NUREG/CR-0370
ORNL/NUREG/TM-242

Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April–June 1978

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Prepared for 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

OAK RIDGE NATIONAL LABORATORY
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Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161

NUREG/CR-0370
ORNL/NUREG/TM-242
Dist. Category R3

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

QUARTERLY PROGRESS REPORT ON FISSION PRODUCT BEHAVIOR IN LWRs FOR THE PERIOD APRIL-JUNE 1978

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Manuscript Submitted: August 31, 1978

Date Published - September 1978

Prepared for the
U.S. Nuclear Regulatory Commission
Division of Reactor Safety Research
Office of Nuclear Regulatory Research
Washington, D.C. 20555
Under Interagency Agreements DOE 40-551-75 and 40-552-75

NRC FIN No. B0127

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FOREWORD

This report documents progress made during the period April-June 1978. 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).

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16. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Source Terms for the LWR Loss-of-Coolant Accident: Summary Report, NUREG/CR-0091 (ORNL/NUREG/TM-206)(June 1978).

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SUMMARY

Analysis of data obtained during High Burnup Fuel Test 12 (HBU-12) is complete; this test was conducted to determine the gap inventories of cesium and iodine by purging a selected H. B. Robinson fuel rod segment with purified helium. Total release of both species was virtually identical when expressed as percent of total inventory. In addition, it was possible to estimate the extent to which diffusion of cesium from the pellet matrix augmented the gap inventory of this species. Correction of the data for this contribution indicated that the gap inventory of cesium for this H. B. Robinson fuel rod amounted to 0.30% of total cesium inventory, as compared with a value of 0.25% which was inferred from determinations of xenon gap inventory made earlier.

Preparations are being made for the conduct of the High Temperature Test Series and for tests with Boiling Water Reactor (BWR) fuel from Peach Bottom-2. Gross gamma activity scans of the Peach Bottom fuel rod which will be employed in the latter tests indicate a buildup of cesium at pellet-pellet interfaces; this suggests that the gap inventory of cesium and iodine is significantly higher than that found in the H. B. Robinson rods which have been used in the High Burnup Fuel Test Series.

Two preliminary designs of the Fission Product Transport Test Facility (FPTTF) have been developed; one of these is for a facility which is capable of operation with steam at 1600°C, and the other with steam at 1000°C. Cost/benefit analyses were made which supported further development of only the 1000°C steam capability.

1.0 INTRODUCTION

Two different types of studies of fission product behavior in light water reactors (LWRs) comprise this program. One is concerned with establishing realistic source terms for selected radiotoxic species which are released from fuel rods that become defected in the course of a loss-of-coolant accident (LOCA) or a spent fuel transportation accident (SFTA). The second activity involves the design of a Fission Product Transport Test Facility (FPTTF) for use in developing and testing computational models of fission product transport and deposition along the primary circuit of LWRs during various postulated core meltdown accident sequences.

Much attention was given to establishing the operational constraints imposed on the FPTTF by state-of-the-art considerations attending the operational characteristics of various system components, and the extent to which these constraints compromise the usefulness of the facility. A superheater which was capable of delivering 1600°C steam to the test section was identified, and a facility limited only by this component was compared with one which was capable of operation to a maximum steam temperature of 1000°C. Greatly increased costs, both for construction and operation of the higher-temperature facility, could not be justified; accordingly, the design of a FPTTF only capable of operation with 1000°C steam is being developed.

A parameter of primary significance in determining fission product release from a defected fuel rod is the gap inventory, i.e., that amount of the total inventory of a fission product which is associated with the interconnected voids within the fuel rod. Previous work has assumed that the gap inventory in low-heat-rated fuel rods is sensibly constant for the long-lived noble gases, volatile species (iodine), and so-called "semi-volatile" species (e.g., cesium). Analysis of the data obtained from an experiment designed to test this assumption has been completed; the assumption appears to be valid for the H. B. Robinson rods which have been employed thus far in this program.

2.0 FISSION PRODUCT RELEASE FROM LWR FUEL

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2.1 High Burnup Fuel Test 12 - Additional Results

In High Burnup Fuel Test 12 (HBU-12), the annular pellet-to-clad gap and other interconnected voids within an H. B. Robinson fuel rod segment were swept with purified helium in order to determine the quantity of fission products, primarily cesium and iodine, that had accumulated in these regions during normal reactor operation. This potentially releasable "gap inventory" is important since it constitutes the principal source of fission product release from failed fuel rods in such events as loss-of-coolant accidents, control rod ejection accidents, fuel handling accidents, and spent fuel transportation accidents. It may also be the principal source of fission products released in an anticipated transient without scram (ATWS).

Details of the test procedure and results of cesium and krypton release have been reported previously.[1] Data concerning iodine release have since been obtained; these data are summarized with the cesium release data in Table 1. Most of the iodine was collected in the thermal gradient tube in the 300 to 400°C temperature range; this suggests deposition as CsI. Very little iodine was collected in the charcoal traps, which indicates an absence of volatile forms of iodine such as I_2, HI, and organic iodides.

Cesium deposition along the thermal gradient tube was characterized by a peak deposition which occurred at approximately 430°C.[1] Although the chemical form of the deposited cesium is unknown, either CsI or CsO_2 is suggested since elemental cesium would have been transported to cooler regions of the collection system.

The pressure of the purified helium purge as applied to the inlet end of the pellet-to-clad gap is depicted graphically in Fig. 1 along with fuel rod temperature and cumulative release of cesium. The radioactive isotopes ^{134}Cs and ^{137}Cs were monitored with a single-channel analyzer, and the concentrations subsequently normalized to the total mass of cesium released after the fuel rod was heated above 500°C. Because

Table 1. Distribution of cesium and iodine in High Burnup Fuel Test 12

Location	Temperature (°C)	Amount found in each location: Cesium, Percent of total[a]	Cesium, Mass (μg Cs)	Iodine, Percent of total[a]	Iodine, Mass[b] (μg I)
Fuel rod (original amounts)	c		$(4.382 \times 10^{5})^{d}$		$(4.315 \times 10^{4})^{d}$
Furnace tube	c				
Quartz liner		1.76×10^{-2}	77.3	1.47×10^{-4}	0.064 ± 0.065
Quartz holder		5.03×10^{-3}	22.1	2.21×10^{-5}	0.0095 ± 0.0005
Thermal gradient tube					
Quartz housing	950-200	2.18×10^{-2}	95.7	1.27×10^{-3}	0.55 ± 0.001
Gold liner: 0-4 in.	950-560	6.69×10^{-3}	29.3	2.32×10^{-4}	0.10 ± 0.05
4-6 in.	560-430	0.149	651.3	1.97×10^{-2}	8.50 ± 0.25
6-9 in.	430-280	0.189	830.5	3.35×10^{-1}	144.68 ± 35.5
9-12 in.	280-200	0.0434	190.3	4.15×10^{-2}	17.9 ± 0.8
Filter pack components	125				
Stainless steel inlet fitting		2.82×10^{-3}	12.4	5.29×10^{-3}	2.28 ± 0.3
First filter paper		1.99×10^{-2}	87.0	3.24×10^{-2}	13.96 ± 0.76
Second and third filter papers		1.27×10^{-8}	5.5×10^{-5}	1.94×10^{-4}	0.084 ± 0.003
Charcoal No. 1a				7.15×10^{-4}	0.31 ± 0.03
Total		0.455	1995.9	0.437	188.44 ± 37.8

[a]Percent of fission product element in fuel rod segment.

[b]Error estimated is based on counting statistics and agreement of duplicate samples.

[c]The temperature-time sequence was conducted as follows: 700°C for 52 min, 900°C for 65 min, 1100°C for 260 min, and 1200°C for 40 min.

[d]Calculated for burnup of 30,745 MWd/metric ton of original uranium, 175.7 g of uranium originally in 11.5-in. segment, and 911 days decay.

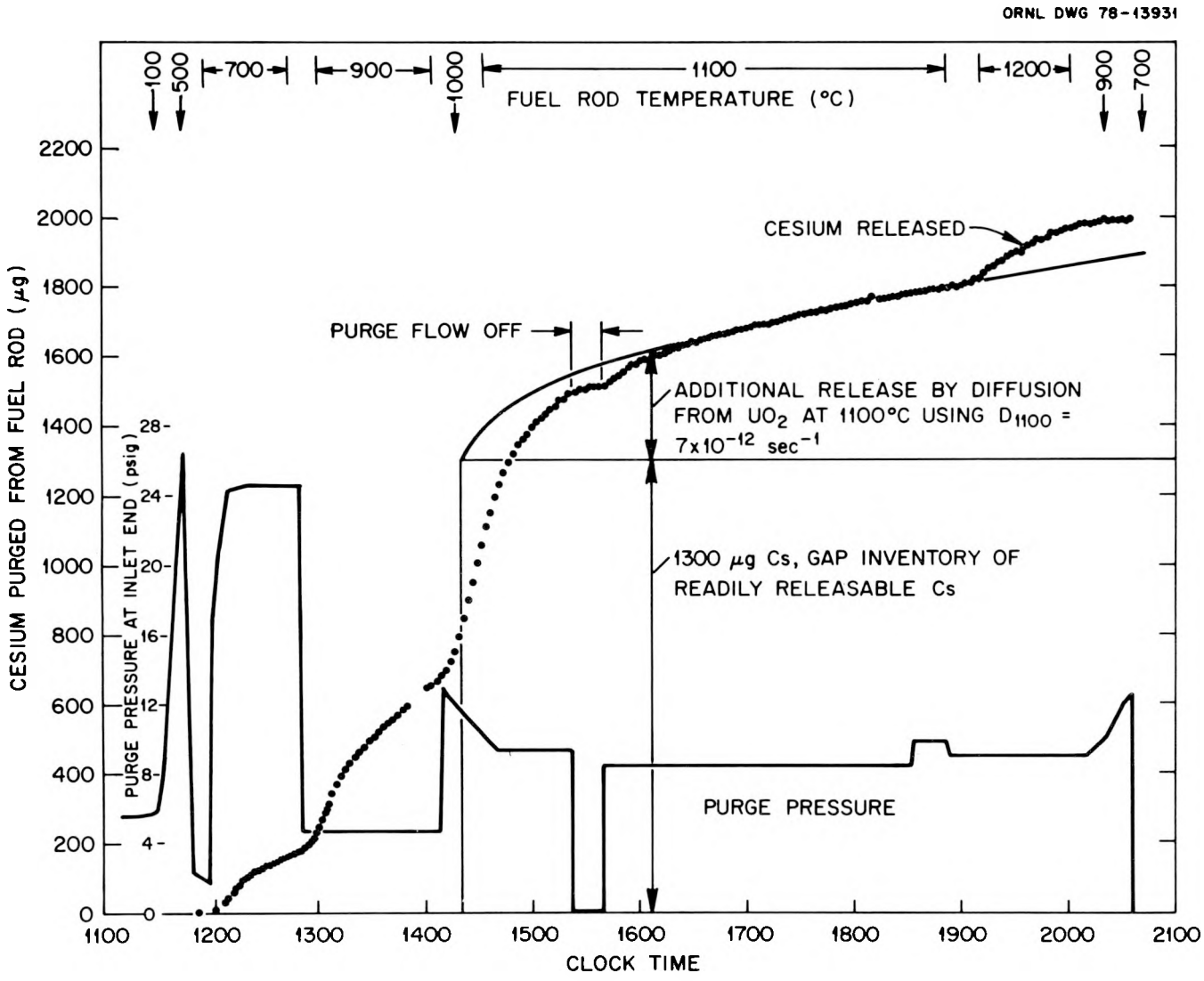

Fig. 1. Purge pressure and cesium release during test HBU-12.

of a leak which developed in the inlet-end stainless steel ferrule fitting as a result of thermal expansion, the flow rate of the helium could not be measured directly. During the heat-up to 500°C, however, it is believed that the leak was insignificant, so that all of the measured purge flow passed through the pellet-to-clad gap space. For this period of operation, it was possible to calculate values of the gap width from the purge flow data according to a method developed previously.[2,3] The results, given in Table 2, indicate that the radial gap width remained unchanged at about 40 μm. This is somewhat larger than that determined with other segments of H. B. Robinson fuel which were taken from bundle B05.[2,3]

By using the measured purge pressure and assuming that the gap size remained constant at higher temperatures, the purge flow rate was calculated for several test periods as listed in Table 3. The cesium purge rate was obtained from the data displayed in Fig. 1; the purge data were then employed to calculate corresponding values of cesium concentration. Since the helium purge rate was not measured directly, these concentrations are only approximate. However, it is clear that, as a given temperature was attained, the initial cesium concentration was greater than the later "stabilized" value. At 1100°C, the low cesium concentrations indicated gradual depletion of the available cesium "gap inventory."

Table 2. Calculated width of radial gap, Test HBU-12

Fuel rod temperature (°C)	Inlet end pressure[a] (atm)	Helium flow rate (cm^3, STP/min)	Helium viscosity (10^6 g/cm.sec)	Calculated gap width (μm)
70	1.38	63.5	220	43
200	1.73	76.3	267	41
300	2.10	90.2	305	41
400	2.49	104.3	342	41
500	2.76	124.2	376	43

[a]Outlet end pressure, 1.02 atm.

Table 3. Cesium purge rates during test HBU-12[a]

Time interval (clock time)	Temperature (°C)	Inlet pressure[b] (atm)	Helium flow rate[b] (cm^3/min, STP)	Cesium purge rate (µg Cs/min)	Cesium concentration[c] (µg Cs/cm^3 He)	Comments
1210-1220	700	2.66	69.7	6.0	0.086	Maximum rate at 700°C
1230-1246	700	2.67	70.1	2.1	0.030	Stabilized rate at 700°C
1300-1310	900	1.31	5.05	14.0	2.77	Maximum rate at 900°C
1330-1405	900	1.31	5.05	4.7	0.93	Stabilized rate at 900°C
1420-1430	1060	1.74	12.4	21.0	1.69	Maximum release rate
1430-1440	1100	1.67	10.2	19.0	1.86	Maximum rate at 1100°C
1750-1850	1100	1.61	8.66	0.83	0.096	Stabilized rate at 1100°C
1910-1930	1200	1.61	7.67	3.4	0.44	Maximum rate at 1200°C

[a]Assumes constant radial gap width of 41 µm.

[b]Outlet pressure ranged from 1.02 atm at 700°C to 1.14 atm at 1100°C (initial).

[c]Helium flow rate and cesium concentration are only approximate since helium flow rate could not be measured directly.

2.2 Diffusion of Cesium from the UO_2 Pellet Matrix

In test HBU-12 the concentration of cesium in the purge stream at 1100°C gradually decreased to a value less than that at 900°C, thus suggesting that the gap inventory of cesium was being depleted. Based on previous tests[4] with irradiated UO_2, additional cesium should diffuse from the UO_2 matrix at a slow but measurable rate.

Booth and Rymer[5] derived equations for diffusion from sintered UO_2 by assuming the pellets to be composed of an agglomeration of independent spheres. For uniform initial concentration of the diffusing species, the fraction released when the pellet was heated to a given temperature was estimated by

$$f = \frac{6}{(\pi)^{1/2}} (D't)^{1/2} , \qquad (1)$$

in which

f = fraction released by diffusion at time t (only for $f \leq 0.1$),

D' = diffusion parameter, D/a^2, sec^{-1},

D = diffusion coefficient, cm^2/sec,

a = radius of equivalent sphere, cm, and

t = time at temperature, sec.

During the 4.5-hr period at 1100°C, some of the cesium purged from the gap space may represent diffusional release from the UO_2 matrix. By trial and error, a diffusion parameter (D') value of 7.0×10^{-12} sec^{-1} was found to fit the observed release curve reasonably well if the initial gap inventory of cesium was taken to be 1300 µg.

The calculated amounts released by diffusion from the UO_2 matrix are listed in Table 4. The sum of the amount diffused and the initial gap inventory is plotted in Fig. 1 along with the measured release amounts, assuming that all of the cesium was released at the same rate as the ^{134}Cs and ^{137}Cs collected in the thermal gradient tube and filter pack. Clock time 1420 (fuel rod temperature, 1025°C) was chosen as the starting time for diffusional release. Obviously, the gap inventory (i.e., 1300 µg cesium) was not completely released by clock time 1420.

According to this simple two-component release model, the initial gap inventory of cesium was 1300 µg, an amount equivalent to about 0.30%

Table 4. Calculated amount of cesium released by diffusion at 1100°C

Time at 1100°C		Amount of cesium released[a]	
sec	hr	Fraction of total inventory, f	Mass (μg)
600	0.167	2.194×10^{-4}	96
3,600	1	5.374×10^{-4}	235
7,200	2	7.599×10^{-4}	333
10,800	3	9.307×10^{-4}	408
14,400	4	1.075×10^{-3}	471
18,000	5	1.202×10^{-3}	526
21,600	6	1.316×10^{-3}	577

[a]Calculated for $D' = 7.0 \times 10^{-12}$ sec^{-1}.

of the total cesium inventory in the segment. This can be compared with the in-reactor xenon release to the fuel rod voids and plenum of 0.25% that was measured at room temperature. (Heating to 1200°C during the HBU-12 test released an additional 1.36% of the fission gas.)

The best-fit diffusion parameter for cesium also compares reasonably well with that for fission gas release as determined by others during post-irradiation annealing tests. Parker et al.[4] determined for xenon a D' value at 1100°C of 2.1×10^{-12} sec^{-1} for trace-irradiated fuel, and approximately 1.2×10^{-11} sec^{-1} for fuel irradiated in the 1000- to 4000-MWd/MT burnup range. The ASTM-5.4 subcommittee[6] observed that $D'_{1000°C}$ for xenon ranged from 7.5×10^{-12} sec^{-1} to 8.3×10^{-11} sec^{-1} for LWR fuel irradiated mainly to burnups of 1000 to 10,000 MWd/MT. The relative rates of release of cesium and xenon from UO_2 have been observed to vary widely,[6] whereas post-irradiation annealing studies[4] indicate that the release of iodine from UO_2, as compared with xenon release, averages approximately 2.5 times greater, i.e., D' for iodine is about six times larger than the corresponding value for xenon.

For high-temperature, long-term exposures, the diffusion of cesium and iodine from the UO_2 matrix should probably be treated as a replenishment of the gap inventory and not as being directly released from the rod. This effect was demonstrated in test HBU-12 between clock times 1522 and 1539 when shutoff of the purge flow resulted in a large decrease in the rate of cesium escape from the test fuel rod segment.

2.3 The High Temperature Test Series

A series of seven tests is planned to investigate fission product release from highly irradiated light water reactor fuel in steam in the temperature range 1200 to 1600°C. (The maximum temperature actually attained will be determined by the extent to which the Zircaloy cladding will lose coupling with the induction heating coil because of oxidation by steam or reaction with the UO_2 fuel.) Eight segments, each 30.5 cm (12 in.) long, were prepared from longer sections of H. B. Robinson fuel rods D-12 and H-15 of bundle B05.

A cladding expansion rig has been designed to uniformly expand the cladding of the irradiated fuel segments to 1.138 cm (0.448 in.) diam over a 15.24-cm (6-in.) length (the length of the fuel rod that is induction-heated to near-uniform temperature). This will create a radial gap width of about 380 μm (0.015 in.). The expansion will be performed by pressurizing the fuel rod segment to 1000 psig and heating to 700°C while the rod is positioned in a restrainer which is machined to permit only the desired amount of expansion.

Following the expansion, a 0.159-cm (0.0625-in.)-diam hole is to be drilled through the cladding to form a reproducibly defected rod segment with fission product diffusional release characteristics similar to those of a pressure-ruptured fuel rod. (Pressure-ruptured cladding behaves poorly at high temperatures; localized induction heating and steam oxidation cause enlargement of the thin-walled rupture opening accompanied by continually decreased heating in the vicinity of the opening. On the other hand, cladding with a small hole drilled through the full wall thickness can withstand induction heating in steam at high temperature for an acceptable period of time.)

2.4 Fission Product Release from Boiling Water Reactor Fuel

A series of fission product release tests in the temperature range 700 to 1200°C will be performed with Boiling Water Reactor (BWR) fuel following the High Temperature Series of experiments. The BWR tests will use fuel removed from Peach Bottom-2 BWR that was operated at higher heat ratings than the H. B. Robinson Pressurized Water Reactor (PWR) fuel used in the High Burnup Fuel Test Series. Although the Peach Bottom-2

fuel selected for the new series (rod F-6 from bundle PH-006) has a lower burnup than the Robinson fuel, preliminary information indicates that the fission gas release to the plenum and void spaces (and therefore the release of cesium and iodine to the pellet-to-clad gap space) was much higher. We anticipate that the gap inventory for cesium and iodine will be a factor of ten higher. Use of this fuel should more clearly define the effect of gap inventory on cesium and iodine release from ruptured fuel rods. (Our preliminary model[7] correlates fractional release with gap inventory raised to the 0.8 power.)

Characterization of the test rod has begun; a gross gamma scan is shown in Fig. 2. Fuel lost from locations where the rod was cut before scanning causes irregular depressions in the scan. The detailed scans show cesium deficiency between pellets in the low-power, low-burnup end regions and cesium activity peaks between pellets in portions of the high-burnup, high-power regions. Fission gas released to the plenum and void spaces was measured by EG&G Idaho[8] before the rod was cut. Gas release fractions will be calculated following burnup determination. (Other fuel rods from the same bundle will receive further characterization at EG&G Idaho.[8]) The BWR test series will include measurement of the gap inventory of cesium and iodine by the same purge-volatilization technique employed in test HBU-12.

3.0 FISSION PRODUCT TRANSPORT TEST FACILITY

S. K. Whatley

A feasibility study was conducted to determine if the Fission Product Transport Test Facility (FPTTF) could be designed for experiments with high-temperature steam (to about 1600°C). Since the state-of-the-art temperature for commercial superheaters (utilizing a tube heat exchanger) and the materials of construction for the proposed test components for the FPTTF is 1000°C, two preliminary designs were considered — one for 1000°C steam at the test section and the other for 1600°C steam. Following discussions with NRC on the two designs, it was decided that design work on only the 1000°C concept would be continued.

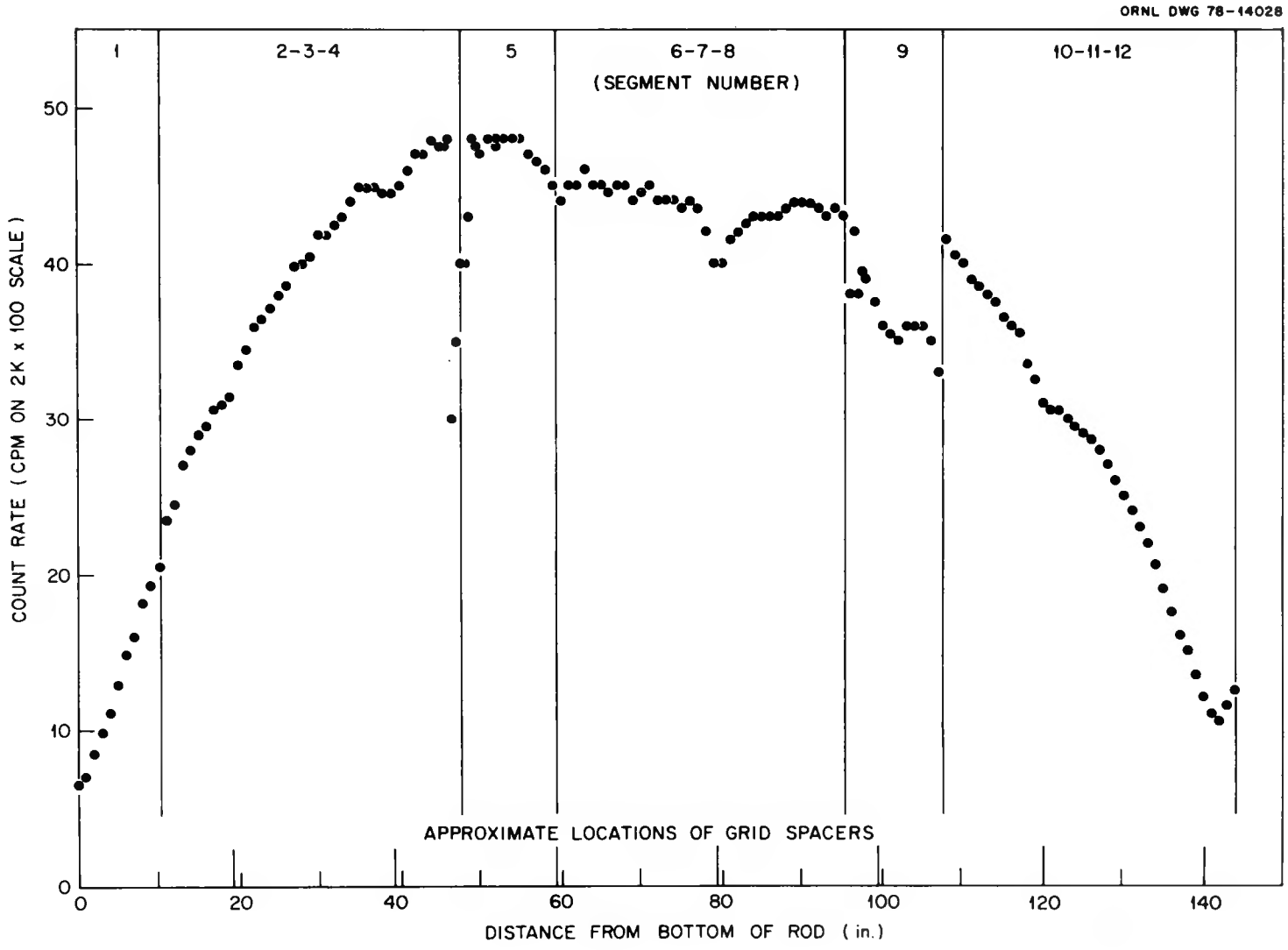

Fig. 2. Gamma scan of rod F-6 from bundle PH-006, Peach Bottom-2.

Design effort was concentrated on development of the fission product injector systems and the proposed test components for both a pressurized and a boiling water reactor.

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