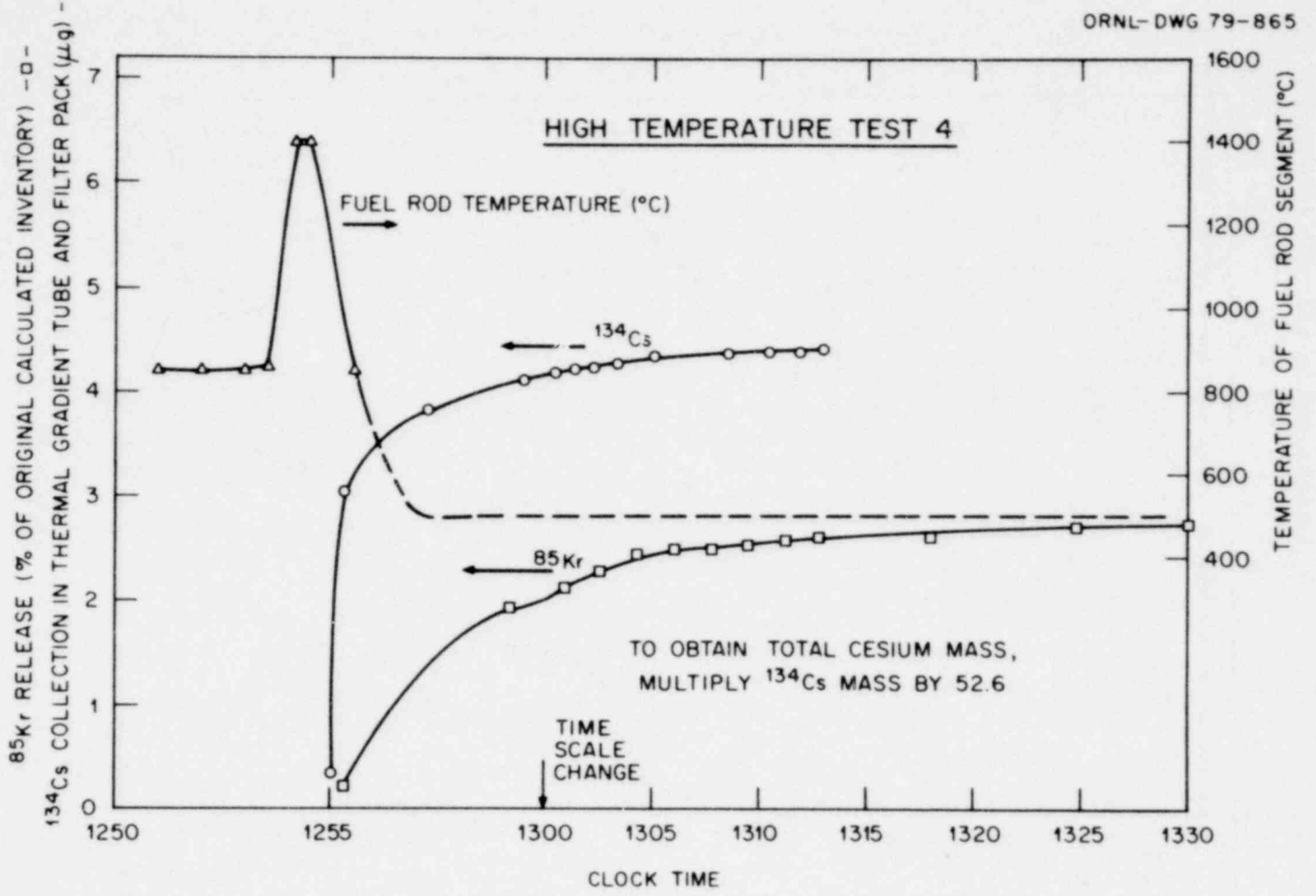

Fig. 2. Collection of ^{134}Cs in the thermal gradient tube and filter pack and of ^{85}Kr in the charcoal traps during test HT-4.

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than that expected on the basis of the amount of uranium found in the filter paper. The chemical and physical forms of the europium released in this test are therefore uncertain.

3. TESTS WITH BOILING WATER REACTOR FUEL

Fission product release tests with Boiling Water Reactor (BWR) fuel (the BWR Fuel Test Series) have begun. In this series of tests, segments cut from a fuel rod that was irradiated during cycle 1 of the Peach Bottom-2 Reactor are being used to measure fission product release throughout the temperature range 850 to 2200°C. This fuel rod was selected because it is believed to be representative of LWR fuel rods which have relatively high concentrations of cesium and iodine in the pellet-to-clad gap space. Our preliminary source-term model[3] correlates fractional release with gap inventory raised to the 0.8 power; previous tests with irradiated fuel were conducted with low gap-inventory fuel.

3.1 Characteristics of the Peach Bottom-2 Reactor Fuel

The fuel used in the BWR Test Series is commercial fuel which was irradiated during cycle 1 of the Peach Bottom-2 Reactor.[4] The dimensions and other characteristics of this fuel are summarized in Table 3. An axial scan of radioactivity in the energy range 0.55 to 0.75 MeV, 2.25 years after shutdown, is shown in Fig. 3.[5] This should provide a good representation of the distribution of radioactive cesium. Segment numbers correspond to approximately 30.5-cm (12-in.) lengths which were cut to form test samples. Segment 11 was dissolved, and the burnup was estimated to be 7730 MWd/MT.[6] The burnup values of the other segments were assumed to be in proportion to the measured gamma activity; these values are listed in Table 4. A calculation for the Peach Bottom irradiation conditions was performed using an updated (April 9, 1979) version of ORIGEN.[7] The resulting inventory of fission products for a burnup of 13,000 MWd/MT and a 3-year decay period is given in Tables 5 and 6.

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Table 3. Characteristics of Peach Bottom-2 Reactor fuel used in BWR Test Series

Rod serial number:	DG-2986	
Assembly data:	Assembly type 1, rod type 1, location F-6 in assembly PH-006	
Dimensions:	Zircaloy-2	1.430 cm (0.563 in.) outer diameter
		1.267 cm (0.499 in.) inner diameter
	Pellet	1.237 cm (0.487 in.) diameter
	Initial radial gap	0.015 cm (0.0059 in.)
	Plenum length	40.64 cm (16.0 in.)
Fuel:	UO_2 density	10.42 g/cm^3
	Uranium enrichment	1.33%
	UO_2 stack density	10.34 g/cm^3
	UO_2 stack length	365.8 cm (144.0 in.)
	UO_2 stack mass	4548 g
	U stack mass	4009 g
Irradiation:	Cycle 1 (January 12, 1974 to March 26, 1976)	
	Core average burnup, 10,000 MWd/MT	
	Rod DG-2986 average burnup, ∿9860 MWd/MT	
	Rod DG-2986 peak burnup in 30.5-cm length, ∿13,000 MWd/MT	

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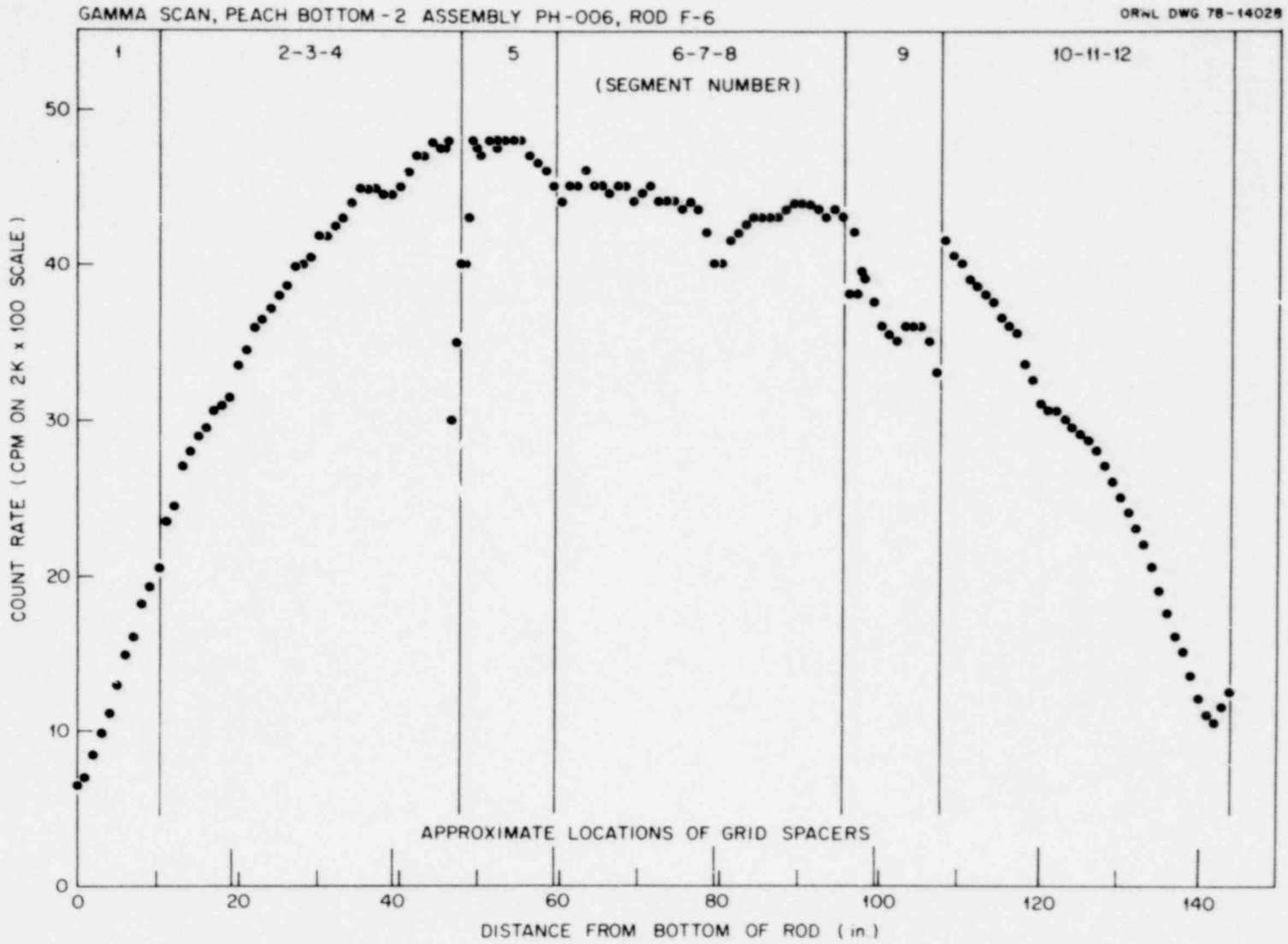

Fig. 3. Gamma scan of rod F-6 from bundle PH-006, Peach Bottom-2 Reactor.

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Table 4. Axial distribution of burnup and fission gas release for Peach Bottom-2 Reactor fuel

Segment No.	Relative burnup[a]	Estimated burnup (MWd/MT)	Estimated fission gas release (% of segment inventory)	Estimated fission gas release (% of total rod inventory)
1	0.395	3,890	0.2	0.006
2	0.870	8,780	0.9	0.064
3	1.152	11,350	6.2	0.595
4[b]	1.293	12,740	13.1	1.410
5[b]	1.319	13,000	1[illegible]	1.656
6[b]	1.259	12,410	11.4	1.192
7[b]	1.196	11,790	7.9	0.789
8	1.217	11,990	8.9	0.899
9	1.041	10,260	3.2	0.276
10	1.050	10,350	3.4	0.294
11	0.785	7,730	0.4	0.022
12	0.424	4,170	0.2	0.006
Rod average	1.000	9,860		7.20

[a]Obtained from gamma scan.

[b]Gamma scan indicated small peaks of activity between some pellets.

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Table 5. Amounts of principal fission product elements in peak burnup region of Peach Bottom-2 Reactor fuel[a]

Element	Mass		Element	Mass	
	g/MT	mg/30.5 cm		g/MT	mg/30.5 cm
Br	9.838	3.287	Te	239.6	80.05
Kr	153.1	51.15	I	124.6	41.63
Rb	142.8	47.71	Xe	2493.0	832.9
Sr	348.8	116.5	Cs	1248.0	416.9
Y	186.8	62.41	Ba	674.9	225.5
Zr	1538.0	513.8	La	557.8	186.4
Mo	1561.0	521.5	Ce	1087.0	363.1
Tc	376.7	125.8	Pr	532.5	177.9
Ru	1078.0	360.1	Nd	1812.0	605.4
Rh	278.2	92.94	Sm	396.5	132.5
Pd	740.6	247.4	Eu	69.27	23.14
Sn	47.27	15.79	U	976,600.0	326,270.0[b]
Sb	13.65	4.560	Pu	7155.0	2390.0

[a]Calculated by ORIGEN computer program for 13,000-MWd/MT burnup and 3-year decay period.

[b]Initial uranium content of 30.5 cm (12 in.) of fuel was calculated to be 334.1 g.

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Table 6. Principal radioactive components of high-burnup region of Peach Bottom-2 fuel[a]

Isotope	Amount per MT of initial uranium		Amount per 30.5-cm length of fuel rod	
	Ci	g	Ci	mg
^{85}Kr	3,295	8,394	1.101	2.804
^{90}Sr	27,980	205.1	9.348	68.52
^{90}Y	27,980	0.0514	9.349	0.1072
^{95}Zr[b]	5.61	2.61×10^{-5}	1.87×10^{-3}	8.72×10^{-6}
^{95}Nb[b]	12.47	3.18×10^{-4}	1.17×10^{-3}	1.06×10^{-4}
^{106}Ru	43,810	13.09	14.64	4.373
^{106}Rh	43,810	1.23×10^{-5}	14.64	4.11×10^{-6}
^{110m}Ag	90.71	1.91×10^{-2}	3.03×10^{-2}	6.38×10^{-3}
^{125}Sb[b]	3,901	3.777	1.303	1.262
^{125m}Te[b]	951.9	5.28×10^{-2}	0.318	1.76×10^{-2}
^{127}Te	7.911	3.00×10^{-6}	2.64×10^{-3}	1.00×10^{-6}
^{129}I	0.01664	94.20	5.56×10^{-6}	31.47
^{134}Cs	18,310[c]	14.14[c]	6.117[c]	4.724[c]
^{137}Cs	45,690	525.0	15.26	175.4
^{137m}Ba	43,220	8.03×10^{-5}	14.44	2.68×10^{-5}
^{144}Ce	41,130	12.89	13.74	4.306
^{144}Pr	41,130	5.44×10^{-4}	13.74	1.82×10^{-4}
^{144m}Pr	493.6	2.72×10^{-6}	0.165	9.09×10^{-7}
^{147}Pm	38,850	41.89	12.98	13.99
^{154}Eu	3,261	12.07	1.089	4.032

[a]Calculated by ORIGEN computer program for 13,000-MWd/MT burnup and 3-year decay period.

[b]Fission product activity only. Additional quantities of these isotopes will be produced in the Zircaloy cladding.

[c]These values are probably 25% high based on experimentally measured ratios of $^{134}Cs/^{137}Cs$, and assuming ^{137}Cs to be correct.

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3.2 Fission Gas Release While in the Peach Bottom-2 Reactor

Fuel rod DG-2986 was punctured and the contents of the plenum and void spaces analyzed by EG&G Idaho.[8] The results of the analysis are presented in Table 7. The total void volume was measured to be 74.0 cm^3; this compares well with the volume calculated from the data given in Table 3: plenum volume = 51.2 cm^3, gap volume = 21.6 cm^3, and pellet end void volume = 3.3 cm^3, for a total of 76.1 cm^3.

Table 7. Analysis of gas removed from plenum and void spaces of Peach Bottom-2 fuel rod DG-2986

Component	Fraction of total (mole %)	Volume[a] (cm^3, STP)
H_2	<0.1	<0.2
He	43.6	79.1
N_2	0.1	0.2
O_2	<0.1	<0.2
Ar	1.1	2.0
CO_2	<0.1	<0.2
Kr	5.4	9.8
Xe	49.7	90.2
Total		181.4 $\pm$ 1.8 cm^3

[a]Rod void volume measured at 74.0 $\pm$ 1.0 cm^3.

Using the ORIGEN-calculated inventory and assuming a rod average burnup of 9860 MWd/MT, we calculate that the 100.0 cm^3 of noble gas (krypton and xenon) is equivalent to 7.20% release of the total inventory to the void spaces and plenum while in the Peach Bottom-2 Reactor. Releases of individual isotopes of xenon and krypton are given in Table 8.

It is necessary to estimate the fission gas release from each individual segment since these release values are used, in turn, to obtain estimates of the cesium and iodine gap inventories. To obtain these estimates, we used a previously determined correlation of fission gas

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Table 8. Fission gas released to plenum of Peach Bottom-2 rod DG-2986

Isotope	Amount found in plenum[a]			Total produced; ORIGEN calculation[b] (g-atoms/MT)	Amount released to plenum (%)
	Percent of element	cm^3 (STP)	g-atoms MT		
^{83}Kr	12.99	1.269	0.0141	0.1788	7.89
^{84}Kr	29.77	2.907	0.0324	0.4179	7.75
^{85}Kr	5.81	0.567	0.0063	0.0749	8.41
^{86}Kr	51.42	5.021	0.0559	0.6920	8.08
Total Kr		9.764	0.1087	1.367	7.95
^{128}Xe	0.02	0.018	0.0002	0.006	3.23
^{130}Xe	0.10	0.090	0.0010	0.024	4.13
^{131}Xe	9.84	8.871	0.0988	1.367	7.22
^{132}Xe	19.09	17.211	0.1917	2.809	6.83
^{134}Xe	28.11	25.343	0.2822	3.845	7.34
^{136}Xe	42.84	38.623	0.4301	6.042	7.12
Total Xe		90.156	1.0040	14.092	7.12
Total Xe + Kr		99.92	1.113	15.459	7.20

[a]Quantities calculated for 3-year decay period (to March 26, 1979).

[b]Calculated for 9860 MWd/MT by linear decrease from an ORIGEN computer inventory calculation for 13,000-MWd/MT burnup.

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release as a function of linear heat rating and irradiation time.[9] The percentage fission gas release predicted by this method is shown graphically in Fig. 4. We assumed that the axial linear power distribution in rod DG-2986 was the same as the burnup distribution presented in Table 4. In order to obtain the total fission gas release of 7.20%, it was necessary to assume that segment 5 operated at 423 W/cm, with the other segments at proportionally lower heat ratings. The gas release for each heat rating was obtained from the curve in Fig. 4 and is listed in column 4 of Table 4. From the burnup in each segment, we then calculated the percentage of the total fission gas release resulting from each segment; these values are listed in column 5.

The heat ratings shown in Fig. 4 are significantly higher than the average for this rod while in the Peach Bottom-2 Reactor. Moreover, it is probable that the power variations in the reactor resulted in considerably more fission gas release than was experienced during the constant power operation of the fuel rods which formed the basis of the correlation shown in Fig. 4.[9] However, this difference is not important, since only the relative fission gas release as a function of linear power is involved in the estimation procedure.

3.3 Test BWR-1 (970°C)

Segment 5 (as identified in Fig. 3) was used in test BWR-1, the first in the Boiling Water Reactor Fuel Test Series. The experimental apparatus was the same as that employed in previous test series.[10] Fuel rod segments in the BWR series were capped with specially manufactured, all-Zircaloy ferrule fittings in order to eliminate leaks due to differential thermal expansion.

After installation in the apparatus, the test segment was pressurized at room temperature three times to 400 psi with purified helium, each time allowing 10 min for mixing before venting in order to remove residual air. After this operation, the segment was heated to 500°C for 30 min, then to 750°C. At this point, the rod was pressurized to 180 psig with helium. The temperature of the test piece was then increased by induction heating

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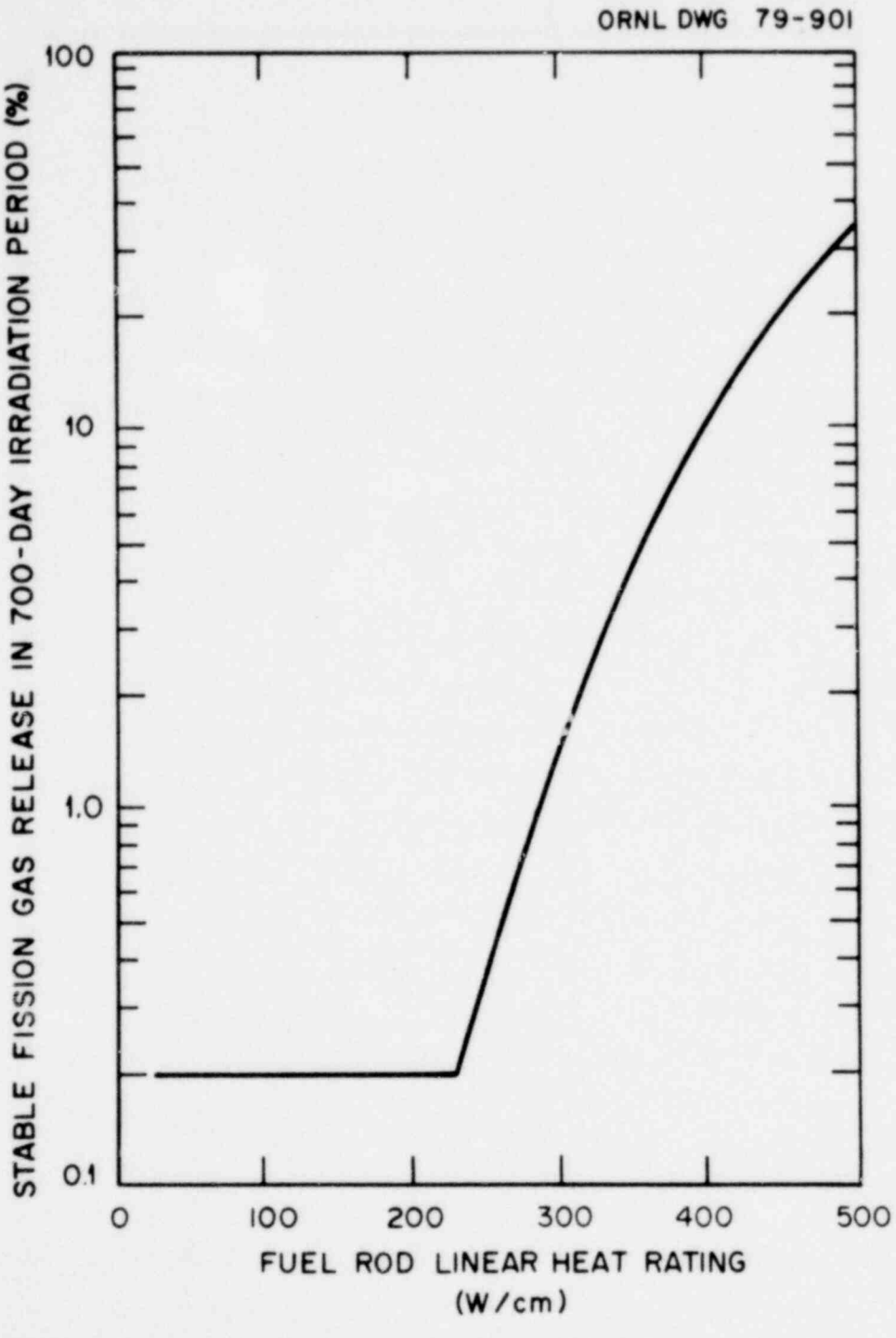

Fig. 4. Stab' fission gas release estimated as a function of linear heat rating.

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while in a flowing steam-helium atmosphere. The cladding ruptured at 970°C; after 1 min at 970°C, induction heating was terminated.

The distribution of cesium released in the test is summarized in Table 9. Approximately 1.71% of the total cesium inventory was released. An additional amount, about 0.01%, was released with fuel dust which was ejected at the time of cladding rupture. The distribution of cesium that had deposited in the thermal gradient tube is shown graphically in Fig. 5. The peak in the deposition profile lies between 400 and 450°C; this is typical of both cesium and iodine deposition, as noted in the previous High Burnup Fuel Test Series.

The cesium released from the test piece in this experiment may be compared with that released during test HBU-7,[11] in which a test segment of H. B. Robinson-2 Reactor fuel was ruptured at 900°C in a steam-helium atmosphere and maintained at that temperature for 1 min. In test HBU-7, only 0.029% of the total cesium invertory was released. Although the 60-fold larger release of cesium inventory in test BWR-1 is believed to be partly a result of the higher rupture temperature and a slightly larger volume of vented gas, the primary effect is probably due to the higher inventory of cesium in the pellet-clad gap space (at 900 to 970°C, only the cesium already in the gap space can escape from the ruptured cladding in a short-term test in steam). The release of cesium to the gap space of the fuel rod segment used in BWR-1 may have been as high as 20 to 40% of the total cesium produced. This contrasts with the HBU-7 test segment, which contained only 0.3% of the total cesium in the gap space. The approximately 100-fold difference in gap inventories (on a percentage basis) is therefore expected to account for most of the 60-fold difference in measured cesium release.

The release of ^{85}Kr in test BWR-1 was approximately 1.7% of the total inventory. In test HBU-7, the corresponding release was 1.0%.

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Table 9. Distribution of cesium in Test BWR-1[a]

Location	Temperature (°C)	Amount of ^{137}Cs found in each location: (μg)[b]	Percent of total[c]	Percent of released	Total cesium found (μg)
Fuel rod segment	970	(1.754×10^{5})[d]			(4.169×10^{5})[d]
Furnace tube					
Quartz liner	e	2210.71	1.26	73.55	5255.79
Quartz fuel rod holder		653.07	3.72×10^{-1}	21.73	1552.25
Thermal gradient tube	750-220	64.64	3.69×10^{-2}	2.15	153.64
Filter pack components	125				
Stainless-steel inlet fitting		10.08	5.75×10^{-3}	0.34	23.96
First filter paper		67.15	3.84×10^{-2}	2.23	159.61
Second and third filter papers		4.44×10^{-4}	2.53×10^{-7}	1.48×10^{-5}	0.01
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. 3		0.0	0.0	0.0	0.0
AgX		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		3005.65	1.71	100.00	7145.70

[a]Steam flow rate, 1806 cm^3/min (STP); helium flow rate, 205 cm^3/min (STP); pressure, 760 torr. Decay time, 3 years (to March 26, 1979). Some additional cesium was released in the form of UO_2 fuel dust.

[b]Amounts less than 1.0×10^{-5} μg are given as 0.0.

[c]Percent of radioactive nuclide in fuel rod segment.

[d]Calculated for 13,000-MWd/MT burnup of original uranium, 334 g of uranium originally in 12-in. segment, and 3-year decay period.

[e]Approximately 750°C maximum at center and 600°C at outlet end.

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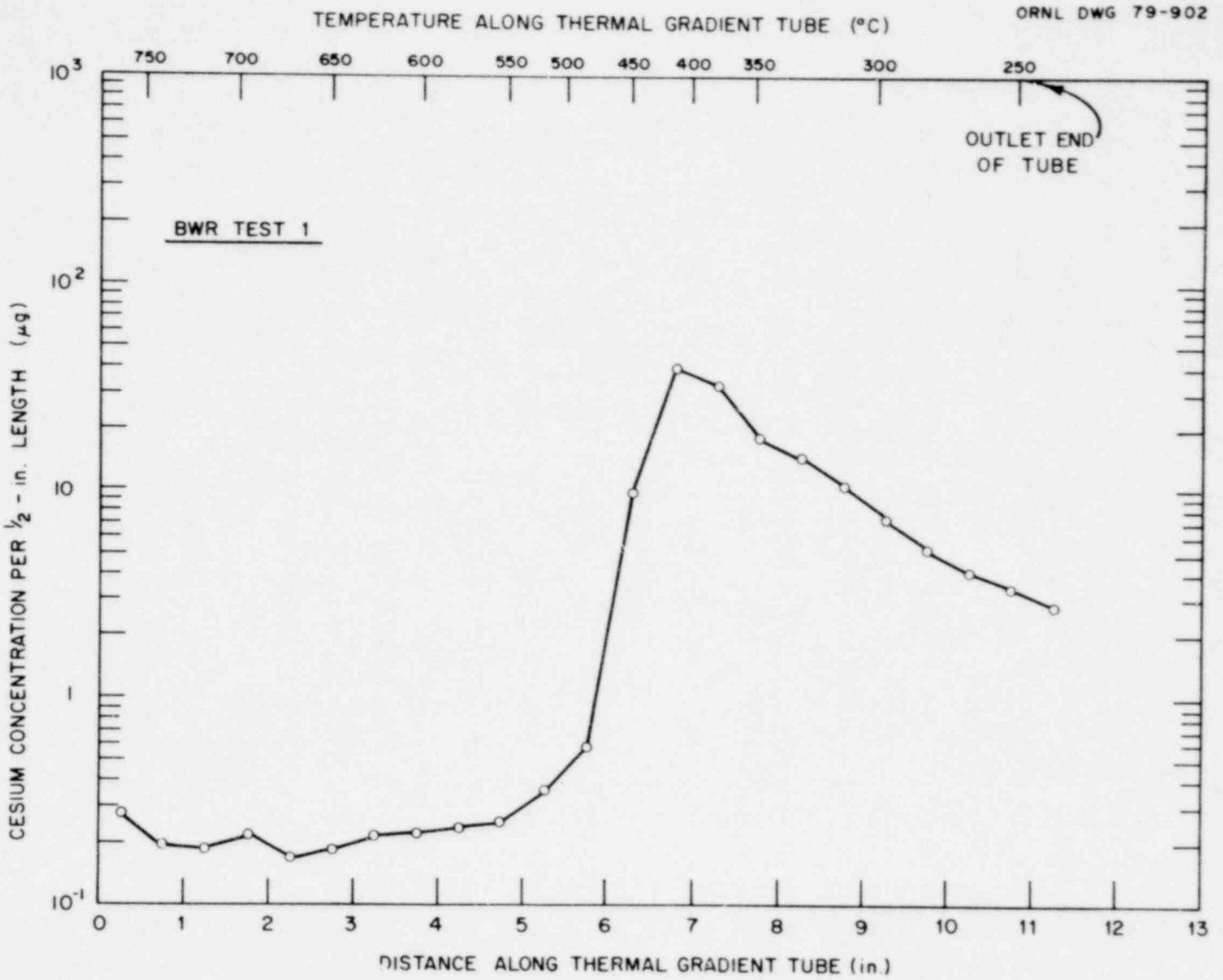

Fig. 5. Distribution of cesium collected in the thermal gradient tube during test BWR-1.

4. REFERENCES

1. 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).

2. 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).

3. 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).

4. N. H. Larsen, Core Design and Operating Data for Cycles 1 and 2 of Peach Bottom 2, EPRI NP-563 (June 1978).

5. R. L. Lines, ORNL, personal communication, June 28, 1978.

6. D. O. Campbell, ORNL, personal communication, April 1979.

7. A. G. Croff, ORNL, personal communication, April 9, 1979.

8. V. W. Storhok, EG&G Idaho, personal communication, March 12, 1979.

9. R. A. Lorenz and G. W. Parker, "Calculation of Amount of Radioactivity in Fuel Rod Void Spaces," in Nuclear Safety Program Annual Progress Report for Period Ending December 31, 1967, ORNL-4228 (April 1968).

10. 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).

11. 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).

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