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Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April–June 1977

A. P. Malinauskas

Prepared for the U.S. Nuclear Regulatory Commission
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CHEMICAL TECHNOLOGY DIVISION

QUARTERLY PROGRESS REPORT ON FISSION PRODUCT BEHAVIOR IN LWRs FOR THE PERIOD APRIL-JUNE 1977

A. P. Malinauskas, Program Manager

R. A. Lorenz
R. P. Wichner
J. L. Collins
S. K. Whatley
O. L. Kirkland
R. L. Towns

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Prepared by the
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Oak Ridge, Tennessee 37830
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SUMMARY

Two fission product release experiments were conducted in an air atmosphere. One of these, High Burnup Fuel Test 5 (HBU-5), was conducted over a 20-hr period at 500°C. In this experiment, 0.5×10^{-6}% of the total cesium inventory and 91×10^{-6}% of the ruthenium inventory were released from the rod. The second test, High Burnup Fuel Test 6 (HBU-6), was run over a 5-hr period at 700°C. The experiment yielded a cesium release amounting to 14.3×10^{-4}% of inventory, whereas the ruthenium release was 7.3×10^{-4}%. The ruthenium releases are markedly greater than observed previously in corresponding tests in steam.

Design of the fission product transport test facility has progressed to the point at which estimates of the costs of construction can be made.

1.0 INTRODUCTION

This program is comprised of two major activities: (1) studies of fission product release from light water reactor (LWR) fuel, and (2) the conceptual design of a fission product transport test facility.

The objective of the fission product release studies is to determine the chemical and physical states of fission product radionuclides which escape from defected fuel rods in steam and air at temperatures characteristic of spent fuel transportation accidents (SFTA) and loss-of-coolant accidents (LOCA) in light water reactors. The experimental work has advanced to the stage wherein all of the testing is now being conducted with irradiated fuel obtained from a commercial reactor. Experiments in the range 500-900°C are performed with fuel rod segments containing mechanically defected claddings, whereas additional tests at temperatures of 900°C and higher are conducted with rod segments which are ruptured during the test by internal pressurization. Descriptions of the apparatus and of the test specimens have been reported previously.[1,2] In this report the results of two tests in an air atmosphere are presented; one of these tests was conducted at 500°C and the other at 700°C.

The current objective of the second major activity is to establish the need and the cost of a fission product transport test facility (FPTTF) for use in experimental validations of computer models which are being developed to describe fission product behavior in an LWR primary coolant circuit during a reactor accident. The accident modes which are to be considered range from controlled loss-of-coolant accidents to core meltdown. During this report period, the schematic representation of the facility which was described previously[3] has been developed to the stage at which estimates of the costs of construction can be made.

2.0 FISSION PRODUCT RELEASE FROM LWR FUEL

R. A. Lorenz
J. L. Collins
O. L. Kirkland
R. L. Towns

Two additional tests with H. B. Robinson fuel were conducted during this report period. As in previous tests, a 0.159-cm-diam hole was drilled

into each of the two rod segments at the midpoint location to simulate a defect. The specimens were heated with a resistance heater and maintained in a flowing dry air atmosphere. (Steam-argon atmospheres were employed in earlier tests of this, the High Burnup Fuel Test Series.) Pertinent information about the fuel rod and the test segments which were machined from it has been presented in an earlier report.[2]

2.1 High Burnup Fuel Test 5 (500°C, Dry Air)

This experiment was performed at 500°C for 20 hr. The fuel specimen (A-5 from fuel rod D-12) was previously employed in High Burnup Fuel Test 4, which was conducted at 500°C over a 20-hr period in a flowing steam-argon atmosphere.[3]

The post-test appearance of the fuel rod segment is shown in Fig. 1. The swollen region in the vicinity of the hole was caused by oxidation of the UO_2 fuel. A small amount of oxidized fuel can be seen protruding from the hole.

Approximately 0.53% (7.2 mCi) of the original ^{85}Kr inventory was released during the test. The chronology of this release is displayed graphically in Fig. 2. Since this rod segment was employed in a previous 20-hr test at 500°C, during which time about 0.63% (8.6 mCi) of the original ^{85}Kr escaped, the additional release observed in this test is probably due to oxidation of the fuel. As is evident in Fig. 2, cessation of krypton release occurred after 4 hr of testing; this suggests that the defect hole became plugged, thus preventing further oxidation of the fuel. Furthermore, it appears likely that the release of other fission product nuclides was similarly halted.

The post-test distribution of ^{134}Cs in the collection system is summarized in Table 1. These data indicate that 5.1×10^{-7}% (2.4×10^{-3} μg) of the total cesium inventory was released. If it is assumed that little additional cesium were released after 4 hr, as is likely, then the linear release rate would be 1.3×10^{-7}%/hr (as compared with a linear release rate of 1.8×10^{-7}%/hr in High Burnup Fuel Test 4).

Small particles of oxidized fuel were present on the fuel rod holder. Determinations of ^{144}Ce (assumed to remain unseparated from the fuel) were made which indicated that approximately 5.84 mg of fuel remained on the holder even after loose particles were removed. This amount of fuel

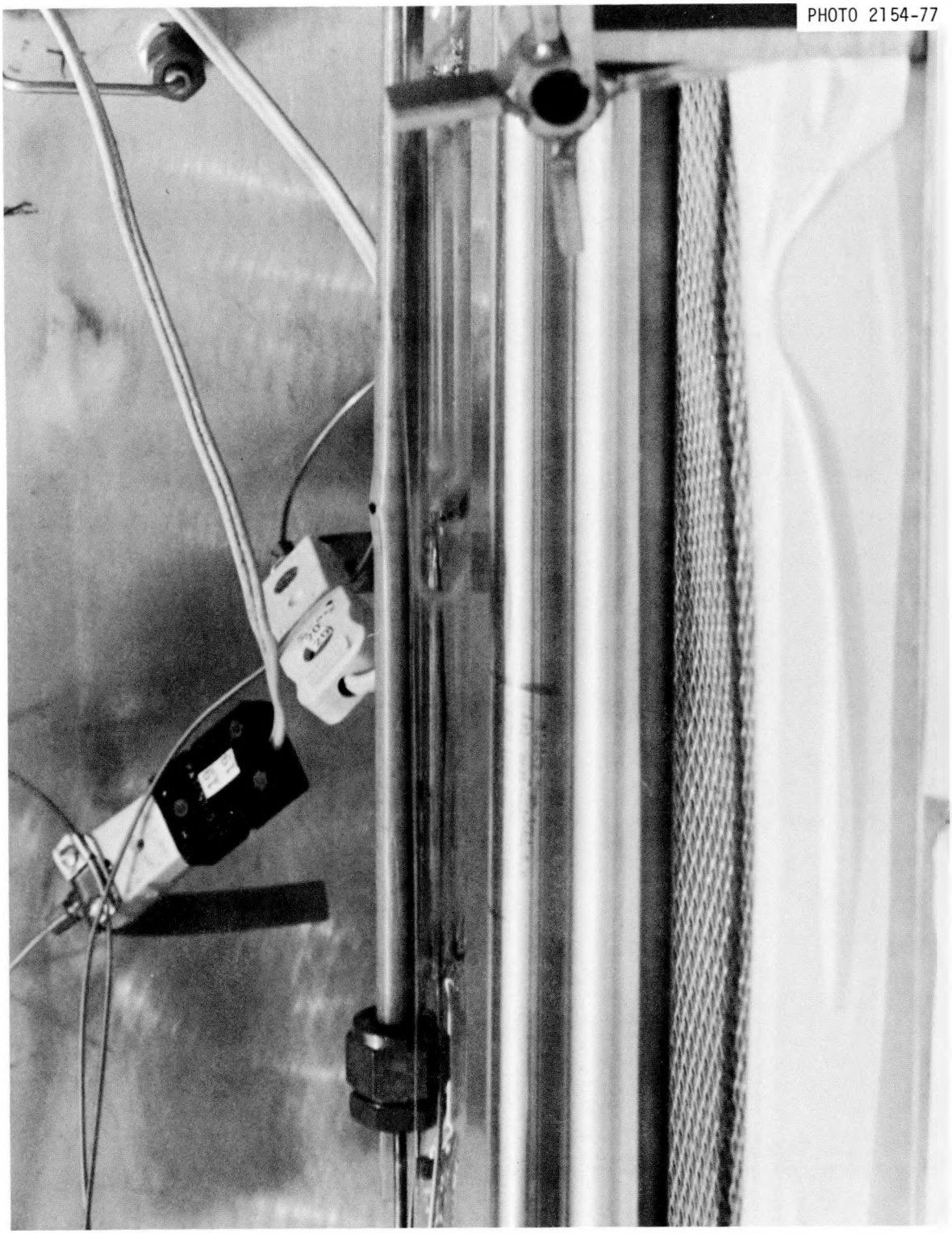

Fig. 1. Fuel rod segment A-5 of rod D-12 after a 20-hr exposure to air at 500°C.

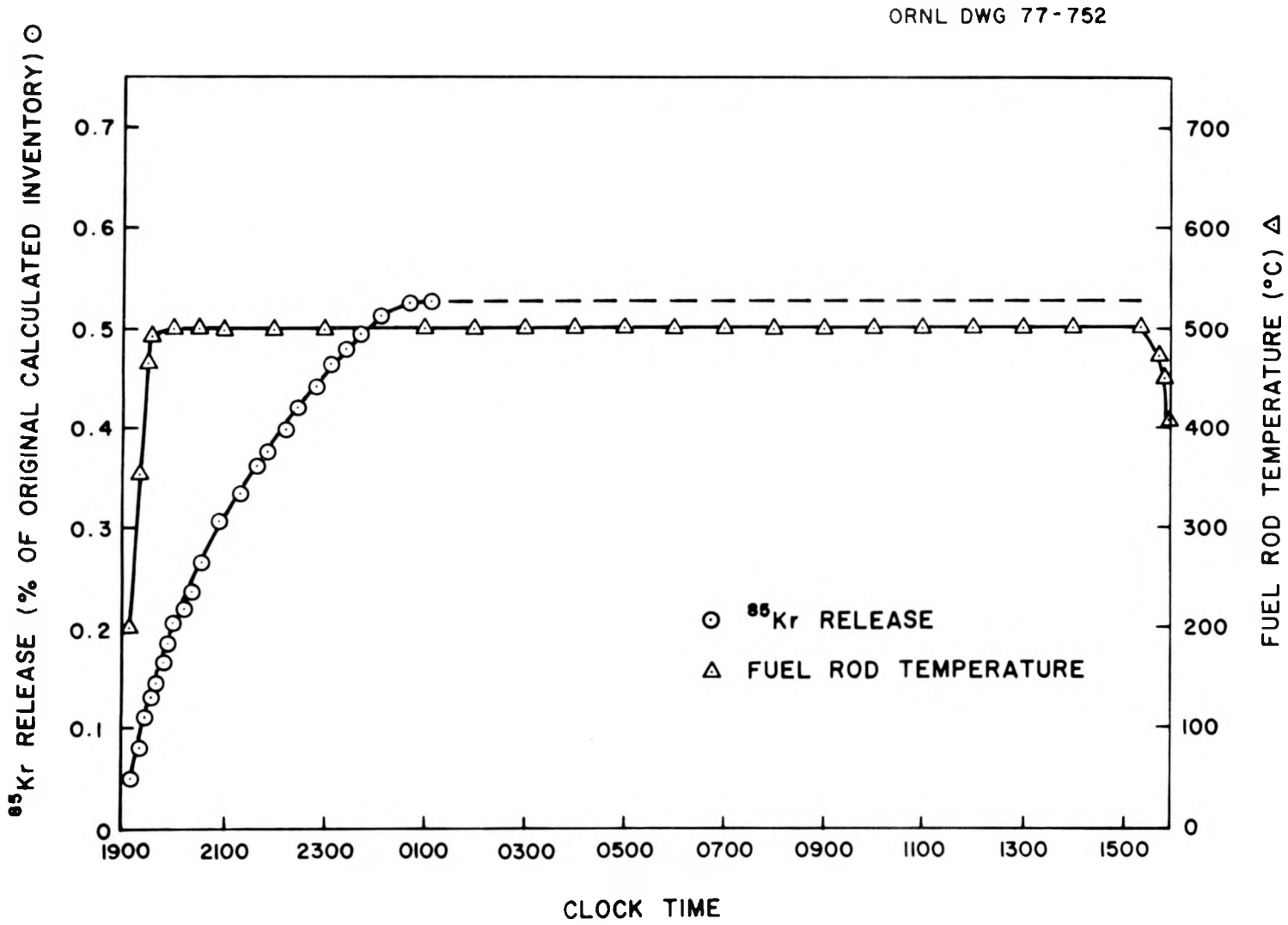

Fig. 2. Krypton-85 release and fuel rod temperature during test HBU-5.

Table 1. Distribution of ^{134}Cs in High Burnup Fuel Test No. 5[a]

Location	Temperature (°C)	Amount of ^{134}Cs found in each location: μg[b]	Percent of total[c]	Percent of released	Total Cs (μg)
Fuel rod	500	8.91×10^{3}[d]			4.69×10^{5}
Furnace tube quartz liner	500	1.93×10^{-5}	2.17×10^{-7}	42.20	1.02×10^{-3}
Fuel rod holder assembly	500				
Quartz fuel rod holder		1.55×10^{-5}[e]	1.74×10^{-7}	33.89	8.16×10^{-4}
Thermocouple No. 1 (inlet end)		0.0			
Thermocouple No. 2 (center)		0.0			
Thermal gradient tube	640-200	6.07×10^{-6}	6.80×10^{-8}	13.27	3.20×10^{-4}
Filter pack components	125				
Stainless steel inlet fitting		1.4×10^{-6}[f]	1.57×10^{-8}	3.06	7.37×10^{-5}
Other housing components		0.0			
First filter paper		3.42×10^{-6}	3.84×10^{-8}	7.48	1.80×10^{-4}
Second filter paper		9.90×10^{-9}		0.02	
Third filter paper		9.25×10^{-9}	1.11×10^{-10}	0.02	5.21×10^{-7}
Charcoal No. 1a		2.45×10^{-8}	2.75×10^{-10}	0.05	1.29×10^{-6}
Charcoal No. 1b		0.0			
Charcoal No. 1c		0.0			
Charcoal No. 2a		0.0			
Charcoal No. 2b		0.0			
Charcoal No. 3		0.0			
AgX		0.0			
Freeze trap	-78				
Cold charcoal traps (two)	-78				

[a] Dry air flow rate, 354 cm^3/min (STP); system pressure, 760 torr. Decay time, 911 days (to November 2, 1976).

[b] Amounts less than 1.0×10^{-9} μg are given as 0.0.

[c] Percentage of radioactive nuclide in fuel rod.

[d] Calculated for burnup of 30,756 MWd/metric ton of original uranium, 183.3 g of uranium originally in 12-in. segment, and 911 days decay.

[e] Estimated amount of vapor-deposited ^{134}Cs. Excludes 0.25 μg present with adhering small particles of fuel.

[f] Estimated amount. Cesium-134 could not be detected because of high ^{106}Ru activity.

originally contained 5.83 μg of ^{137}Cs, compared with 5.83 μg which was found by counting radioactive ^{137}Cs. Therefore, in order to obtain an estimate of the amount of cesium deposited on the fuel rod holder from transported vapor, we used the ratio of cesium on the holder to cesium on the quartz liner as measured in HBU-6 (a dry air test at 700°C). By this method we calculated that 0.00082 μg of cesium deposited on the fuel rod holder from the vapor phase compared with 5.83 μg present within adhering particles of fuel.

The most notable result in this experiment was the relatively large release of ruthenium. Only trace amounts were released in the three prior tests, which were conducted in steam. The ruthenium distribution data, which are presented in Table 2, indicate that 9.04×10^{-5}% (0.377 μg) of the total ruthenium inventory was released. Also, about 99% of the released ruthenium deposited on the cooler surfaces of the collection apparatus, especially at the outlet end of the gold thermal gradient tube (300 to 200°C) as illustrated in Fig. 3, in the stainless steel inlet to the filter pack (125°C), and on the first filter paper (125°C). This suggests a highly volatile vapor species, such as RuO_4.

2.2 High Burnup Fuel Test 6 (700°C, Dry Air)

HBU-6 was the second experiment conducted in the High Burnup Fuel Test Series in which a dry air atmosphere was employed. The fuel rod segment (A-2 of fuel rod D-12) was maintained at 700°C for 5 hr by means of a resistance heater which surrounded the furnace tube. Unlike the rod segment employed in HBU-5, no previous testing was performed on this fuel specimen. Determination of radial gap size via a gas flow technique[3] indicated a pellet-to-clad gap space in the fuel rod segment of about 18 μm. The distribution of radionuclides ^{134}Cs and ^{106}Ru at the conclusion of the test is summarized in Tables 3 and 4, respectively.

As can be seen in the photograph in Fig. 4, some swelling occurred in the region of the hole as a result of fuel oxidation. Also, as in HBU-5, a small quantity of fuel can be seen protruding from the defect hole.

The exterior coloration of the cladding was copper-to-pink, and the surface texture appeared to be smooth and unbroken. This cladding surface phenomenon has been observed in all our tests (both irradiated and unirradiated) conducted at 700°C irrespective of the test atmosphere (steam or air).

Table 2. Distribution of ^{106}Ru in High Burnup Fuel Test No. 5[a]

Location	Temperature (°C)	Amount of ^{106}Ru found in each location: μg[b]	Percent of total[c]	Percent of released	Total Cs (μg)
Fuel rod	500	5.93×10^{3}[d]			4.17×10^{5}
Furnace tube quartz liner	500	6.14×10^{-5}	1.04×10^{-6}	1.15	4.32×10^{-3}
Fuel rod holder assembly	500				
Quartz fuel rod holder		0.0	0.0	0.0	0.0
Thermocouple No. 1 (inlet end)		0.0	0.0	0.0	0.0
Thermocouple No. 2 (center)		0.0	0.0	0.0	0.0
Thermal gradient tube	640-200	2.36×10^{-3}	3.98×10^{-5}	44.04	1.66×10^{-1}
Filter pack components	125				
Stainless steel inlet fitting		1.21×10^{-3}	2.04×10^{-5}	22.62	8.51×10^{-2}
Other housing components		9.99×10^{-7}	1.68×10^{-8}	0.019	7.02×10^{-5}
First filter paper		1.72×10^{-3}	2.90×10^{-5}	32.03	1.21×10^{-1}
Second filter paper		3.22×10^{-6}	5.43×10^{-8}	0.060	2.26×10^{-4}
Third filter paper		1.45×10^{-6}	2.45×10^{-8}	0.027	1.02×10^{-4}
Charcoal No. 1a		2.27×10^{-6}	3.83×10^{-8}	0.042	1.60×10^{-4}
Charcoal No. 1b		5.64×10^{-8}	9.51×10^{-10}	0.001	3.97×10^{-6}
Charcoal No. 1c		2.54×10^{-8}	4.28×10^{-10}	0.001	1.79×10^{-6}
Charcoal No. 2a		5.30×10^{-8}	8.94×10^{-10}	0.001	3.73×10^{-6}
Charcoal No. 2b		5.91×10^{-8}	9.97×10^{-9}	0.001	4.16×10^{-6}
Charcoal No. 3		7.69×10^{-8}	1.30×10^{-10}	0.001	5.41×10^{-6}
AgX		1.30×10^{-8}	2.19×10^{-10}	<0.001	9.14×10^{-7}
Freeze trap	-78				
Cold charcoal traps (two)	-78				

[a] Dry air flow rate, 354 cm^3/min (STP); system pressure, 760 torr. Decay time, 911 days (to November 2, 1976).

[b] Amounts less than 1.0×10^{-8} μg are given as 0.0.

[c] Percentage of radioactive nuclide in fuel rod.

[d] Calculated for burnup of 30,756 MWd/metric ton of original uranium, 183.3 g of uranium originally in 12-in. segment, and 911 days decay.

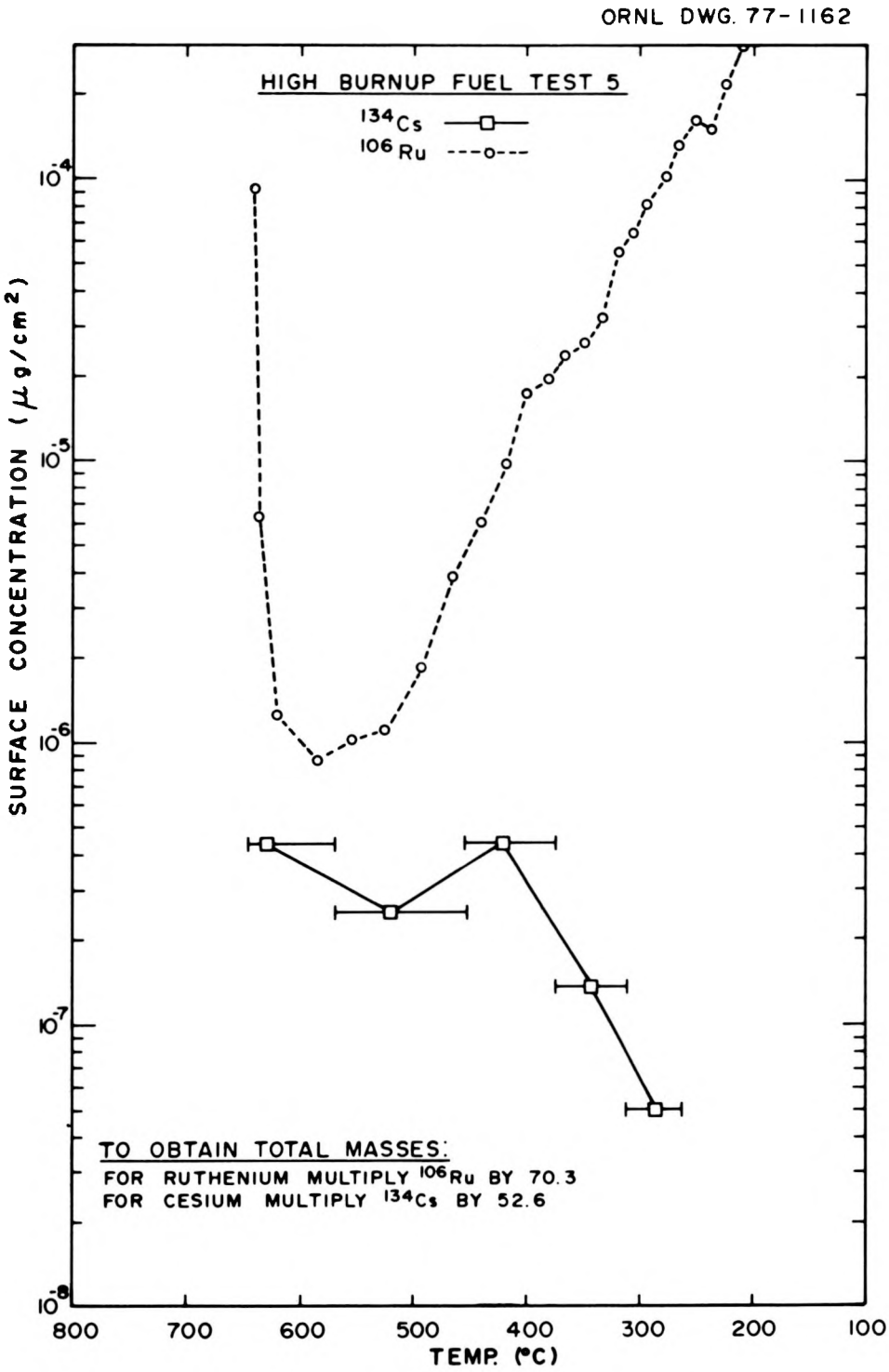

Fig. 3. Distribution of ^{134}Cs and ^{106}Ru deposited in the thermal gradient tube during run HBU-5.

Table 3. Distribution of ^{134}Cs in High Burnup Fuel Test No. 6[a]

Location	Temperature (°C)	Amount of ^{134}Cs found in each location			Total Cs (μg)
		μg[b]	Percent of total[c]	Percent of released	
Fuel rod	700	8.20×10^{3}[d]			3.84×10^{5}
Furnace tube quartz liner	700	1.93×10^{-2}	2.36×10^{-4}	16.48	1.016
Fuel rod holder assembly	700				
Quartz fuel rod holder		1.55×10^{-2}	1.89×10^{-4}	13.24	8.16×10^{-1}
Thermocouple No. 1 (inlet end)		2.41×10^{-6}	2.94×10^{-8}	0.002	1.27×10^{-4}
Thermocouple No. 2 (center)		3.76×10^{-5}	4.59×10^{-7}	0.032	1.98×10^{-3}
Thermal gradient tube	700-210	7.97×10^{-2}	9.73×10^{-4}	68.06	4.195
Filter pack components	125				
Stainless steel inlet fitting		7.10×10^{-4}	8.66×10^{-6}	0.61	3.74×10^{-2}
Other housing components					
First filter paper		1.85×10^{-3}	2.26×10^{-5}	1.58	9.74×10^{-2}
Second filter paper		0.0			
Third filter paper		0.0			
Charcoal No. 1a		0.0			
Charcoal No. 1b		0.0			
Charcoal No. 1c		0.0			
Charcoal No. 2a		0.0			
Charcoal No. 2b		0.0			
AgX		0.0			
Freeze trap	-78	0.0			
Cold charcoal traps (two)	-78	0.0			

[a] Dry air flow rate, 363 cm^3/min (STP); system pressure, 760 torr. Decay time, 911 days (to November 2, 1976).
[b] Amounts less than 1.0×10^{-8} μg are given as 0.0.
[c] Percentage of radioactive nuclide in fuel rod.
[d] Calculated for burnup of 28,285 MWd/metric ton of original uranium, 183.3 g of uranium originally in 12-in. segment, and 911 days decay.

Table 4. Distribution of ^{106}Ru in High Burnup Fuel Test No. 6[a]

Location	Temperature (°C)	Amount of ^{106}Ru found in each location: μg[b]	Percent of total[c]	Percent of released	Total Ru
Fuel rod	700	5.45×10^{3}			3.83×10^{5}[d]
Furnace tube quartz liner	700	0.0	0.0	0.0	0.0
Fuel rod holder assembly	700	0.0	0.0	0.0	0.0
Gold thermal gradient tube	700-210	2.91×10^{-3}	5.34×10^{-5}	7.35	2.05×10^{-1}
Filter pack components	125				
Stainless steel inlet fitting		1.59×10^{-2}	2.29×10^{-4}	40.13	1.116
Other housing components		0.0	0.0	0.0	0.0
First filter paper		2.08×10^{-2}	3.82×10^{-4}	52.50	1.462
Second filter paper		5.01×10^{-6}	9.19×10^{-8}	0.013	3.52×10^{-4}
Third filter paper		5.82×10^{-7}	1.07×10^{-8}	0.002	4.09×10^{-5}
Charcoal No. 1a		1.15×10^{-6}	2.11×10^{-8}	0.003	8.07×10^{-5}
Charcoal No. 1b		2.10×10^{-8}	3.85×10^{-10}	0.00005	1.48×10^{-6}
Charcoal No. 1c		1.44×10^{-8}	2.64×10^{-10}	0.00004	1.01×10^{-6}
Charcoal No. 2a		1.68×10^{-8}	3.08×10^{-10}	0.00004	1.18×10^{-6}
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
Freeze trap	-78	0.0	0.0	0.0	0.0
Cold charcoal traps (two)	-78	0.0	0.0	0.0	0.0

[a] Dry air flow rate, 363 cm^3/min (STP); system pressure, 760 torr. Decay time, 911 days (to November 2, 1976).

[b] Amounts less than 1.0×10^{-8} µg are given as 0.0.

[c] Percentage of radioactive nuclide in fuel rod.

[d] Calculated for burnup of 28,285 MWd/metric ton of original uranium, 183.3 g of uranium originally in 12-in. segment, and 911 days decay.

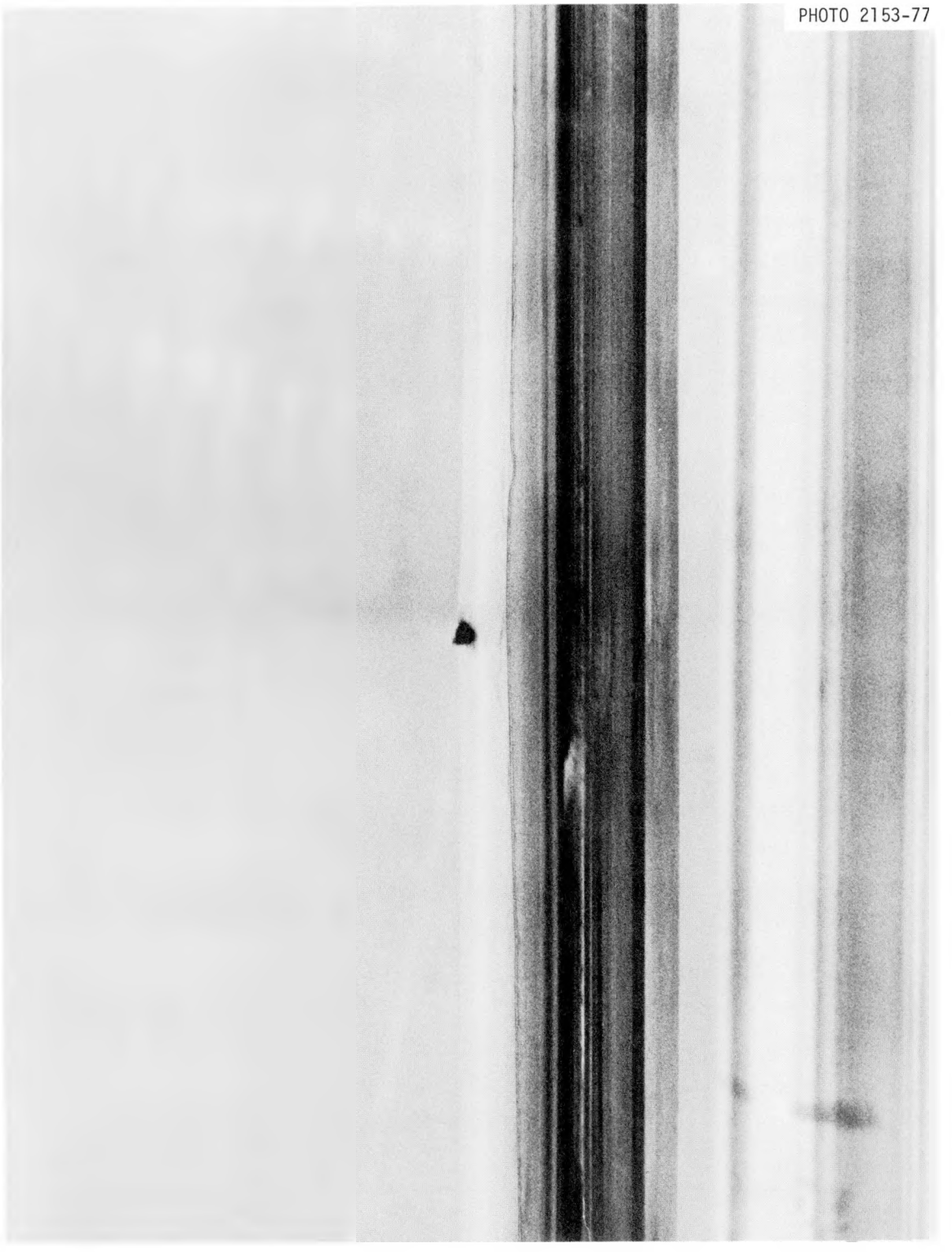

Fig. 4. Fuel rod segment A-2 of rod D-12 after a 5-hr exposure to air at 700°C.

The release of ^{85}Kr during the experiment (see Fig. 5) amounted to approximately 1.24% (15.5 mCi) of the original inventory. Since no other tests were performed with the fuel specimen, this value represents the release of recoil-implanted krypton (releasable at $\leq$ 700°C) from the fuel and cladding surface layers of the gap region, as well as krypton released by fuel oxidation.

Approximately 1.43×10^{-3}% (6.16 µg) of the total cesium inventory of the rod segment was released. Contrary to the results obtained for rods tested in steam, where less than 0.2% of the released cesium was transported beyond the quartz furnace tube, about 70% of the released cesium did so in this test. Most of it deposited in the gold thermal gradient tube. Concentration profiles of ^{134}Cs and ^{106}Ru along the thermal gradient tube are presented in Fig. 6. The cesium peak occurs at about 400°C. Continuous monitoring of the thermal gradient tube and filter pack, as is depicted graphically in Fig. 5, indicates that little cesium deposited there in the first hour after the test temperature (700°C) was reached. Subsequently, a deposition rate of about 0.02 µg of ^{134}Cs per hr (or 1.08 µg of Cs per hr) resulted. The released cesium appears to have initially reacted with the quartz liner in a manner which passivates it, thus allowing additional released cesium species to be transported downstream. The ^{134}Cs distribution displayed in Fig. 7 indicates that the cesium was more or less evenly deposited on the furnace tube liner downstream of the defect hole.

As in HBU-5, the ruthenium release was considerable; approximately 7.27×10^{-4}% (2.79 µg) of the total ruthenium inventory was released. Similarly, the distribution data presented in Table 4 indicate that the ruthenium released is in a very volatile form. In effect, all of the released ruthenium was transported downstream to the cool end (300 to 125°C) of the collection train before being deposited. About 7% deposited at the end of the gold thermal gradient tube, yielding a profile displayed in Fig. 5; the remainder deposited on the stainless steel inlet fitting and the first filter paper, both at about 125°C.

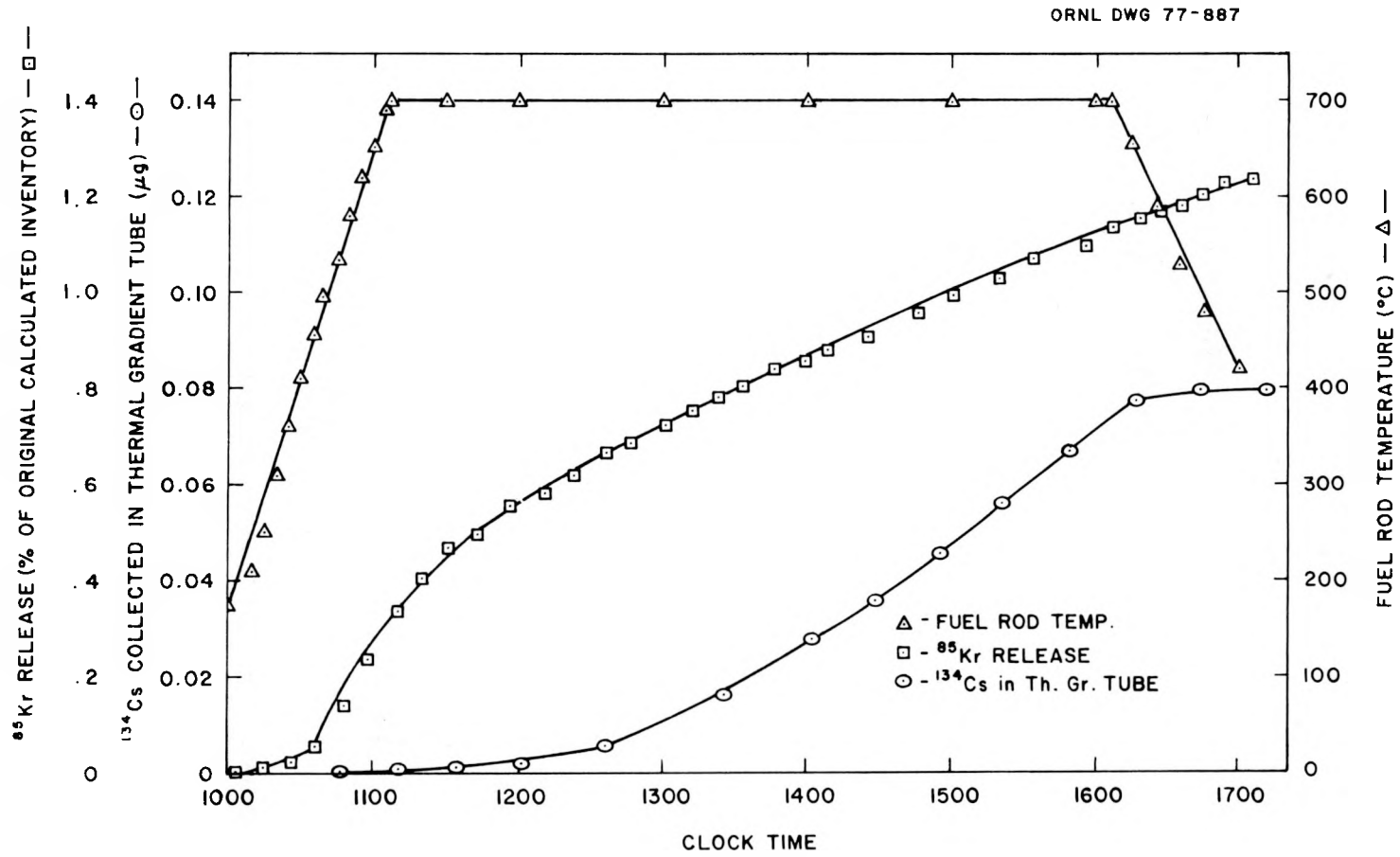

Fig. 5. Temperature and release of ^{85}Kr and ^{134}Cs during test HBU-6.

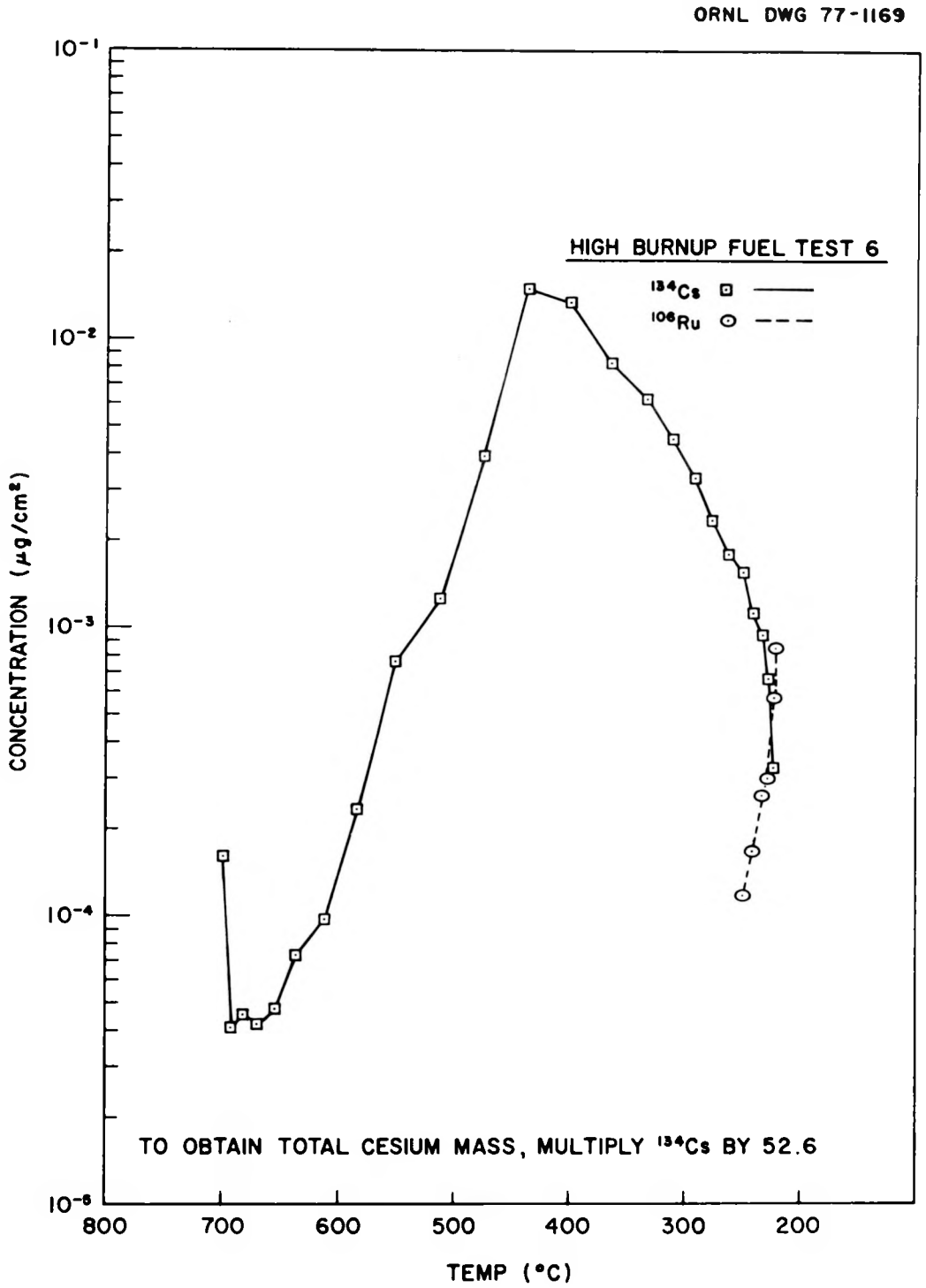

Fig. 6. Distribution of ^{134}Cs and ^{106}Ru deposited in the thermal gradient tube during run HBU-6.

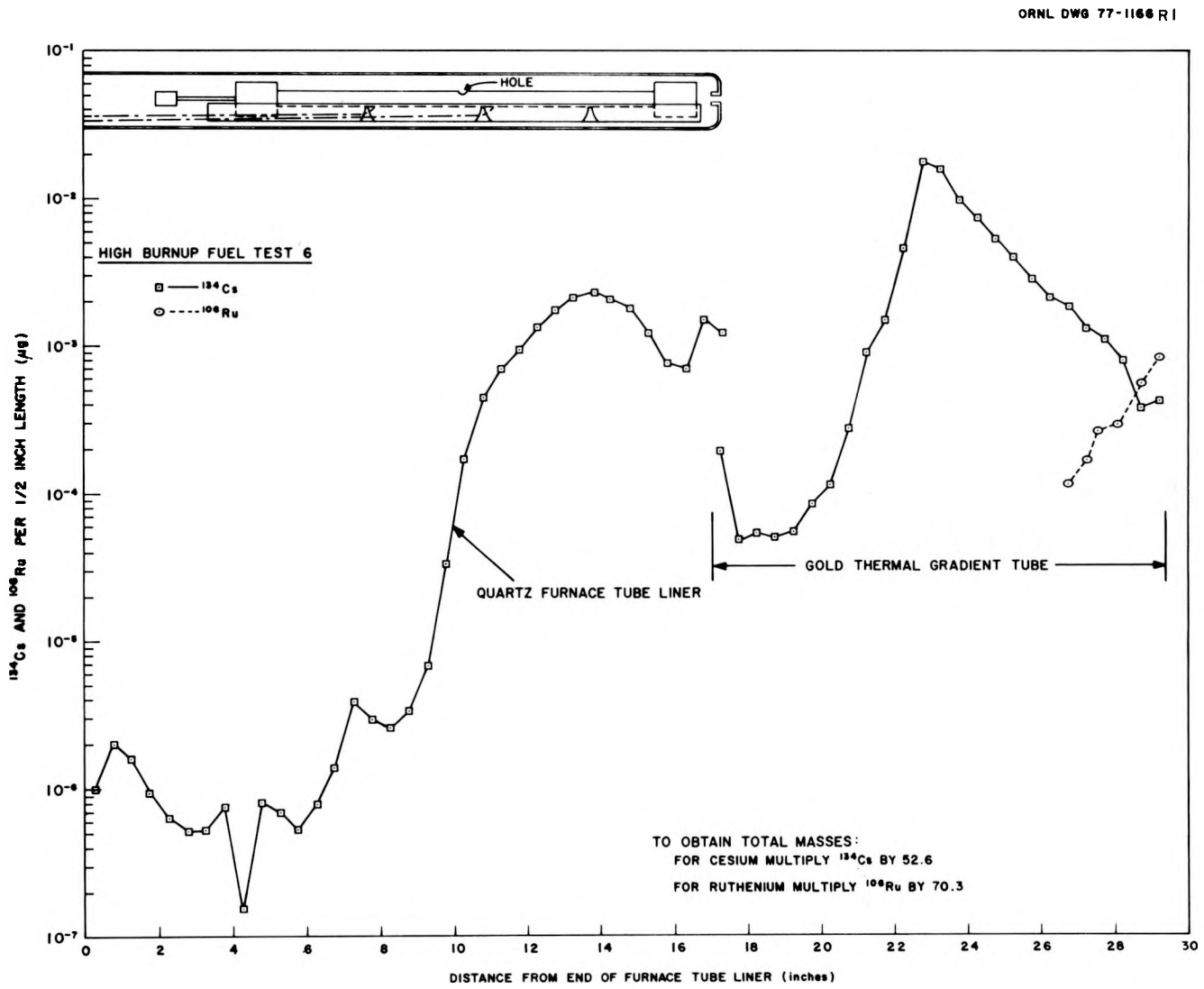

Fig. 7. Distribution of ^{134}Cs and ^{106}Ru in the furnace tube liner and thermal gradient tube in HBU-6.

2.3 Comparison of Releases in Steam and Air

Releases of ^{85}Kr, ^{134}Cs, and ^{106}Ru in steam and air atmospheres are compared in Table 5. (The tests in steam have been described previously.[2,3]) The amount of cesium released at 500°C in air appears unusually low compared with that observed at 700°C, as well as that measured in the Implant Test Series.[4,5] In the implant tests, approximately ten times as much cesium was released from the gap space in a dry air atmosphere as in a steam atmosphere at 700°C. The release of ^{85}Kr in HBU-5 stopped after 4 hr, suggesting that oxidation of the UO_2 may have ceased, perhaps as a result of plugging of the drilled hole. Unfortunately, the release of ^{85}Kr is not a reliable indicator of oxidation. Voloxidation experiments,[6] in which irradiated fuel samples are completely oxidized at 480°C, yield less than 10% release of ^{85}Kr.

Table 5. Comparison of releases in steam and air

Fuel rod segment number	High burnup test number	Test atmosphere	Temp. (°C)	Test period (hr)	Percent of original total inventory released		
					^{85}Kr	^{134}Cs	^{106}Ru
A-5	5	Steam	500	20	0.63	3.6×10^{-6}	0.15×10^{-6}
	5	Dry air	500	20	0.53	0.51×10^{-6}	91×10^{-6}
A-7	1	Steam	700	5	~1	26×10^{-6}	0.04×10^{-6}
A-9	6	Dry air	700	5	1.24	1430×10^{-6}	727×10^{-6}

3.0 FISSION PRODUCT TRANSPORT TEST FACILITY

R. P. Wichner
S. K. Whatley

As mentioned previously, the initial objective of this task is to establish the need and estimate the cost of a Fission Product Transport Test Facility (FPTTF) for the purpose of experimentally validating computer models which predict fission product holdup in the LWR primary loop in accident modes ranging from controlled loss-of-coolant accidents to core meltdown. The evaluation of need is being made on the basis of (1) advantages of such a facility with respect to reactor licensing procedures, (2) the most critical test conditions for computer model verification purposes, and (3) the availability (or nonavailability) of existing facilities which may be employed for this objective.

3.1 Conceptual Design

The bulk of the conceptual design work for the FPTTF was completed during this period. The Instrument Application Diagram, shown in Fig. 8, is an expanded version of the schematic flow diagram presented previously.[3] Each of the following individual components were included in the conceptual design:

Dwg. No.	Component
I-11757-QC-001-D-0	Instrument Application Diagram
X2E-11757-005	Typical Test Section
X2E-11757-006	Pressurized Hot Water Tank
X2E-11757-007	Steam-Water Separator
X3E-11757-008	Clean-Up Tank
X2E-11757-009	Condenser
X2E-11757-010	Steam Generator
X2E-11757-011	Pump Simulator
X2E-11757-012	Weigh Tank
ASK-PJF-5090	Fission Product Injection System

Four different configurations (Dwg. Nos. X2E-11757-001 through 004) were prepared for the equipment layout and piping — an integral

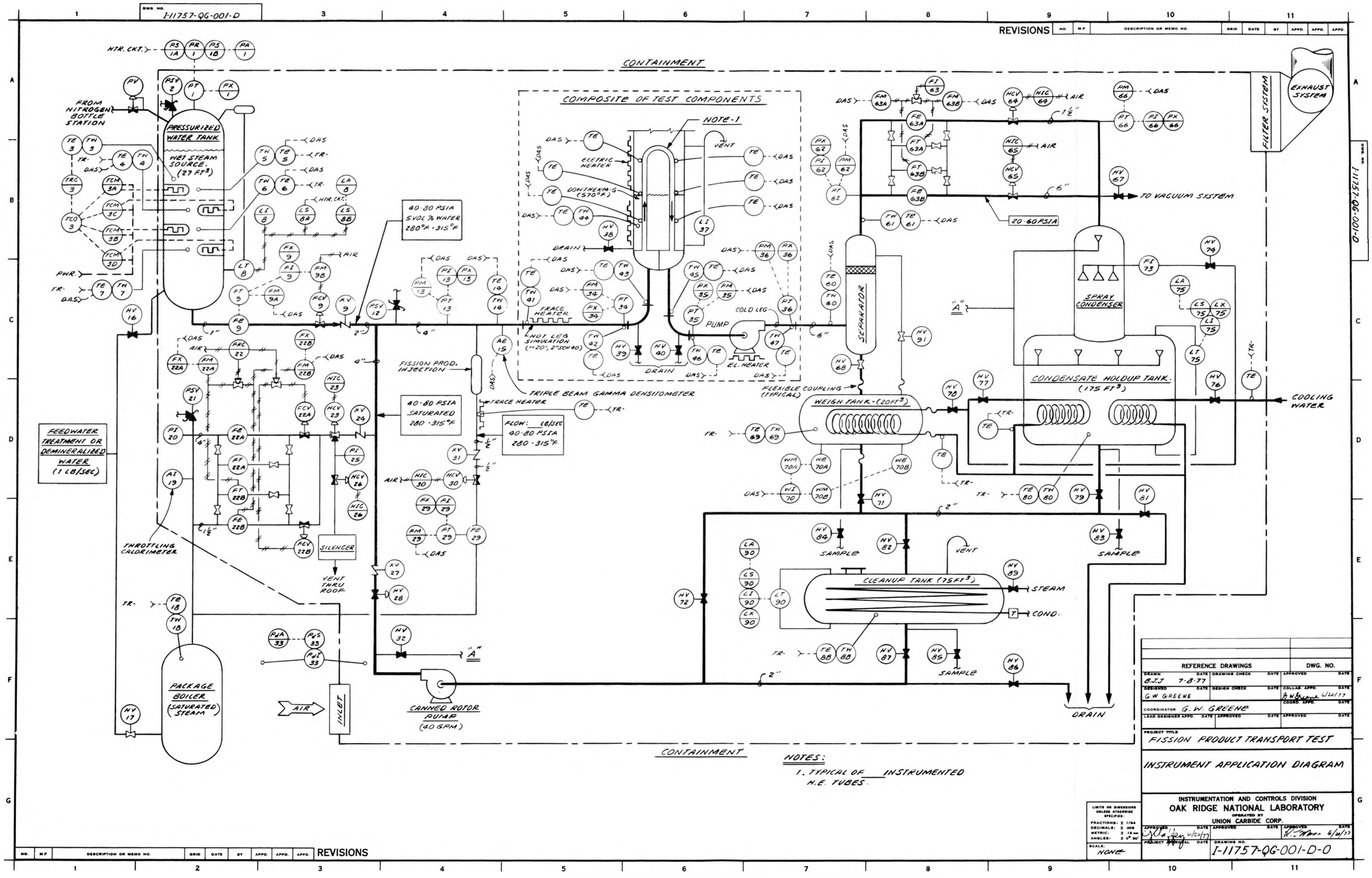

Fig. 8. Instrument application diagram of the fission product transport test facility.

test facility with all components in the primary coolant loop, and individual layouts for the steam generator, the pump simulator, and a typical test section with pipe inserts (typical of a hot or cold leg). The current design provides only for the injection of cesium as a solution of CsOH through an atomizing spray nozzle into dry steam. The injector pump will be commercially procured. Also, either a commercially available package boiler or an on-site steam supply is proposed for the main steam supply system.

In addition to the test component and equipment layout designs, the following drawings were also completed for the conceptual design:

Dwg. No.	Drawing
B-SK-PFJ-5089	FPTTF Flow Diagram
B-SK-CBC-5097	Electrical Diagram
ASK-PJF-5091	Ventilation System

3.2 Cost Estimates

Cost estimates of the FPTTF are being prepared by UCC-ND Engineering on three different bases: (1) fabrication and installation at a selected Y-12 (ORNL) experimental area; (2) fabrication and installation at a "typical" industrial eastern location; and (3) fabrication and installation at a "typical" west coast location. The Y-12 (ORNL) location, specifically selected for its suitability for this type of experimentation, is equipped with building steam supply and data acquisition equipment which were assumed to be unavailable at the other sites.

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