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Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April–June 1979

A. P. Malinauskas

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QUARTERLY PROGRESS REPORT ON FISSION PRODUCT BEHAVIOR IN LWRs FOR THE PERIOD APRIL-JUNE 1979

A. P. Malinauskas, Program Manager

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

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FOREWORD

This report documents progress made during the period April-June 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).

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SUMMARY

Tests BWR-2 and BWR-3 were performed during this report period; each test employed segment 4 from fuel rod F-6, bundle PH-006, which had been irradiated to ~12,700 MWd/MT during cycle 1 of the Peach Bottom-2 boiling water reactor (BWR). The test segment was pressure-ruptured at 850°C in test BWR-2 and later heated at 1200°C for 25 min in flowing steam in test BWR-3. The results obtained to date in the BWR Test Series can be summarized as follows:

	Test conditions		Percent of total inventory release			
Test	Rupture	Diffusion	^{85}Kr	^{137}Cs	^{129}I	UO_2
BWR-1	960°C at 1.23 MPa (179 psi)	960°C for 1 min	1.7[a]	1.71	1.18	0.02
BWR-2	850°C at 3.40 MPa (500 psi)	850°C for 1 min	1.0[b]	1.70	2.54	0.02
BWR-3	c	1200°C for 25 min	1.1[d]	1.85	—	0.00

[a]An additional amount, estimated at 15.1% of the total inventory, was released from this fuel rod segment during operation in the Peach Bottom-2 Reactor (ref. 1).

[b]An additional amount, estimated at 13.1% of the total inventory, was released from this fuel rod segment during operation in the Peach Bottom-2 Reactor.

[c]This segment was the same one that was ruptured at 850°C in test BWR-2.

[d]This amount of ^{85}Kr is in addition to the 1.0% released during test BWR-2 and the 13.1% estimated to have been released while in the Peach Bottom-2 Reactor.

1. INTRODUCTION

The BWR Fuel Test Series utilizes 30.5 cm-long fuel rod segments cut from rod F-6, bundle PH-006, which had been irradiated during cycle 1 of the Peach Bottom-2 Reactor. This fuel rod, whose characteristics have been described previously,[1] was selected because it is believed to be representative of light water reactor fuel rods which have relatively high concentrations of cesium and iodine in the pellet-to-clad gap space. Our preliminary source-term model[2] correlates mass release with gap inventory raised to the 0.8 power; however, this correlation is largely based on earlier tests which were conducted with irradiated fuel rod segments having low gap inventories.[3]

2. TEST BWR-1 (960°C RUPTURE)

Additional results have been obtained for test BWR-1. Reexamination of the temperature measurements showed that the rupture temperature was 960°C rather than 970°C as previously reported.[1] Based on the analysis of ^{129}I, 1.18% of the total idoine was released (see Table 1). The peak deposit of iodine in the thermal gradient tube corresponded to the location of the cesium peak. Only 0.36% of the released iodine reached the charcoal cartridges, thus indicating a very low release of the reactive forms of iodine. The amounts of ^{137}Cs collected in the thermal gradient tube and filter pack, and of ^{85}Kr collected in the charcoal traps, are presented graphically in Fig. 1 as a function of time and fuel rod temperature.

Some cesium and iodine were also released in the form of fuel dust that was ejected at the time of rupture; 119 μg of ^{137}Cs was measured in loose fuel particles tapped off the furnace tube liner. Analyses of ^{154}Eu as a tracer for fuel indicated that these particles contained 44.4 mg of UO_2. Since this amount of fuel should have contained only 21 μg of ^{137}Cs, it appears that the ejected fuel dust may have been enriched in cesium as a result of its proximity to the pellet surface. The total amount of ejected dust, as determined from ^{154}Eu concentration measurements, amounted to 69 mg of UO_2.

3. TEST BWR-2 (850°C RUPTURE)

Test BWR-2 was performed by pressurizing segment 4 of rod F-6[1] to 515 psig (3.54 MPa) with purified argon while holding it at a temperature of ~600°C in the test apparatus; the rod was then heated to 850°C to cause rupture of the Zircaloy cladding in a flowing steam atmosphere. The

Table 1. Distribution of iodine in test BWR-1[a]

Location	Temperature (°C)	Amount of ^{129}I found in each location: Mass (μg)[b]	Percent of total[c]	Percent of released	Total Iodine found (μg)
Fuel rod segment	960	(3.147×10^4)[d]			(4.163×10^4)[d]
Furnace tube	e	169.73 ± 3.90[f]	5.39×10^{-1}	45.69	224.53
Thermal gradient tube	750-220	78.61 ± 0.13	2.50×10^{-1}	21.16	103.99
Filter pack components	125				
Stainless steel inlet fitting		5.35 ± 0.02	1.70×10^{-2}	1.44	7.08
First filter paper		116.40 ± 1.70	3.70×10^{-1}	31.33	153.98
Second and third filter papers		0.07 ± 0.001	2.22×10^{-4}	0.02	0.09
Charcoal No. 1a		1.30 ± 0.01	4.13×10^{-3}	0.35	1.72
Charcoal No. 1b		0.03 ± 0.001	9.53×10^{-5}	0.01	0.04
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		371.49 ± 5.76	1.18	100.00	491.4 ± 7.6

[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 iodine was released in the form of UO_2 dust.

[b]Amounts less than $1.0 \times 10^3 \times 10^3$ 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 a 3-year decay period.

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

[f]Error estimates are based solely on the results of replicate chemical analyses.

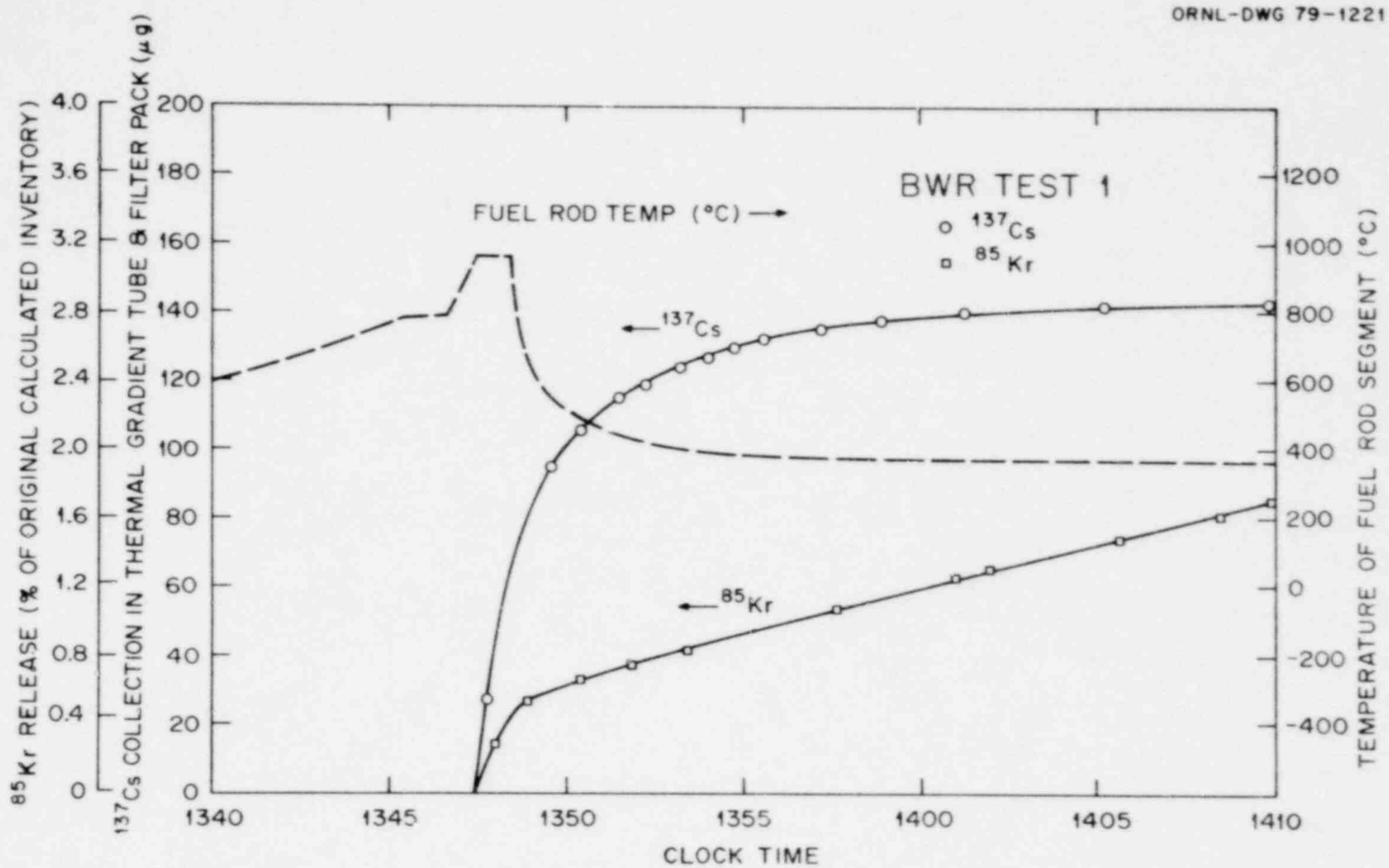

Fig. 1. Collection of ^{137}Cs in the thermal gradient tube and filter pack, and of ^{85}Kr in the charcoal traps, during test BWR-1.

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temperature was maintained for 1 min before turning off the induction heating. Figure 2 is a photograph of the ruptured rod. The amount of cladding expansion and the hole size (∿2 mm^2) are similar to those observed in previous experiments in the same apparatus.

The distributions of cesium and iodine which were collected in the different components of the apparatus are presented in Tables 2 and 3. These distributions were typical for pressure-ruptured tests. Approximately 1.70% and 2.54% of the total cesium and the total iodine inventories, respectively, were found.

The amounts of cesium collected in the thermal gradient tube and filter pack, and of ^{85}Kr collected in the charcoal traps, are illustrated graphically in Fig. 3 as a function of time and fuel rod temperature. The distributions of cesium and iodine in the thermal gradient tube are shown in Fig. 4. (Peak deposits in the 350 to 450°C temperature range are typical for both cesium and iodine in our tests.) The release of ^{85}Kr (11.4 mCi) amounted to 1.01% of the total inventory.

4. TEST BWR-3 (DIFFUSIONAL RELEASE AT 1200°C)

Test BWR-3 was performed to measure the release of fission products at 1200°C from a previously pressure-ruptured fuel rod segment. The rod used in this test was the same as that which was ruptured in test BWR-2. An electric resistance heater surrounding the furnace tube was substituted for the induction heater so that the entire 30.5-cm length could be heated nearly uniformly.

Approximately 1.85% of the total cesium inventory in the test piece was released. The distribution of cesium on the apparatus components is given in Table 4. The amounts of ^{137}Cs collected in the thermal gradient tube and filter pack, and the ^{85}Kr collected in the charcoal traps, are presented in Fig. 5 as a function of time and fuel rod temperature. Cesium and krypton releases occurred during both the slow heatup and the cooldown periods. The total effective time for cesium release at 1200°C was determined to be 25 min when the heatup and cooldown periods were taken into account. Measurements of gamma activity in the thermal gradient tube

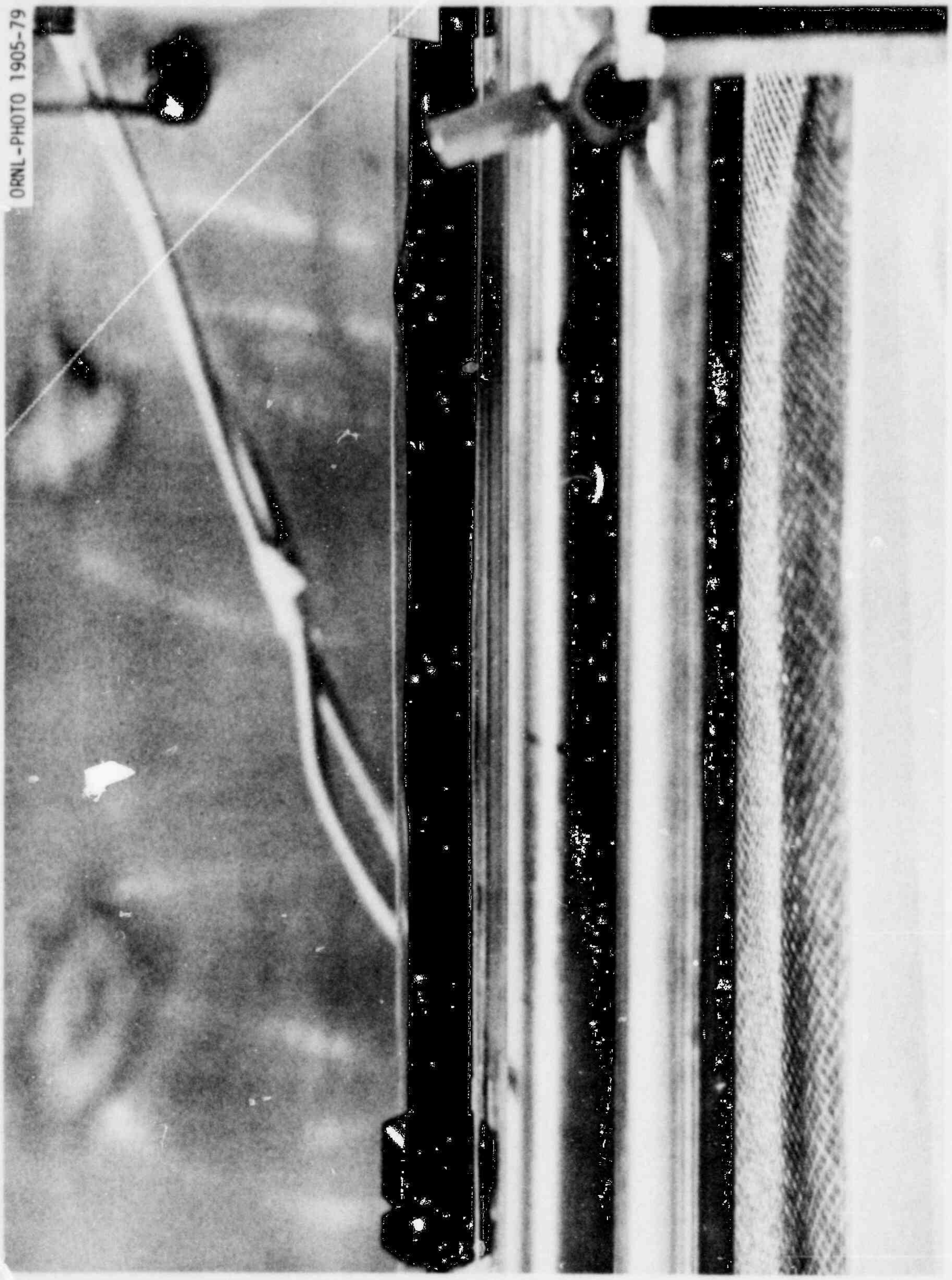

Fig. 2. View of ruptured fuel rod segment from test BWR-2.

Table 2. Distribution of cesium in test BWR-2[a]

Location	Temperature (°C)	Amount of ^{137}Cs found in each location: Mass (μg)[b]	Amount of ^{137}Cs found in each location: Percent of total[c]	Amount of ^{137}Cs found in each location: Percent of released	Total cesium found (μg)
Fuel rod segment	850	(1.714×10^5)[d]			(4.074×10^5)[d]
Furnace tube					
Quartz liner	e	2039.00	1.19	70.01	4846.5
Quartz fuel rod holder		447.30	0.26	15.36	1063.2
Thermal gradient tube	740-275	89.36	5.21×10^{-2}	3.07	212.40
Filter pack components	130				
Stainless steel inlet fitting		16.14	9.42×10^{-3}	0.55	38.36
First filter paper		320.104	0.19	10.99	760.85
Second filter paper		0.087	5.08×10^{-5}	0.003	0.21
Third filter paper		0.105	6.13×10^{-5}	0.004	0.25
Charcoal No. 1a		0.141	8.23×10^{-5}	0.005	0.34
Charcoal No. 1b		0.038	2.22×10^{-5}	0.0013	0.09
Charcoal No. 1c		0.016	9.33×10^{-6}	0.0005	0.04
Charcoal No. 2a		0.009	5.25×10^{-6}	0.0003	0.02
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		2912.3	1.699	100.00	6922.2

[a]Steam flow rate, 1375 cm^3/min (STP); helium flow rate, 245 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 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 12,740-MWd/MT burnup of original uranium, 334 g of uranium originally in 12-in. segment, and a 3-year decay period.

[e]Approximately 650°C maximum at center; ∿500°C at outlet end.

Table 3. Distribution of iodine in test BWR-2[a]

Location	Temperature (°C)	Amount of ^{129}I found in each location: Mass (μg)[b]	Percent of total[c]	Percent of released	Total iodine found (μg)
Fuel rod segment	850	(3.084×10^4)[d]			(4.080×10^4)[d]
Furnace tube	e	571.09 ± 26.30[f]	1.852	73.05	755.46
Thermal gradient tube	740-275	47.72 ± 0.45	1.55×10^{-1}	6.10	63.13
Filter pack components	130				
Stainless steel inlet fitting		6.55 ± 0.22	2.12×10^{-2}	0.84	8.66
First filter paper		156.00 ± 6.00	5.06×10^{-1}	19.95	206.36
Second and third filter papers		0.22 ± 0.01	7.13×10^{-4}	0.03	0.29
Charcoal No. 1a		0.24 ± 0.01	7.78×10^{-4}	0.03	0.32
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		781.82 ± 32.98	2.535	100.00	1034.2 ± 43.6

[a]Steam flow rate, 1375 cm^3/min (STP); helium flow rate, 245 cm^2/min (STP); pressure, 760 torr. Decay time, 3 years (to March 26, 1979).

[b]Amounts less than 1.0×10^{-3} μg are given as 0.0.

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

[d]Calculated for 12,740-MWd/MT burnup of original uranium, 334 g of uranium originally in 12-in. segment, and a 3-year decay period.

[e]Approximately 650°C maximum at center; ~ 500°C at outlet end.

[f]Error estimates are based solely on the results of replicate chemical analyses.

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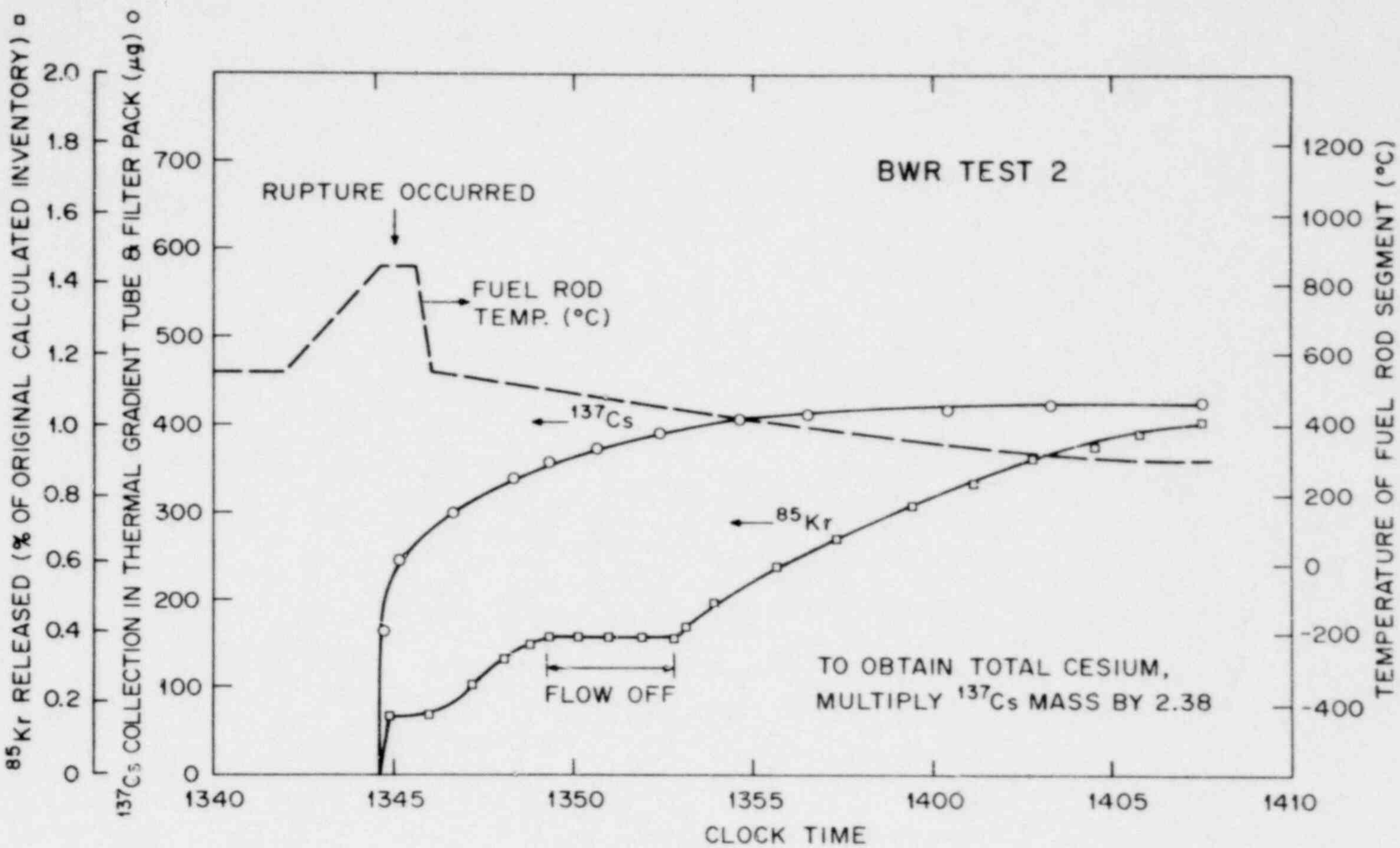

Fig. 3. Collection of ^{137}Cs in the thermal gradient tube and filter pack, and of ^{85}Kr in the charcoal traps, during test BWR-2.

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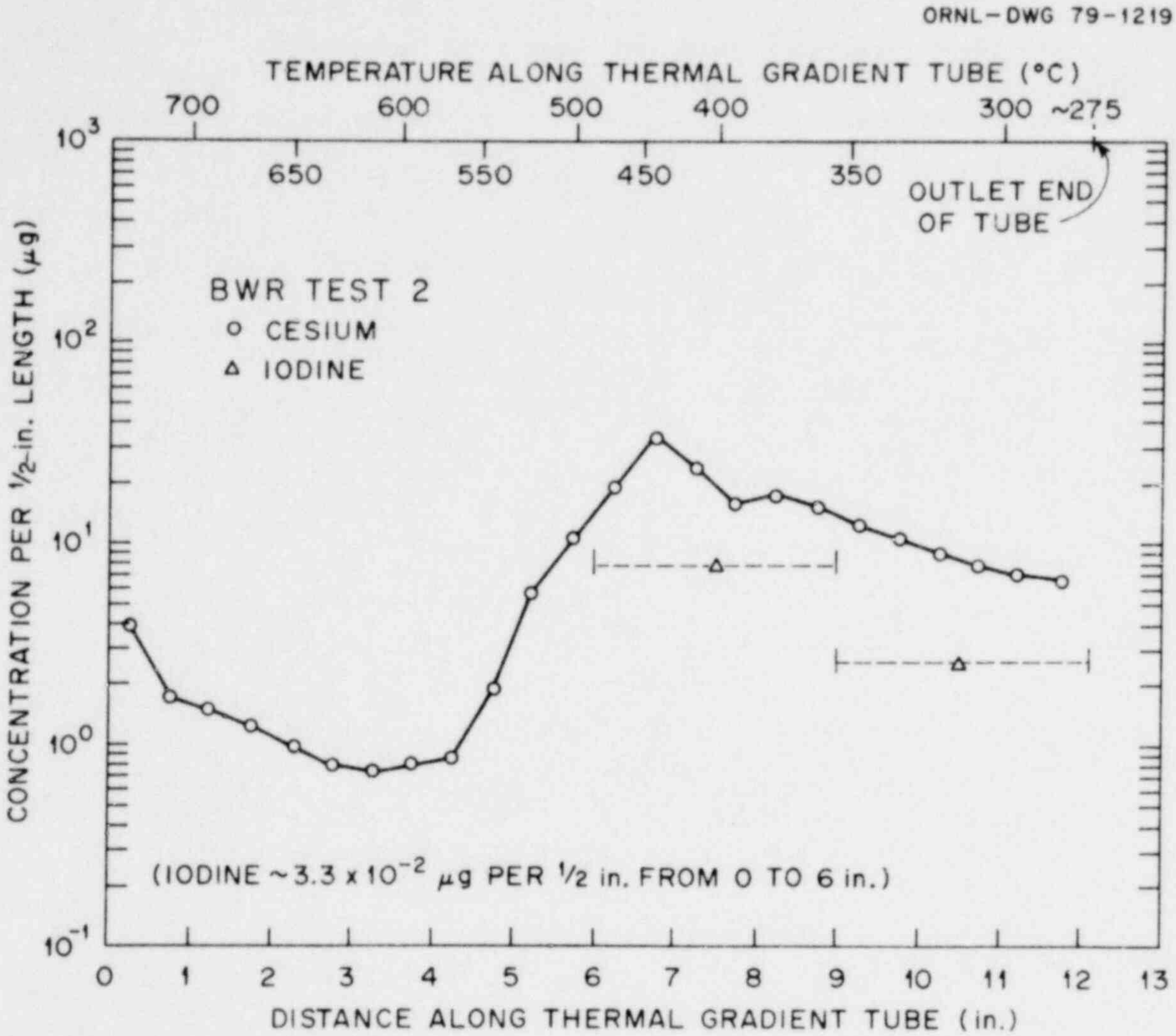

Fig. 4. Concentration profiles of cesium and iodine in the thermal gradient tube from test BWR-2.

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Table 4. Distribution of cesium in test BWR-3[a]

Location	Temperature (°C)	Amount of ^{137}Cs found in each location: Mass (µg)[b]	Percent of total[c]	Percent of release	Total cesium found (µg)
Fuel rod segment	1200	(1.685×10^5)[d]			(4.005×10^5)[d]
Furnace tube	1200				
Quartz liner		444.41	2.64×10^{-1}	14.29	1056.30
Quartz fuel rod holder		904.50	5.37×10^{-1}	29.09	2149.87
Thermal gradient tube	1050-200	765.82	4.55×10^{-1}	24.63	1820.24
Filter pack components	125				
Stainless steel inlet fitting		40.80	2.42×10^{-2}	1.31	96.98
First filter paper		954.11	5.66×10^{-1}	30.68	2267.78
Second filter paper		0.08	4.75×10^{-5}	2.57×10^{-3}	0.19
Third filter paper		0.05	2.97×10^{-5}	1.61×10^{-3}	0.12
Charcoal No. 1a		2.0×10^{-6}	1.19×10^{-9}	6.43×10^{-8}	4.65×10^{-6}
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		3109.77	1.846	100.00	7391.47

[a] Steam flow rate, 1000 cm^3/min (STP); helium flow rate, 300 cm^3/min (STP); pressure, 760 torr. Decay time, 3 years (to March 26, 1979). This fuel rod segment was also used in test BWR-2.

[b] Amounts less than 1.0×10^{-6} µg are given as 0.0.

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

[d] Calculated for burnup of 12,740 MWd/MT of original uranium, 334 g of uranium originally in 12-in. segment, and a 3-year decay period.

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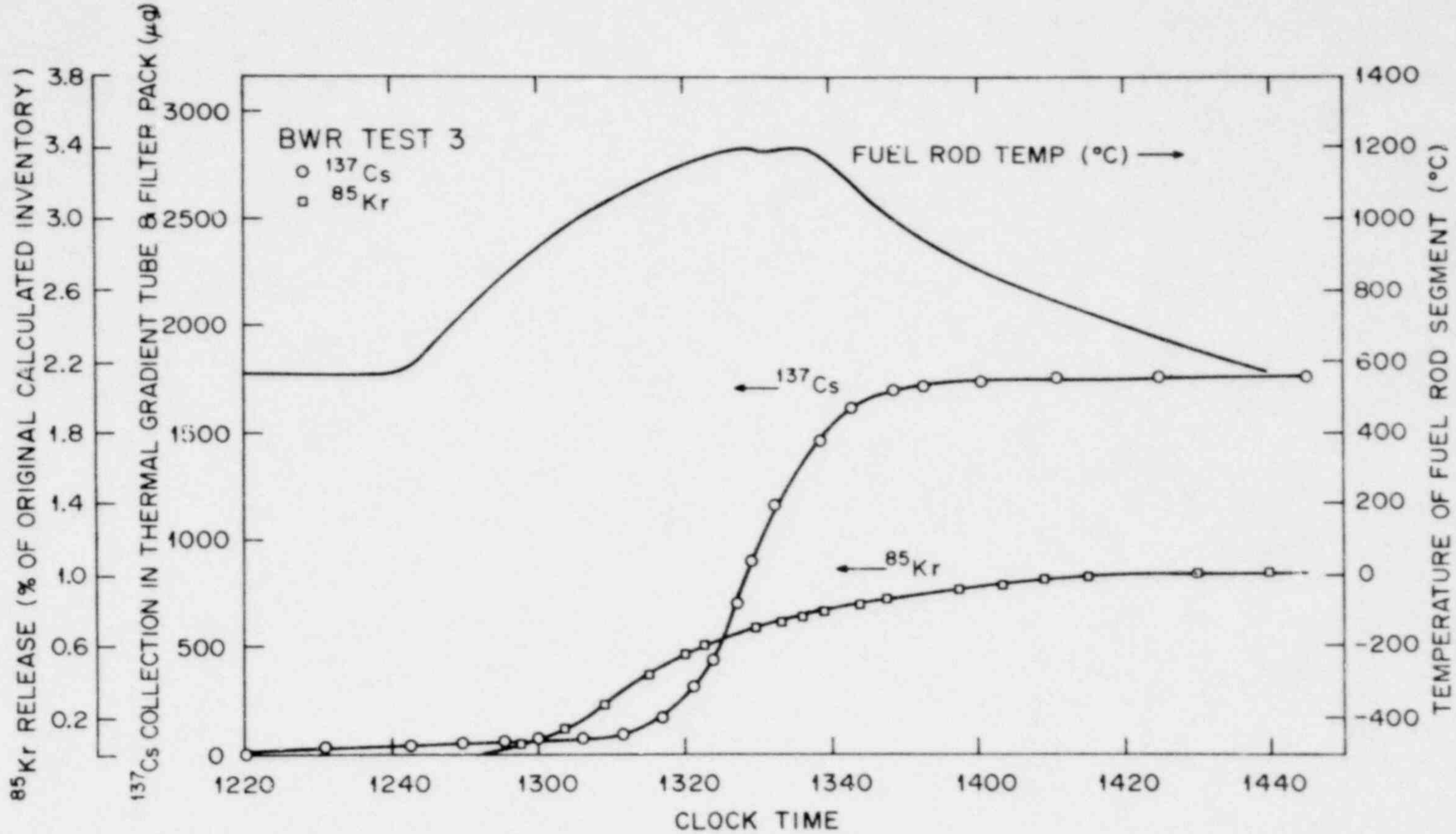

Fig. 5. Collection of ^{137}Cs in the thermal gradient tube and filter pack, and of ^{85}Kr in the charcoal traps, during test BWR-3.

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and filter pack indicated that half of the cesium was released in the 13-min period during which the fuel rod was at a temperature >1180°C.

The distribution of cesium in the thermal gradient tube is shown in Fig. 6. The small broad peak at 600 to 650°C is similar to the one that was observed in test HBU-11.[3] (Test HBU-11 was essentially identical to test BWR-3 except for the use of high-burnup PWR fuel from the H. B. Robinson-2 Reactor.)

Significant krypton release began when the temperature of the fuel rod reached about 900°C. (Note that this segment had previously been heated to a maximum of 850°C in test BWR-2.) The total releases of ^{85}Kr were 1.08% in test BWR-3, 1.01% in test BWR-2, and an estimated 13.1% while in the Peach Bottom-2 Reactor.

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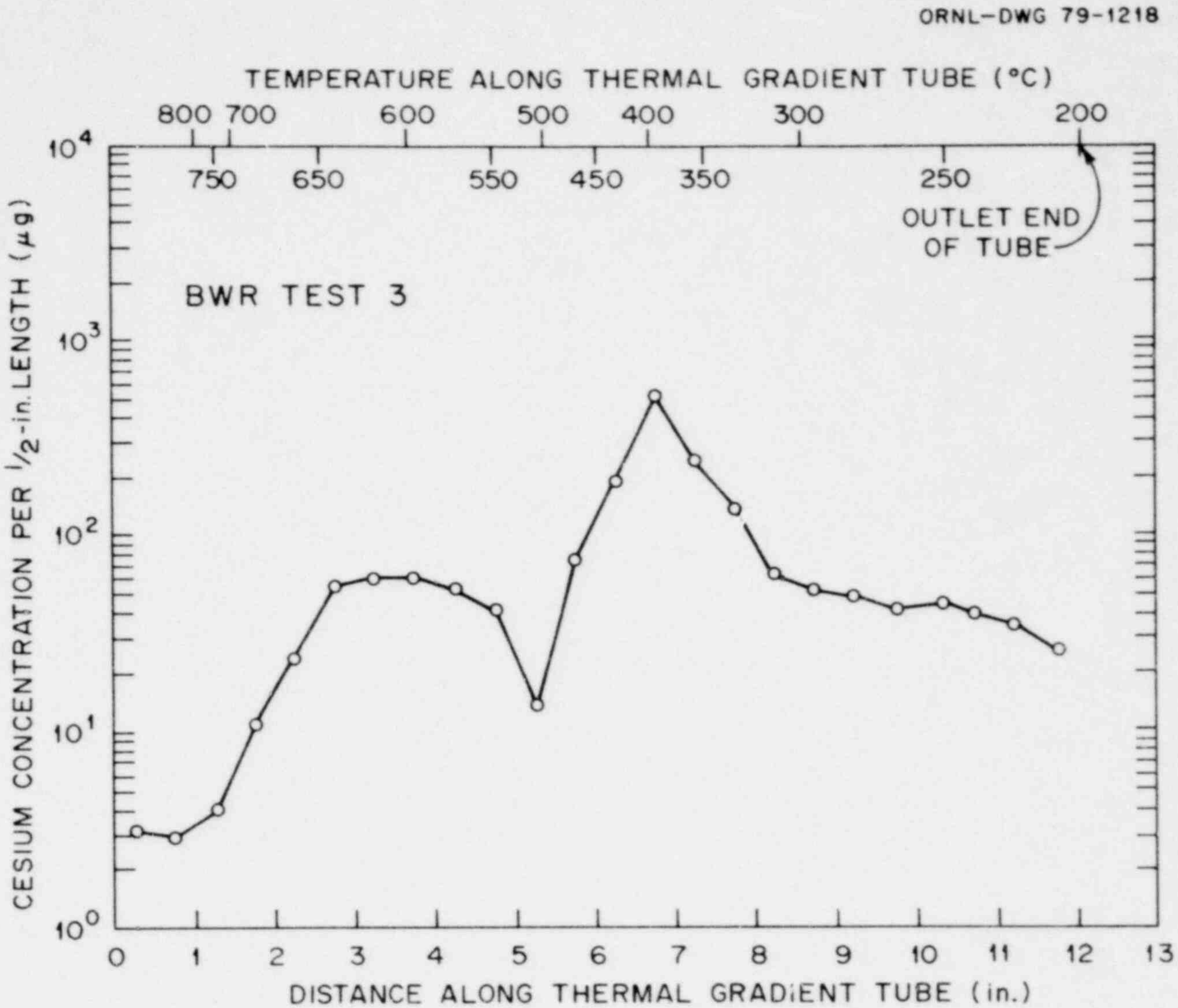

Fig. 6. Distribution of cesium in the thermal gradient tube during test BWR-3.

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