

**Quarterly Progress Report on Fission
Product Behavior in LWRs for the
Period October-December 1977**

A. P. Malinauskas
R. A. Lorenz
J. L. Collins
M. F. Osborne
S. K. Whatley
R. L. Towns

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

ORNL/NUREG/TM-186
Dist. Category NRC-3

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

QUARTERLY PROGRESS REPORT ON FISSION PRODUCT BEHAVIOR IN LWRs FOR THE
PERIOD OCTOBER-DECEMBER 1977

A. P. Malinauskas, Program Manager

R. A. Lorenz	S. K. Whatley
J. L. Collins	R. L. Towns
M. F. Osborne	

Manuscript Completed - February 1978

Date Published - March 1978

NOTICE: This document contains information of a preliminary nature.
It is subject to revision or correction and therefore does not
represent a final report.

Prepared for the
U. S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
Under Interagency Agreements DOE 4G-551-75 and 4G-552-75

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
DEPARTMENT OF ENERGY

NOTICE
This report was prepared as an account of work
sponsored by the United States Government. Neither the
United States nor the United States Department of
Energy, nor any of their employees, nor any of their
contractors, subcontractors, or their employers, makes
any warranty, express or implied, or assumes any legal
liability or responsibility for the accuracy, completeness
or usefulness of any information, apparatus, product or
process disclosed, or represents that its use would not
infringe privately owned rights.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

TABLE OF CONTENTS

	PAGE
FOREWORD	v
SUMMARY	1
1.0 INTRODUCTION	2
2.0 FISSION PRODUCT RELEASE FROM LWR FUEL	2
2.1 High Burnup Fuel Test 10 (1200°C)	3
2.2 Comparison of Cesium Release in Steam	8
3.0 FISSION PRODUCT TRANSPORT TEST FACILITY	12
3.1 Objective of the Facility	12
3.2 Design Criteria	13
3.3 Description of the FPTTF	13
3.4 Conclusions and Recommendations	16
4.0 REFERENCES	18

FOREWORD

This report documents progress made during the period October-December 1977. 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. 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-5250 (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-Uranium Interactions, ORNL/NUREG/TM-24 (June 1976).
8. R. A. Lorenz, M. F. Osborne, J. L. Collins, S. R. Manning, and A. P. Malinauskas, Behavior of Iodine, Methyl Iodide, Cesium Oxide, and Cesium Iodide in Steam and Argon, ORNL/NUREG/TM-25 (July 1976).
9. R. A. Lorenz, J. L. Collins, S. R. Manning, and A. P. Malinauskas, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period January-March 1976, ORNL/NUREG/TM-30 (July 1976).
10. R. A. Lorenz, J. L. Collins, S. R. Manning, O. L. Kirkland, and A. P. Malinauskas, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period April-June 1976, ORNL/NUREG/TM-44 (August 1976).

11. R. A. Lorenz, J. L. Collins, and O. L. Kirkland, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period July-September 1976, ORNL/NUREG/TM-73 (December 1976).
12. R. A. Lorenz, J. L. Collins, and O. L. Kirkland, Quarterly Progress Report on Fission Product Release from LWR Fuel for the Period October-December 1976, ORNL/NUREG/TM-88 (March 1977).
13. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period January-March 1977, ORNL/NUREG/TM-122 (June 1977).
14. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period April-June 1977, ORNL/NUREG/TM 139 (September 1977).
15. A. P. Malinauskas et al., Quarterly Progress Report on Fission Product Behavior in LWRs for the Period July-September 1977, ORNL/NUREG/TM-170 (January 1978).

SUMMARY

Analysis of release data obtained during High Burnup Fuel Test 10 (HBU-10) has been completed. In this test the fuel rod segment was ruptured by internal pressurization at 900°C, at which time the temperature was rapidly increased to 1200°C and maintained at this temperature for 10 min. Approximately 0.061% of the total cesium inventory in the rod segment was released; this was accompanied by the release of about 1.69% of the total ⁸⁵Kr inventory. Moreover, about 0.022% of the fuel was ejected from the rod as particulates.

The feasibility and conceptual design study for an experimental facility to verify fission product transport computational models has been completed. Although the study focused entirely on a facility which would be employed to verify those aspects of the transport models that dealt with fission product source-term attenuation factors in the primary coolant circuit external to the pressure vessel, the need for a second facility, to simulate conditions within the pressure vessel, was also indicated.

1.0 INTRODUCTION

This program is presently comprised of two activities. One of these concerns studies of fission product release from defected light water reactor (LWR) fuel rods. The objective of the studies is to provide fission product source-term data for use in analyses of spent fuel transportation accidents and loss-of-coolant accidents in light water reactors.

Results to date indicate that release from the fuel-clad gap region of defected rods can be described as a superposition of two processes, a "burst release" which occurs during the period of rod rupture and a "diffusive release" process which occurs subsequently. The magnitude of the latter, relative to burst release, is dependent upon both time and temperature, and current studies have been directed toward a quantitative description of these dependences. Although some consideration is given to this aspect in the report, much more effort was expended in completing the analysis of data obtained during and after the conduct of High Burnup Fuel Test HBU-10, and this is reflected in the document.

The second activity associated with this program involves a study that is being made to establish the need and costs of construction and operation of a fission product transport test facility (FPTTF) which would be used for the experimental validation of computational models for fission product transport. These models, which are being developed to describe fission product behavior in a LWR primary coolant circuit during a reactor accident, are expected to describe accidents that range from controlled loss-of-coolant accidents to core meltdown.

During this period, the conceptual design and preliminary cost estimate of a scaled model facility were completed and formally transmitted to NRC.

2.0 FISSION PRODUCT RELEASE FROM LWR FUEL

R. A. Lorenz
J. L. Collins

M. F. Osborne
R. L. Towns

High Burnup Fuel Test 10 was the final test of a series of experiments which were conducted using irradiated fuel rod segments machined from the H. B. Robinson Reactor fuel rods. Details about the rods and the test segments have been presented in an earlier report.¹

2.1 High Burnup Fuel Test 10 (1200°C)

This test, which utilized segment A-6 of fuel rod D-12, was conducted over a 10-min period at 1200°C in a flowing steam-helium atmosphere following rupture at approximately 900°C. In the prerupture stage of the test, 308 psig (2.12 MPa) helium pressure was added to the segment at about 745°C. Subsequently, the temperature of the segment was raised at a rate of 5.1°C/sec to 898°C, where rupture occurred; at this point, the pressure was 291 psig (2.01 MPa). A photograph of the ruptured rod is shown in Fig. 1. Details of the rupture parameters have been given previously.²

A scan of the fuel rod segment was made with a mobile, automatic optical pyrometer about 5 min after rupture. This scan yielded the axial temperature profile shown in Fig. 2. As in previous rupture tests, a depression in the temperature occurred at the rupture location because of a loss of inductive coupling. After rupture, there was a gradual but steady decrease in temperature at the rupture fissure; this was probably caused by axial expansion of this opening and by oxidation of the thinned cladding.

A summary of the distribution of released ¹³⁴Cs (deposited as vapor) in the experimental apparatus at the conclusion of the test is given in Table 1. These data show that 0.0611% (280 µg) of the total cesium inventory was released. This is about a factor of 2.5 more cesium released than in test HBU-9, which was conducted at 1100°C. Approximately 88.3% of the released cesium deposited in the furnace tube. The remaining 11.7% was transported downstream to the thermal gradient tube and first filter paper of the filter pack.

As in previous experiments, in which the fuel rod segments were ruptured with helium pressure, fuel dust was ejected. On the basis of ¹⁵⁴Eu analysis (assuming no loss of europium from the fuel particles), it is estimated that 0.022% (46.0 mg) of the fuel was ejected from the rod segment. The manner in which the dust was dispersed throughout the apparatus is summarized in Table 2. About 2% of the ejected fuel was found on the first filter paper of the filter pack. The remainder was found in the quartz furnace tube liner near the rupture location.

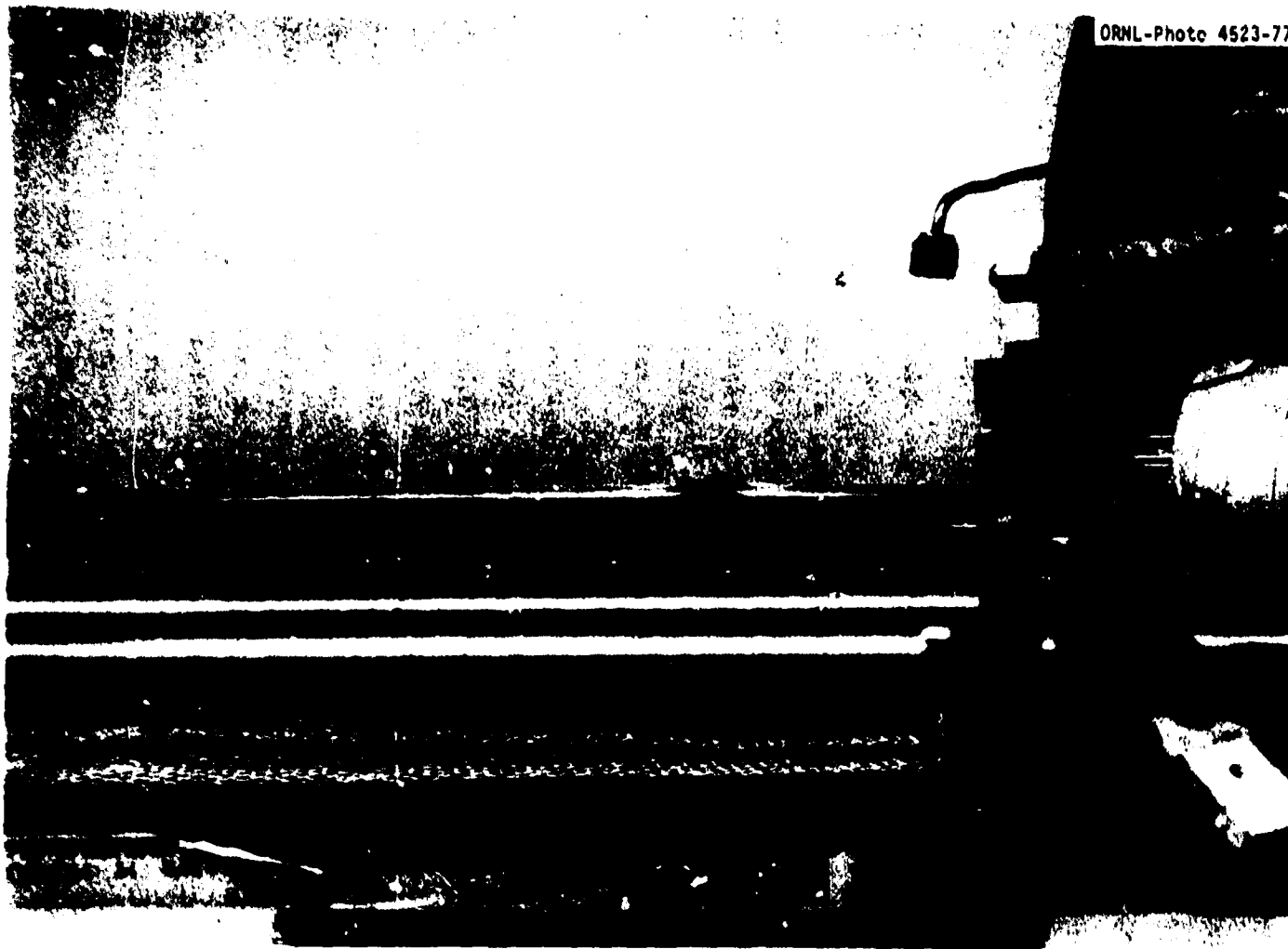


Fig. 1. Post-test view of the rod segment used in test HBU-10, as seen through the cell window.

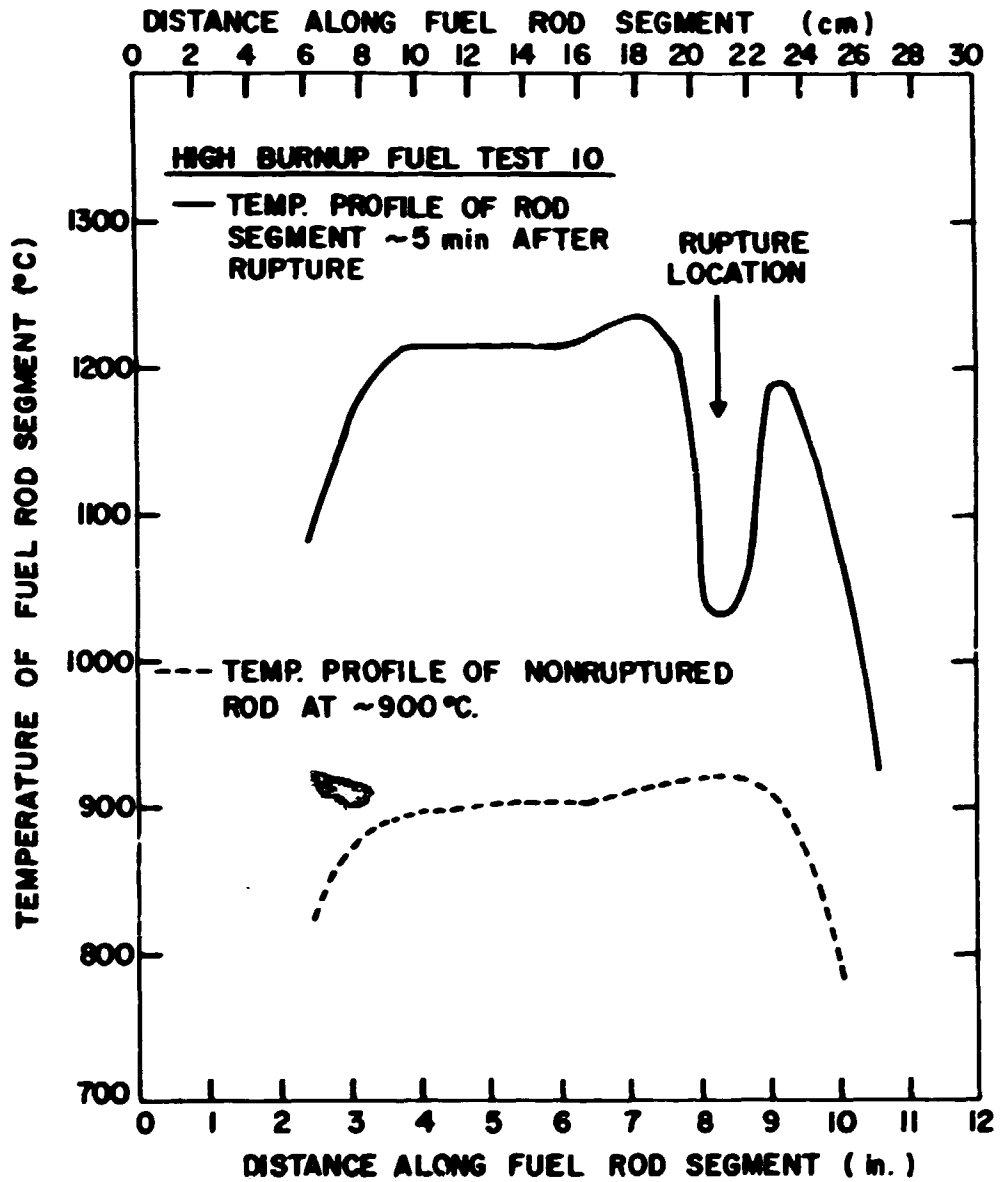


Fig. 2. Axial temperature profile of HBU-10 fuel rod segment during test.

Table 1. Distribution of ^{134}Cs in High Burnup Fuel Test 10^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	900-1200	(8685) ^d			4.572×10^5 ^d
Furnace tube	~ 200-900				
Quartz liner		4.460	5.14×10^{-2}	84.01	234.766
Quartz fuel rod holder		0.228	2.63×10^{-3}	4.29	12.001
Thermal gradient tube	700-225	0.326	3.75×10^{-3}	6.14	17.160
Filter pack components	130				
Stainless steel inlet fitting		0.019	2.19×10^{-4}	0.36	1.000
Other housing components		7.46×10^{-6}	3.44×10^{-7}	1.41×10^{-4}	3.927×10^{-4}
First and second filter papers		0.276	3.18×10^{-3}	5.20	14.528
Third filter paper		0.0	0.0	0.0	0.0
Charcoal No. 1a		1.93×10^{-7}	2.22×10^{-9}	3.64×10^{-6}	1.016×10^{-5}
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 released		5.309	6.11×10^{-2}	100.0	279.455

^a Steam flow rate, 1246 cm³/min (STP); helium flow rate, 368 cm³/min (STP); system pressure, 760 torr.

^b Decay time, 911 days (to November 2, 1976).

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

^d Percent of radioactive nuclide in fuel rod.

^e Calculated for burnup of 29,987 MWd/metric ton of original uranium originally in 12-in. segment, and 911 days decay.

Table 2. Distribution of UO₂ fuel particles released at the time of rupture in High Burnup Fuel Test 10

Location	UO ₂ fuel particles ^a		¹³⁴ Cs present in fuel particles ^b (μg)
	mg	% of released	
Furnace tube			
Quartz liner	41.04 ^c	89.17	1.714
Fuel rod holder	3.64	7.91	0.152
Thermal gradient tube	0.22	0.47	0.09
Filter pack			
Stainless steel inlet fitting	0.12	0.26	0.005
First filter paper	1.01	2.19	0.042
Total	46.03	100.0	1.922 ^d

^aEstimated on the basis of ¹⁵⁴Eu analysis. It was assumed that there was no loss of europium from the UO₂ particles.

^bCalculated based on ¹³⁴Cs/¹⁵⁴Eu ratio in loose fuel particles.

^cLoose particles, 23.52 mg; adhering fuel particles, 18.25 mg.

^dEquivalent to 101 μg of the total cesium.

The distribution of ^{134}Cs (in both vapor-deposited and fuel dust-associated forms) along the furnace tube liner, fuel rod holder, and thermal gradient tube is presented graphically in Fig. 3. (Loose fuel particles were removed from the furnace tube liner prior to gamma scanning.) Analytical estimates indicate that about 16.4% of the ^{134}Cs in the liner, 66.7% on the fuel rod holder, and 2.8% in the thermal gradient tube were associated with fuel dust. Most of the cesium in the furnace tube deposited on the quartz surface of the furnace tube liner in the region of the rupture. (As noted in previous tests, the released cesium reacted readily with the quartz components.) Approximately 6% of the cesium deposited in the thermal gradient tube.

The rate of deposition of ^{134}Cs in the thermal gradient tube and filter pack is graphically presented in Fig. 4. Approximately 8% of this cesium was associated with fuel particles which were found on the first filter paper in the filter pack. [A sample of the particles has been submitted for examination by a scanning electron microscope (SEM) to determine particle size.]

The rate of collection of ^{85}Kr in the cold charcoal traps is also displayed in Fig. 4. It was determined that 1.69% (22.4 mCi) of the total ^{85}Kr in the rod segment was released.

2.2 Comparison of Cesium Release in Steam

Data on the release of cesium as vapor which were determined in the tests with the high-burnup fuel rod segments are summarized graphically in Fig. 5. In experiments that were conducted with already defected rod segments (a 1/16-in.-diam hole was drilled in the cladding at midcapsule), the releases were controlled primarily by gas phase diffusion. The percent cesium released at 900°C in the test with the rod containing the drilled hole was about 50 times less than that released in a 1-min test at 900°C with a pressure-ruptured rod segment. In a second pressure-rupture test at 900°C, which lasted 60 min, a factor of three less cesium was released. For the tests conducted at 900°C, it is obvious that the release of cesium caused by the rapidly venting "plenum" gas at the time of rupture is paramount, and that the amount of cesium released at the time of rupture can vary considerably.

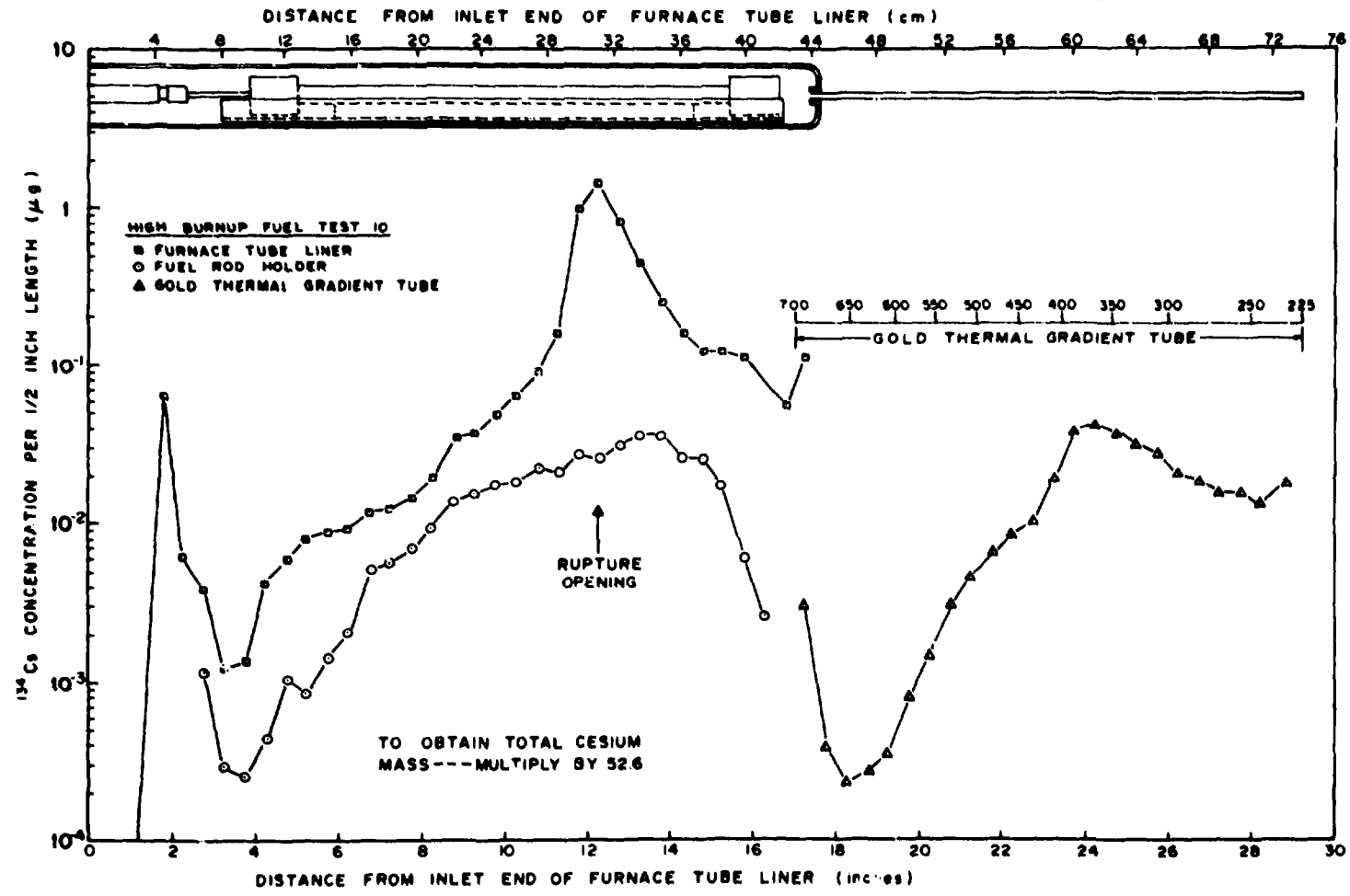


Fig. 3. Distribution of ¹³⁴Cs in furnace tube and thermal gradient tube, test HBU-10.

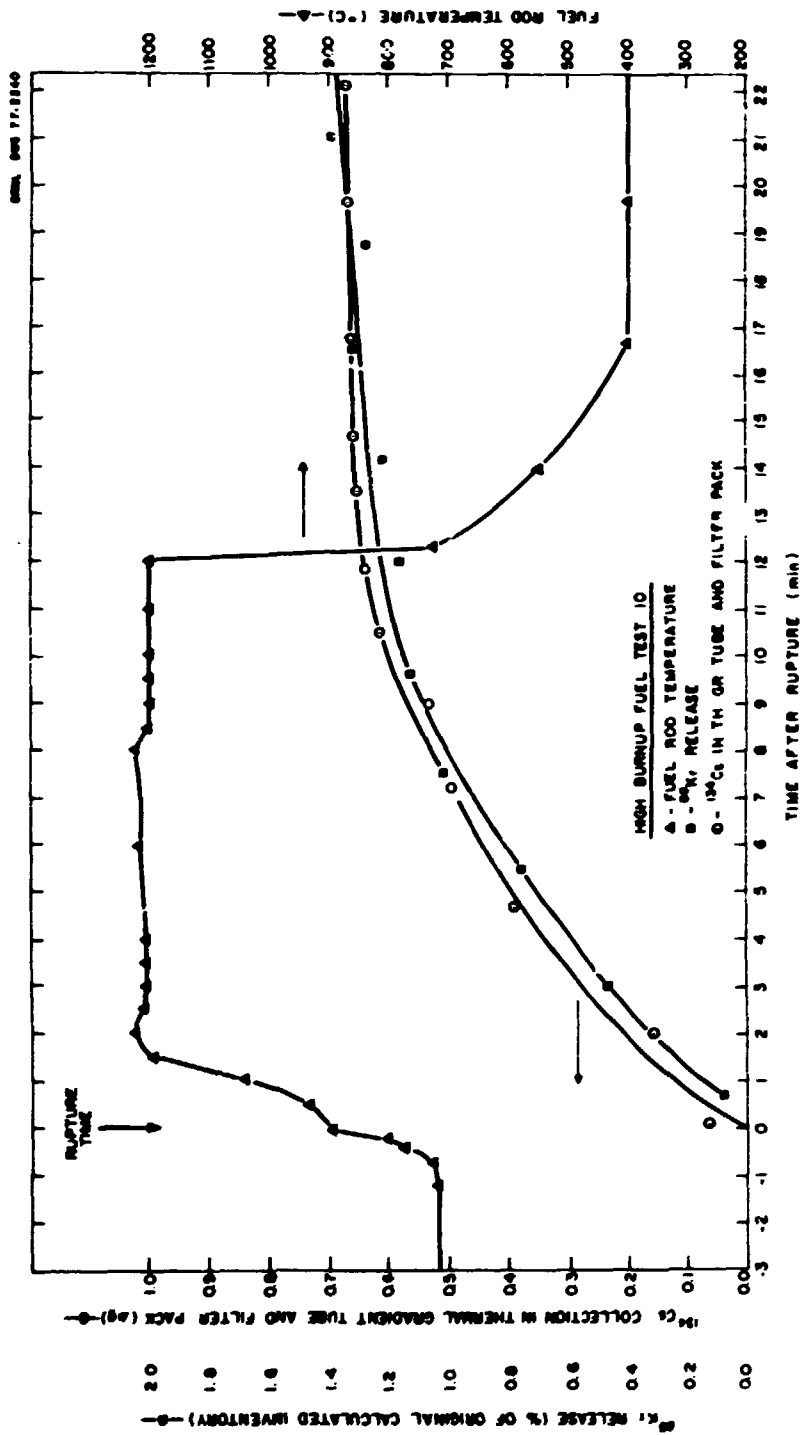


Fig. 4. Collection of ¹³⁴Cs in the thermal gradient tube and filter pack and of ⁸⁵Kr in the cold charcoal traps during High Burnup Fuel Test 10.

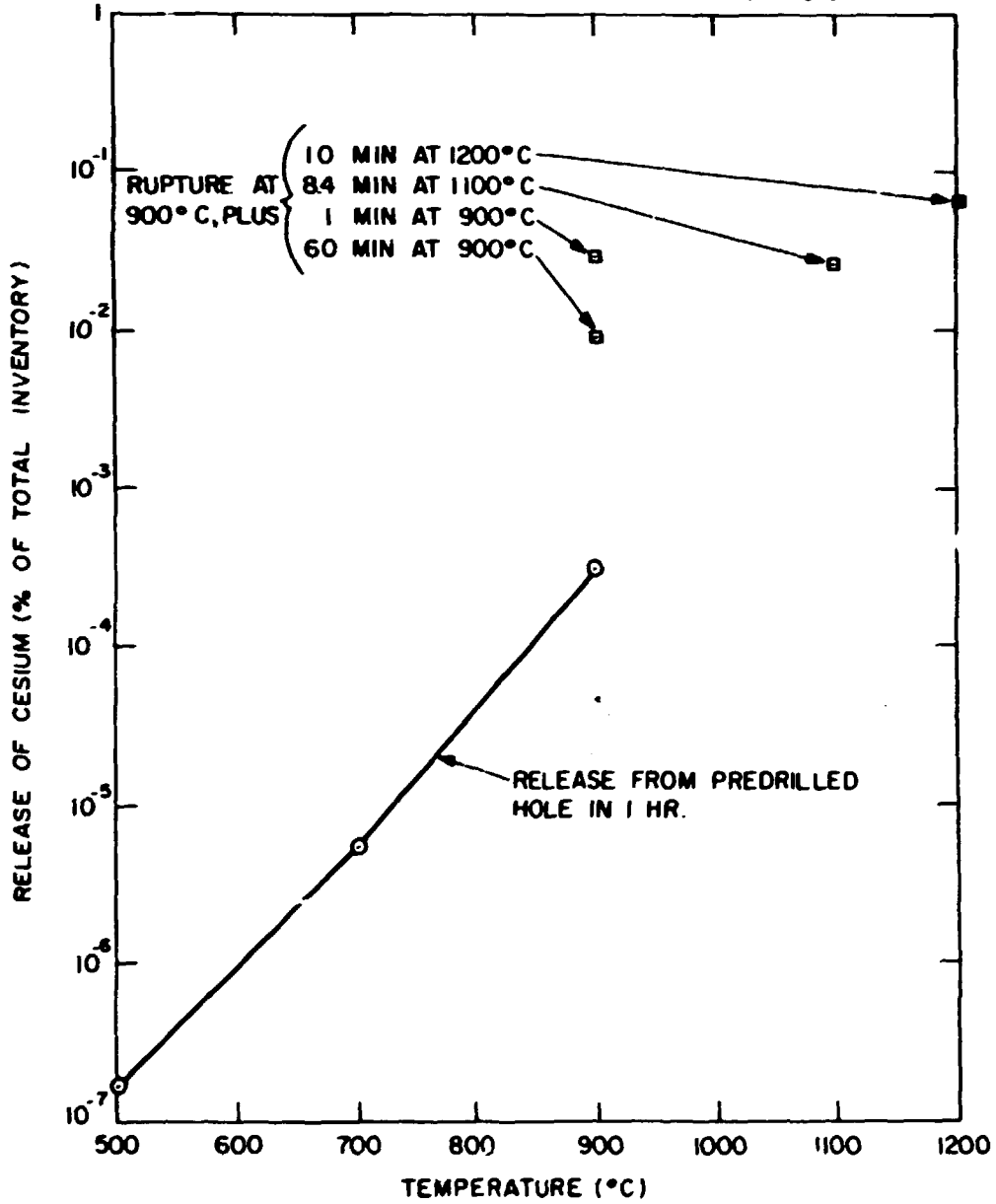


Fig. 5. Release of cesium in vapor form into steam from highly irradiated fuel rod segments.

Diffusive release seems to be of greater importance in the 1200°C test. This is suggested by the two-fold greater release over the 900°C and 1100°C test values. In order to better define the diffusive escape of fission products from fuel rods with expanded and ruptured cladding, we plan to use the fuel rod segment from test HBU-7 (ruptured at 900°C and held at 900°C for only 1 min) in a special experiment. Diffusive release at high temperature will be measured by heating this fuel rod segment to 1200°C for 10 min, using a resistance furnace and a flowing steam atmosphere.

3.0 FISSION PRODUCT TRANSPORT TEST FACILITY

S. K. Whatley

The conceptual design and preliminary cost estimate of a scaled model facility, the Fission Product Transport Test Facility (FPTTF), which could be employed to validate computational models of fission product behavior in the primary circuit of a light water reactor (LWR) under hypothetical accident conditions, were completed and formally transmitted to NRC. A meeting of the Fission Product Review Group has been set to discuss the study.

3.1 Objective of the Facility

The primary objective of the FPTTF is to provide experimental verification of the TRAP (Transport of Radioactivity in Primaries) computer code being developed by Battelle Columbus Laboratories. This code is being developed to describe the behavior of key fission products in the primary system of an LWR during hypothetical accidents which range from controlled loss-of-coolant accidents (LOCAs) to core meltdown. Because of the complexity of the interacting chemical and hydrodynamic phenomena involved, validation of the code on this basis is considered essential. Although TRAP is capable of predicting the amount of fission product deposition within the reactor vessel's internal structure (a part of the primary coolant circuit), the conceptual design of the FPTTF is concerned only with the attenuation of the fission product source term due to processes which occur between the reactor pressure vessel and a postulated rupture in the primary circuit piping.

In addition to utilizing the facility to validate the TRAP code, it should also be possible to utilize the facility to test some of the detailed models incorporated in the computer code.

3.2 Design Criteria

A 1/200 model scale based on cross-sectional flow area and experimental conditions that might be expected at the end of the blowdown phase of a design-basis LOCA for a pressurized water reactor (PWR) was selected for the conceptual design of the FPTTF. With the selected scale and anticipated conditions, the criteria shown in Table 3 were utilized for the preliminary conceptual design. Although emphasis later shifted to core meltdown conditions, the design was too far advanced to make major changes and still meet programmatic commitments. However, areas where design changes would be necessary were examined.

3.3 Description of the FPTTF

A flow diagram of the proposed FPTTF is displayed in Fig. 6. The facility would operate with a steady flow and pressure with test runs up to about 10 min. A radiotracer source would be used to simulate the released fission products.

The test components shown in Fig. 6 (dashed section) simulate a break in the cold leg of a PWR; included are the hot and cold leg pipes, the steam generator, and the primary loop circulator. [At a later date it may be desirable to add a PWR pressurizer and/or boiling water reactor (BWR) components to the test facility.] Although the test components are shown assembled for testing as a single unit in Fig. 6, they can be tested individually, with only minor piping modifications, or can be rearranged to simulate other breaks.

Prior to the initiation of a test run, the test components would be electrically preheated to the initial temperature condition. During the test run the thermal inertia of the walls would provide passive temperature control. Dowtherm is proposed for use on the shell side of the steam generator to provide passive temperature control. (The use of Dowtherm would alleviate the necessity for high-pressure equipment.)

**Table 3. Design criteria based on loss-of-coolant accident
(using 1/200 scale)**

Mass flow rate:	1 lb/sec
Steam conditions:	5 vol % water to dry steam
Steam temperature:	~260°C (500°F)
Initial wall temperature:	~260°C (500°F)
Pressure:	20 to 60 psia

Mass flows through the loop would be controlled by fixing the steam supplies and exit pressures, the latter ranging from 20 to 60 psig. Provision is made for both wet and dry steam supplies. Wet steam, which would be required for simulating controlled LOCA tests, would be provided by a pressurized hot water tank, while the dry steam would be provided by building steam, if available, or by a purchased package boiler.

Because health physics regulations limit the allowable quantities of radioactive tracer material to levels which are unacceptably low for external monitoring, the test components are designed with removable internal surfaces so they may be scanned directly for activity or so that the deposited activity may be removed by chemical dissolution. The steam generator has two removable tubes, the pump has removable inserts, and both the hot and cold leg pipes have spool pieces with removable inserts.

The fission products exiting the test components would be collected in weigh tanks for subsequent sampling for material balances before disposal. A system for decontaminating the interior surfaces of all parts which will come in contact with the fission products is also provided.

3.4 Conclusions and Recommendations

Based on studies and evaluations during the period when the conceptual design and preliminary cost estimate were being prepared, the following conclusions and recommendations were made:

1. Validation experiments for the TRAP code, or similar codes, are deemed essential before any reliance can be placed upon code predictions of the retention of radiologically significant fission products in the primary system of an LWR
2. Although not specifically addressed in the study, it appears expedient to conduct the validation exercise in two stages, one which is concerned with fuel and fission product behavior in the reactor pressure vessel, and a second which focuses on transport and deposition in the primary system external to the pressure vessel. Of the two associated

facilities required to conduct the validation exercise, the latter is intuitively less expensive. Moreover, construction and operation of such a facility can be reasonably expected to have significant impact on the design, construction, and operation of the companion facility.

3. Justification for the construction of the Fission Product Transport Test Facility resides in a commitment to complete the code validation exercise through the construction and operation of a facility which is designed to verify code predictions of the behavior of fission products within the pressure vessel. Without such a commitment, justification for the FPTTF rests solely on the intent of the Nuclear Regulatory Commission to obtain detailed best estimates of fission product releases for accident scenarios other than worst case accidents.
4. An existing facility which can practically function as the FPTTF was not found.
5. The estimated construction cost for building an integral FPTTF (one-time assembly) with the option to test components separately is approximately \$1,241,000 if constructed at an existing experimental area at the Oak Ridge Y-12 site. Since costs are site-dependent, some variation in cost can be expected if other sites are considered.
6. Minor construction cost differences were found between three fabrication and assembly options which were examined, namely, (a) assembly of all components at one time without option for separate component tests; (b) same as (a) except with provision for separate component testing; and (c) piece-by-piece fabrication, testing one component at a time.
7. One-time fabrication, assembly, and testing, as opposed to component-by-component fabrication, assembly, and testing, has the potential for significant savings in operational costs. One to two years' operational cost savings is estimated if the one-time fabrication procedure were adopted.

8. Based on conclusions (6) and (7) above, a one-time fabrication and assembly of a system which has provisions for separate component tests is recommended.
9. The desirability of adding the PWR pressurizer to the model should be examined.
10. Suitable decontamination conditions for removal of selected tracer materials should be experimentally determined. The present design provides for the worst possibility, that is, concentrated nitric acid at 90°C would be needed for decontamination.

4.0 REFERENCES

1. 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).
2. 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).