6
25-77
ANL-76-121

Dr # 761

ANL-76-121

**LIGHT-WATER-REACTOR SAFETY
RESEARCH PROGRAM:
QUARTERLY PROGRESS REPORT**

July–September 1976

AUA-USERDA

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

Prepared for the U. S. NUCLEAR REGULATORY COMMISSION

Office of Nuclear Regulatory Research

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

The facilities of Argonne National Laboratory are owned by the United States Government. Under the terms of a contract (W-31-109-Eng-38) between the U. S. Energy Research and Development Administration, Argonne Universities Association and The University of Chicago, the University employs the staff and operates the Laboratory in accordance with policies and programs formulated, approved and reviewed by the Association.

MEMBERS OF ARGONNE UNIVERSITIES ASSOCIATION

The University of Arizona	Kansas State University	The Ohio State University
Carnegie-Mellon University	The University of Kansas	Ohio University
Case Western Reserve University	Loyola University	The Pennsylvania State University
The University of Chicago	Marquette University	Purdue University
University of Cincinnati	Michigan State University	Saint Louis University
Illinois Institute of Technology	The University of Michigan	Southern Illinois University
University of Illinois	University of Minnesota	The University of Texas at Austin
Indiana University	University of Missouri	Washington University
Iowa State University	Northwestern University	Wayne State University
The University of Iowa	University of Notre Dame	The University of Wisconsin

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, 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. Mention of commercial products, their manufacturers, or their suppliers in this publication does not imply or connote approval or disapproval of the product by Argonne National Laboratory or the U. S. Energy Research and Development Administration.

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
Price: Printed Copy \$6.00; Microfiche \$3.00

Distribution Categories:
Water Reactor Safety Research—
Fuel Behavior (NRC-3)
Water Reactor Safety Research—
Analysis Development (NRC-4)

ANL-76-121

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

LIGHT-WATER-REACTOR SAFETY
RESEARCH PROGRAM:
QUARTERLY PROGRESS REPORT

July-September 1976

Robert G. Sachs, Laboratory Director
Jack A. Kyger, Associate Laboratory Director

December 1976

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, 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.

Previous reports in this series:

ANL-75-28	January-March 1975
ANL-75-58	April-June 1975
ANL-75-72	July-September 1975
ANL-76-15	October-December 1975
ANL-76-49	January-March 1976
ANL-76-87	April-June 1976

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	xv
I. LOSS-OF-COOLANT ACCIDENT RESEARCH: HEAT TRANSFER AND FLUID DYNAMICS	1
A. Transient Critical Heat Flux	1
1. Objectives and System Performance	1
2. Data Acquisition and Reduction.	3
3. Blowdown Results for Test Section I	4
4. Description of Test Section II.	39
B. Nitrogen-Water Blowdown Tests	43
References.	47
II. TRANSIENT FUEL RESPONSE AND FISSION-PRODUCT RELEASE PROGRAM.	48
A. Modeling of Fuel-Fission-product Behavior	50
1. Steady-state LWR Fuel-rod Performance Code Development.	50
a. LIFE-LWR.	50
b. Model Additions and Revisions.	51
c. Materials Properties for Zircaloy Cladding	53
d. UO ₂ Fuel Properties	54
e. Uncertain Parameters	55
f. Description of Standard Fuel-rod Cases	57
g. Results	57
2. Transient Fission-gas Release.	62
a. Rate of Growth of Coalescing Bubbles	62
b. Burst Release of Fission Gas	63
c. GRASS Results for Fission-gas Release under DEH Test Conditions.	64
d. Model Development	65
3. FRAP Code Development.	66
B. Experimental Program	68
1. Transient-heating Tests	68
a. Test Conditions.	68
b. Fission-gas Release during Transient Heating	70

TABLE OF CONTENTS

	<u>Page</u>
2. Fuel Characterization	72
a. Posttest Characterization	72
b. Radial Distribution of Fission Gas	75
c. Pretest Total Gas and Fission-product Analyses	75
3. Experimental Technique	79
References	79
III. MECHANICAL PROPERTIES OF ZIRCALOY CONTAINING	
OXYGEN.	81
A. Summary	81
B. Mechanical Properties of As-received Zircaloy	82
1. Comparison of the Tensile Properties of Stress-relieved Tubing and Recrystallized Sheet	82
2. Activation Energy for the Deformation of Zircaloy-4 at High Temperatures	84
C. Biaxial Deformation and Rupture Characteristics of As- received Zircaloy-4 Cladding In Vacuo	90
1. Rupture Characteristics of Unconstrained Cladding at a Low Heating Rate	90
2. Effect of Axial Constraint on the Deformation Behavior of Zircaloy Cladding	90
D. Effect of Steam Oxidation on the Biaxial Deformation of Zircaloy-4 Cladding	93
1. Diametral Expansion Characteristics of Zircaloy-4 Cladding in Steam at Different Heating Rates	93
2. Effect of Steam Oxidation on the Deformation Behavior of Constrained and Unconstrained Zircaloy-4 Cladding at a High Heating Rate	94
3. Effect of Steam Oxidation on the Deformation Behavior of Constrained and Unconstrained Cladding at a Low Heating Rate	97
4. Stress-rupture Characteristics of Zircaloy-4 Cladding in Steam and Vacuum Environments	101
References	105

TABLE OF CONTENTS

	<u>Page</u>
IV. STEAM-EXPLOSION STUDIES	107
A. Vapor Explosions at Elevated Pressures	107
B. Combined Physical-Chemical Explosions	110
References	113

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.1.	Schematic of Test Section I	2
I.2.	System Pressure during Test DB-1	5
I.3.	Differential Pressure across Test Section during Test DB-1 . . .	5
I.4.	Differential Pressure across Break Orifice during Test DB-1 . .	6
I.5.	Fluid Temperature during Test DB-1	6
I.6.	Wall-thermocouple Measurements during Test DB-1	7
I.7.	System Pressure during Test DB-2	8
I.8.	Differential Pressure across Test Section during Test DB-2 . . .	9
I.9.	Differential Pressure across Break Orifice during Test DB-2 . .	9
I.10.	Fluid Temperature during Test DB-2	10
I.11.	Wall-thermocouple Measurements during Test DB-2	10
I.12.	System Pressure during Test DB-5	12
I.13.	Differential Pressure across Test Section during Test DB-5 . . .	12
I.14.	Differential Pressure across Break Orifice during Test DB-5 . .	13
I.15.	Turbine-flowmeter Measurement during Test DB-5	13
I.16.	Fluid Temperature during Test DB-5	14
I.17.	Wall-thermocouple Measurements during Test DB-5	14
I.18.	System Pressure during Test DB-6	16
I.19.	Differential Pressure across Test Section during Test DB-6 . . .	16
I.20.	Differential Pressure across Break Orifice during Test DB-6 . .	17
I.21.	Turbine-flowmeter Measurement during Test DB-6	17
I.22.	Fluid Temperature during Test DB-6	18
I.23.	Wall-thermocouple Measurements during Test DB-6	18
I.24.	System Pressure during Test DB-7	20
I.25.	Differential Pressure across Test Section during Test DB-7 . . .	20
I.26.	Differential Pressure across Break Orifice during Test DB-7 . .	21
I.27.	Turbine-flowmeter Measurement during Test DB-7	21
I.28.	Fluid Temperature during Test DB-7	22
I.29.	Wall-thermocouple Measurements during Test DB-7	22

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.30.	System Pressure during Test DB-8	24
I.31.	Differential Pressure across Test Section during Test DB-8 . . .	24
I.32.	Differential Pressure across Break Orifice during Test DB-8 . .	25
I.33.	Turbine-flowmeter Measurement during Test DB-8	25
I.34.	Fluid Temperature during Test DB-8.	26
I.35.	Wall-thermocouple Measurements during Test DB-8	26
I.36.	System Pressure during Test DB-9	28
I.37.	Differential Pressure across Test Section during Test DB-9 . . .	28
I.38.	Differential Pressure across Break Orifice during Test DB-9 . .	29
I.39.	Turbine-flowmeter Measurement during Test DB-9	29
I.40.	Fluid Temperature during Test DB-9.	30
I.41.	Wall-thermocouple Measurements during Test DB-9	30
I.42.	System Pressure during Test AB-5	35
I.43.	Differential Pressure across Test Section during Test AB-5 . . .	36
I.44.	Differential Pressure across Break Orifice during Test AB-5 . .	36
I.45.	Turbine-flowmeter Measurement during Test AB-5	37
I.46.	Fluid Temperature during Test AB-5.	37
I.47.	Wall-thermocouple Measurements during Test AB-5	38
I.48.	Schematic of Test Section II	40
I.49.	Heat-flux Profile for Test Section II	41
I.50.	Dimensions of Test Section II	41
I.51.	Photograph of Thermocouple Bead and Mica	41
I.52.	Thermocouple Responses in a Jet Flame Test	42
I.53.	Dimensions of the Three Discharge Geometries and Locations of Pressure Taps	43
I.54.	Measured Pressures for Test Section B without a Tube Bundle and with Negligible Dissolved Gas	43
I.55.	Measured Pressures for Test Section B without a Tube Bundle and with 24-hr Gas Bubbling Time.	43
I.56.	Measured Pressures for Test Section B with a Tube Bundle and Negligible Dissolved Gas	44

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.57.	Measured Pressures for Test Section B with a Tube Bundle and 24-hr Gas Bubbling Time.	44
I.58.	Measured Pressures for Test Section C without a Tube Bundle and with Negligible Dissolved Gas.	44
I.59.	Measured Pressures for Test Section C without a Tube Bundle and with 24-hr Gas Bubbling Time	44
I.60.	Measured Pressures for Test Section C with a Tube Bundle and Negligible Dissolved Gas.	44
I.61.	Measured Pressures for Test Section C with a Tube Bundle and 24-hr Gas Bubbling Time.	44
II.1.	Fuel Surface and Centerline Temperatures in Three Interrupted Transient Tests.	69
II.2.	Gas Release vs Time of Transient Power Application at the Slowest Heating Rate	71
II.3.	Gas Release vs Heating Rate for Tests to Center Melting	71
II.4.	Fuel from Transient Test 22 Showing Grain-boundary Separations and Tearing without Fuel Melting	72
II.5.	Polarized-light Photograph of Cesium Precipitate at Grain Boundaries of Transient-heated Fuel.	74
II.6.	Autoradiograph of a Polished Cross Section of Transient-heated Fuel.	74
II.7.	Measurements of Radial Fission-gas Distribution by Laser Sampling	75
III.1.	Ultimate Tensile Strength and 0.2% Yield Stress of Zircaloy-4 Stress-relieved Tube and Recrystallized Sheet Material as a Function of Temperature.	83
III.2.	Total Strain of Zircaloy-4 Tube and Recrystallized Sheet as a Function of Temperature	83
III.3.	Uniform Strain of Zircaloy-4 Tube and Recrystallized Sheet as a Function of Temperature	84
III.4.	Strain-rate Dependence of the 0.2% Yield Stress of 5- μ m Grain-size Zircaloy-4 at Several Temperatures between 700 and 950°C	85
III.5.	Strain-rate Dependence of the 0.2% Yield Stress of 11- μ m Grain-size Zircaloy-4 at Several Temperatures between 700 and 1000°C	85

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
III.6.	Temperature Dependence of the Strain Rate for Stage III Deformation of 5- μm Grain-size Zircaloy-4	86
III.7.	Temperature Dependence of the Strain Rate for Stage III Deformation of 11- μm Grain-size Zircaloy-4	86
III.8.	Logarithmic Plot of the Strain Rate as a Function of Grain Size for Stage II Deformation of Zircaloy-4 at 850°C.	87
III.9.	Comparison of the Calculated and Experimental Strain-rate Dependence of the 0.2% Yield Stress for 5- and 11- μm Grain- size Zircaloy-4 at 850 and 900°C.	88
III.10.	Micrograph of the Surface of a 5- μm Grain-size Zircaloy-4 Specimen Deformed at a Strain Rate of $3.3 \times 10^{-4} \text{ s}^{-1}$ at 900°C That Shows Offsets of the Inscribed Scratches at the Grain Boundaries	88
III.11.	Microstructure of an 11- μm Grain-size Specimen Deformed to 0.7% Strain at a Strain Rate of $3.0 \times 10^{-1} \text{ s}^{-1}$ at 850°C	89
III.12.	Maximum Circumferential Strain vs Burst Temperature for Axially Unconstrained Zircaloy-4 Cladding In Vacuo at Heating Rates of 5 and 115°C/s	90
III.13.	Average Axial Strain vs Burst Temperature for Unconstrained Zircaloy-4 Cladding In Vacuo at Heating Rates of 5 and 115°C/s	91
III.14.	Effect of the Degree of Axial Constraint on the Maximum Diametral Strain during the Ballooning Deformation of Zircaloy-4 Cladding In Vacuo at a Heating Rate of 115°C/s	91
III.15.	Maximum Circumferential Strain vs Burst Temperature for Three Axial-constraint Conditions at a Heating Rate of 115°C/s In Vacuo	92
III.16.	Maximum Circumferential Strain vs Burst Temperature for Axially Constrained and Unconstrained Zircaloy-4 Cladding at a Heating Rate of 5°C/s In Vacuo	92
III.17.	Diametral Strain vs Temperature for Axially Constrained Zircaloy-4 Cladding at Different Internal Pressures at a Heating Rate of 115°C/s in Steam	93
III.18.	Diametral Strain vs Temperature for Axially Constrained Zircaloy-4 Cladding at Different Internal Pressures at a Heating Rate of 55°C/s in Steam	93

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
III.19.	Diametral Strain vs Temperature for Axially Constrained Zircaloy-4 Cladding at Different Internal Pressures at a Heating Rate of 5°C/s in Steam	94
III.20.	Burst Temperature as a Function of Initial Internal Pressure for Axially Constrained Zircaloy-4 Cladding in Steam and Vacuo and for Axially Unconstrained Cladding in Steam at a Heating Rate of 115°C/s.	94
III.21.	Maximum Circumferential Strain vs Burst Temperature for Unconstrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 115°C/s.	95
III.22.	Maximum Circumferential Strain vs Burst Temperature for Axially Constrained As-received Cladding in Steam and Vacuo and Preoxidized Cladding Ruptured In Vacuo at a Heating Rate of 115°C/s.	96
III.23.	Cross Section of the Fracture Location of Axially Constrained Zircaloy-4 Cladding that Burst at 825°C In Vacuo and in Steam at a Heating Rate of 115°C/s.	96
III.24.	Scanning-electron Micrographs of the Fracture Edge of the Zircaloy-4 Cladding Shown in Fig. III.23 That Burst In Vacuo and Steam at a Heating Rate of 115°C/s	96
III.25.	"Rupture" and "Fracture" Failure-mode Classifications for the Deformation Data in Fig. III.22 on Axially Constrained Zircaloy-4 Cladding	97
III.26.	Maximum Diametral Strain as a Function of Time before Rupture for Axially Constrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 115°C/s	98
III.27.	Maximum Diametral Strain as a Function of Time before Rupture for Axially Constrained As-received Zircaloy-4 Cladding in Steam and Vacuo and Preoxidized Cladding In Vacuo at a Heating Rate of 115°C/s.	98
III.28.	Burst Temperature as a Function of Initial Internal Pressure for Axially Constrained and Unconstrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 5°C/s.	98
III.29.	Maximum Circumferential Strain vs Burst Temperature for Axially Unconstrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 5°C/s	99

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
III.30.	Maximum Diametral Strain as a Function of Time before Rupture for Axially Constrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 5°C/s	100
III.31.	Schematic Representation of the Effect of Axial Constraint on the Biaxiality and Tangential Strain in the Plastic-instability Region of a Zircaloy Tube Burst in Vacuum and Steam Environments	100
III.32.	Average Axial Strain vs Burst Temperature for Unconstrained Zircaloy-4 Cladding In Vacuo and Steam at a Heating Rate of 5°C/s	101
III.33.	Maximum Diametral Strain as a Function of Time in Isothermal Stress-rupture Tests on Unconstrained As-received and Preoxidized Zircaloy-4 Cladding In Vacuo at 750°C and an Internal Pressure of 5.2 MPa	102
III.34.	Localized Ballooning Characteristics and Diametral and Axial Strains during an Isothermal Stress-rupture Test on Preoxidized Zircaloy-4 Cladding In Vacuo at 751°C and an Internal Pressure of 5.2 MPa	103
III.35.	Maximum Diametral Strain as a Function of Time in Isothermal Stress-rupture Tests on Unconstrained As-received Zircaloy-4 Cladding in Steam and Vacuo and Preoxidized Cladding In Vacuo at 750°C and an Internal Pressure of 4.2 MPa	103
III.36.	Comparison of Maximum Circumferential Strain vs Burst-temperature Data Obtained from Short-term Isothermal Stress-rupture Tests on Constrained Zircaloy-4 Cladding in Steam and Vacuo with Similar Data from Transient-heating Experiments at 115°C/s in the Two Environments	104
III.37.	Photographs of Zircaloy-4 Cladding from the Isothermal Stress-rupture Tests	104
IV.1.	Inertially and Thermally Dominated Bubble-growth Behavior . .	107
IV.2.	High-pressure Apparatus	109
IV.3.	High-ambient-pressure Freon/Mineral-oil Data	110
IV.4.	High-ambient-pressure Freon/Water Data	110
IV.5.	Impact Apparatus	111
IV.6.	Explosion-initiated Apparatus	112

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.1.	Dimensions of Test Section I	3
I.2.	Location of Instrumentation.	3
I.3.	Steady-state Test Conditions in Test Section I before Blowdown.	32
I.4.	Summary of Time to CHF during Blowdown in Test Section I.	34
I.5.	Blowdown Time for Discharge Nozzle B without Tube Bundle inside the Vessel.	45
I.6.	Blowdown Time for Discharge Nozzle B with Tube Bundle inside the Vessel.	46
I.7.	Blowdown Time for Discharge Nozzle C without Tube Bundle inside the Vessel.	46
I.8.	Blowdown Time for Discharge Nozzle C with Tube Bundle inside the Vessel.	46
II.1.	LIFE-LWR Calculated Fuel Centerline Temperature vs Peak Power for Halden IFA-226 Rod AA.	58
II.2.	LIFE-LWR Calculated Change in Cladding Length for Halden IFA-226 Rod AK as a Function of Peak Linear Power for the First Ramp.	58
II.3.	LIFE-LWR Results for Case D Assuming a 7000-h History.	59
II.4.	Comparison between LIFE-LWR Results and Experimental Data for Case D Assuming a 7000-h History.	59
II.5.	LIFE-LWR Results for Case D Assuming the Correct 10,555-h History	60
II.6.	Comparison between LIFE-LWR Results and Experimental Data for Case D Assuming the Correct 10,055-h History	61
II.7.	Preliminary GRASS Sensitivity Analyses for DEH Test Conditions	65
II.8.	Conditions of Interrupted DEH Transient Tests on H. B. Robinson Fuel.	69
II.9.	Fission Gas Collected from Interrupted DEH Transient Tests	70
II.10.	Chemical Analyses of Gases Recovered by Fuel Dissolution	76
II.11.	Quantities of Gas Collected from Dissolutions of 8.6 g of Fuel	76
II.12.	Isotopic Analysis of Fission Gas in H. B. Robinson Fuel.	77

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
II.13.	Quantities of Radionuclides in High-burnup Portion of H. B. Robinson Fuel.	77
II.14.	Loss of Radioactive Isotopes from Fuel Solution during Fission-gas Extraction.	78
II.15.	Fractional Fission-product Release to Quartz Container during a DEH Test	78

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

ABSTRACT

This progress report summarizes the Argonne National Laboratory work performed during July, August, and September 1976 on water-reactor-safety heat-transfer and flow problems. The following research and development areas are covered: (1) Loss-of-coolant Accident Research: Heat Transfer and Fluid Dynamics; (2) Transient Fuel Response and Fission-product Release Program; (3) Mechanical Properties of Zircaloy Containing Oxygen; and (4) Steam-explosion Studies.

I. LOSS-OF-COOLANT ACCIDENT RESEARCH: HEAT TRANSFER AND FLUID DYNAMICS

Responsible Section Managers:

M. A. Grolmes, R. E. Henry, and P. A. Lottes, RAS

A. Transient Critical Heat Flux (J. C. Leung and R. E. Henry, RAS)

1. Objectives and System Performance

One of our major objectives is to perform definitive Freon blow-down experiments in an attempt to understand the basic mechanism governing the rapid thermal excursion or critical heat flux, CHF (less than 500 msec), during blowdown, as experienced in semiscale tests. The present system is small-scale, heavily instrumented, and simple enough for the analysis of the results.

Test Section I, in Fig. I.1, has a 0.91-m (3-ft) heated length, with wall-thermocouple locations as shown. The ultrasonic wall-thickness test gave a mean wall thickness of 0.081 ± 0.002 mm (0.0205 ± 0.0005 in.) and showed no thin spots. As many as nine blowdown tests have been conducted and the results of seven tests are reported, together with an adiabatic blowdown for comparison. Before the transient, the hot Freon-11 flows upward through the heated test section for some time (~5 min) to achieve steady-state conditions. Blowdown was initiated by three quick-acting air-operated ball valves, which were in turn actuated simultaneously by solenoid valves. Their performance in the hot Freon experiment has been satisfactory.

One of the precautions in conducting a test was in the problem of nitrogen coming out of solution. This effect was previously observed in the low-pressure system.¹ The same kinds of measures¹ to eliminate significant dissolved gas were again observed. If dissolved nitrogen gas had come out of solution during steady-state operation, the presence of a two-phase mixture at the top of the section would result in a higher exit-flowmeter output than could be accounted for solely by the liquid-density change. The fact that the mass flow rate at inlet and outlet showed good agreement during these tests demonstrates the success of the precautions taken.¹

A test was performed with the Freon at 110°C (230°F) being pressurized by nitrogen at 2.76 MPa (400 psia) in the supply vessel while a steady 1-m/sec flow was maintained in the heated test section. Wall thermocouples indicated that subcooled boiling was taking place over half the heated section. Initially, there was good agreement between the inlet and exit mass flow rates until the supply tank was two-thirds empty, at which time (~20 min) the exit flowmeter indicated a rapid increase in flow to 2 m/sec while the exit bulk temperature was still subcooled. This indicates significant nitrogen-gas evolution.

Some pertinent dimensions of the test section and the location of various measurements are shown in Tables I.1 and I.2, respectively.

Fig. I.1. Schematic of Test Section I (1 in. = 2.54 cm). ANL Neg. No. 900-76-560.

TABLE I.1. Dimensions of Test Section I

SS 304 heater OD	1.273 cm (0.501 in.)
SS 304 heater ID	1.168 cm (0.460 ± 0.001 in.)
Heated length	91.4 cm (36 in.)
Heated-length L/D	78
Calming length at each end	17.8 cm (7.0 in.)
Calming-length L/D	15
Flow area	1.07 cm ² (0.166 in. ²)
Heated surface area	0.0335 m ² (0.361 ft ²)
Volume of heated zone	98.0 cm ³ (5.98 in. ³)
Volume below heated zone	65.5 cm ³ (4.00 in. ³)
Volume above heated zone for	
DB-1, -2, -5, and -6	61.0 cm ³ (3.72 in. ³)
DB-7	88.8 cm ³ (5.42 in. ³)
DB-8 and -9	177.6 cm ³ (10.84 in. ³)

TABLE I.2. Location of Instrumentation

Instrument	Symbol	Location, ^a cm (in.)
Lower pressure transducer	PT1	-5.1 (-2)
Upper pressure transducer	PT2	+96.5 (+38)
Test-section DP cell	DP1	Between -5.1 (-2) and +96.5 (+38)
Break-orifice DP cell	DP2	across the orifice
Lower turbine flowmeter	TV1	-25.4 (-10)
Upper turbine flowmeter	TV2	+116.8 (+46)
Inlet-fluid thermocouple	TF1	-5.1 (-2)
Outlet-fluid thermocouple	TF2	+96.5 (+38)
Wall thermocouples	TC 'X'	'X' means +X in. above Z = 0 line.

^a+ means above the Z = 0 line (see Fig. I.1); - means below the Z = 0 line. However, radial orientation is not indicated.

2. Data Acquisition and Reduction

The data-acquisition system consists mainly of two 14-channel FM tape recorders running at 1.52 m/sec (60 in./sec). All transient signals listed in Table I.2 were first individually conditioned to the level compatible with the

tape recorders. A 65.6°C (150°F) reference junction box was used as the compensation unit for all the thermocouple signals before they were amplified. A COX flow converter was used with each turbine flowmeter so that the rotor speed in cycles per second is converted and amplified to the 0-5-V range. For each test, the calibration of amplifiers, initial steady-state conditions, and the subsequent transient were all stored on tape. The signals would then be played back onto a visicorder oscillograph, subsequently digitized, and stored on a computer disk or cards. The computer converted into SI and engineering units. The data so reduced were plotted using straight lines joining adjacent digitized points by the IBM plotter.

3. Blowdown Results for Test Section I

For the rig presently constructed, the following parameters could be systematically varied:

- a. Test-section heat flux.
- b. Inlet temperature.
- c. Inlet mass velocity,
- d. Test-section pressure.
- e. Break orifice size.
- f. Volume above the heated section.

Our first objective was to study the effect of initial pressure, and hence the subcooled boiling length, on transient CHF behavior while all other parameters remained constant. Provided the wall heat flux was high enough to cause local subcooled boiling while the exiting bulk liquid still remained subcooled, then the lower initial pressure test would have a longer subcooled boiling length as compared to the high-pressure test during normal steady-state conditions. However, during the early portion of blowdown, one would suspect the pressure decay profile to be comparable in both cases.

The results of seven blowdown tests are presented in Figs. I.2-I.41 and their corresponding steady-state conditions are listed in Table I.3. Test DB-1 was performed with single-phase forced-convection upflow in the heated section during steady state. From a reduced-pressure standpoint, the initial pressure of 3.07 MPa is equivalent to 15.5 MPa (2250 psia) in the water system. Test DB-2 was run at a lower pressure, but higher inlet temperature, resulting in subcooled boiling over the entire heated section.

The pressure transient was very different between these tests. In DB-1, the pressure showed a sharp undershoot and a long recovery period immediately after blowdown. This is indicative of the reactivation of nucleation sites at the heated wall and the subsequent bubble generation.²

Fig. 1.2. System Pressure during Test DB-1

Fig. 1.3. Differential Pressure across Test Section during Test DB-1

Fig. I.4. Differential Pressure across Break Orifice during Test DB-1

Fig. I.5. Fluid Temperature during Test DB-1

Fig. I.6. Wall-thermocouple Measurements during Test DB-1

Fig. I.6 (Contd.)

Fig. I.6 (Contd.)

Fig. I.7. System Pressure during Test DB-2

Fig. I.8. Differential Pressure across Test Section during Test DB-2

Fig. I.9. Differential Pressure across Break Orifice during Test DB-2

Fig. I.10. Fluid Temperature during Test DB-2

Fig. I.11. Wall-thermocouple Measurements during Test DB-2

Fig. I.11 (Contd.)

Fig. I.11 (Contd.)

Fig. 1.12. System Pressure during Test DB-5

Fig. 1.13. Differential Pressure across Test Section during Test DB-5

Fig. I.14. Differential Pressure across Break Orifice during Test DB-5

Fig. I.15. Turbine-flowmeter Measurement during Test DB-5

Fig. I.16. Fluid Temperature during Test DB-5

Fig. I.17. Wall-thermocouple Measurements during Test DB-5

Fig. I.17 (Contd.)

Fig. I.17 (Contd.)

Fig. I.18. System Pressure during Test DB-6

Fig. I.19. Differential Pressure across Test Section during Test DB-6

Fig. I.20. Differential Pressure across Break Orifice during Test DB-6

Fig. I.21. Turbine-flowmeter Measurement during Test DB-6

Fig. I.22. Fluid Temperature during Test DB-6

Fig. I.23. Wall-thermocouple Measurements during Test DB-6

Fig. I.23 (Contd.)

Fig. I.23 (Contd.)

Fig. I.24. System Pressure during Test DB-7

Fig. I.25. Differential Pressure across Test Section during Test DB-7

Fig. I.26. Differential Pressure across Break Orifice during Test DB-7

Fig. I.27. Turbine-flowmeter Measurement during Test DB-7

Fig. I.28. Fluid Temperature during Test DB-7

Fig. I.29. Wall-thermocouple Measurements during Test DB-7

Fig. I.29 (Contd.)

Fig. I.29 (Contd.)

Fig. I.30. System Pressure during Test DB-8

Fig. I.31. Differential Pressure across Test Section during Test DB-8

Fig. I.32. Differential Pressure across Break Orifice during Test DB-8

Fig. I.33. Turbine-flowmeter Measurement during Test DB-8

Fig. 1.34. Fluid Temperature during Test DB-3

Fig. 1.35. Wall-thermocouple Measurements during Test DB-8

Fig. I.35 (Contd.)

Fig. I.35 (Contd.)

Fig. I.36. System Pressure during Test DB-9

Fig. I.37. Differential Pressure across Test Section during Test DB-9

Fig. I.38. Differential Pressure across Break Orifice during Test DB-9

Fig. I.39. Turbine-flowmeter Measurement during Test DB-9

Fig. I.40. Fluid Temperature during Test DB-9

Fig. I.41. Wall-thermocouple Measurements during Test DB-9

Fig. I.41 (Contd.)

Fig. I.41 (Contd.)

TABLE I.3. Steady-state Test Conditions in Test Section I before Blowdown

Test No. (date)	Pressure, kPa (psia)	Fluid Temperature, °C (°F)		Inlet Velocity, m/sec (ft/sec)	Flow Rate, kg/m ² · sec (lb _m /hr · ft ²)	Heat Flux, kW/m ² (Btu/hr · ft ²)
		Inlet	Outlet			
DB-1 (7-8-76)	3070 (445)	128 (262)	139 (282)	1.01 (3.3)	1195 (8.81 × 10 ⁵)	35.7 (11,300)
DB-2 (7-16-76)	2730 (396)	151 (303)	159 (319)	0.98 (3.2)	1066 (7.86 × 10 ⁵)	35.7 (11,300)
DB-5 (7-27-76)	2880 (418)	138 (280)	145 (293)	1.19 (3.9)	1370 (1.01 × 10 ⁶)	65.6 (20,800)
DB-6 (7-27-76)	3050 (443)	138 (280)	145 (293)	1.19 (3.9)	1370 (1.01 × 10 ⁶)	65.6 (20,800)
DD-7 (7-29-76)	3050 (442)	125 (257)	133 (272)	0.85 (2.8)	1001 (7.38 × 10 ⁵)	35.7 (11,300)
DB-8 (8-24-76)	2310 (335)	137 (278)	148 (299)	1.19 (3.9)	1370 (1.01 × 10 ⁶)	44.8 (14,220)
DB-9 (8-27-76)	3030 (440)	138 (280)	148 (298)	1.19 (3.9)	1350 (9.95 × 10 ⁵)	46.2 (14,660)

To gain an appreciation, one can estimate the size of surface cavities that remained unflooded during the steady-state operation assuming a simple reentrant cavity model. Thus the Laplace equation for deactivation is

$$r_c = \frac{2\sigma}{P_l - P_v}, \quad (1)$$

where r_c is the critical cavity radius, σ is the surface tension, P is the pressure, and the subscripts l and v represent liquid and vapor, respectively. The value of r_c is on the order of 8×10^{-7} cm or 80 Å. For these sites to be activated during depressurization, the pressure has to drop significantly so as to satisfy the equation for activation,

$$r_c = \frac{2\sigma}{P_v - P_l}. \quad (2)$$

The initial wall temperature in the middle of the heated section was 154°C (310°F), which corresponds to a vapor pressure of 2.28 MPa (330 psia). The system pressure for activation of an 80-Å cavity was calculated to be 1.38 MPa (200 psia), which agrees well with experimental measurement. Test DB-2, however, showed little pressure undershoot and a very short recovery, because there were preexisting nucleation sites during subcooled boiling before the transient. Because, as a result of the higher initial exit temperature, the pressure at the end of subcooled blowdown in DB-2 was much higher than that in DB-1, the break flow was higher, resulting in a shorter transient.

Tests DB-5 and -6 were conducted with identical conditions, except for a slight difference in operating pressure. The initial pressure in DB-5 was

156 kPa (23 psi) higher, equivalent to a 4°C (7°F) difference in saturation temperature. The transient data indicate that the system pressure, the test-section pressure drop, the pressure differential across the break, and the upper-turbine-flowmeter measurement are almost identical for these two tests, while the lower turbine flowmeter exhibited good agreement during the initial 2 sec. No significant difference was observed in the transient wall-temperature behavior.

During flow reversal, the rotor in the turbine flowmeter is expected to reverse its direction. But since the magnetic pickup coil does not differentiate the direction of rotation, one would not observe a change in polarity of the output voltage. Hence, an ideal flow reversal would be indicated by a sharp drop to zero and an immediate rise in the flow measurement. But the lower turbine flowmeter only showed a slight dip in output. This is thought to have been caused by the attenuation during signal processing when the flow converter actually integrated the input pulse by an operational amplifier to provide an analog voltage output.³ The time interval of flow reversal was therefore too short for the circuit to detect the flow stagnation. It is difficult to interpret the actual flow direction at the upper turbine meter during the initial 0.5 sec when the flow was low and, in fact, beyond the operating range of the meter.

At the end of subcooled blowdown, the highest potential region for vapor generation is found at the upper portion of the heated section, since the wall temperature is highest. The pressure in the system would then be determined by the rate of discharge at the break and the rate of volume generation in the test section. Until the heated section lost much of its liquid, the liquid, which was initially stored above the heated zone, would remain stagnant. This time is indicated by a sharp rise of the upper flowmeter at about 300 msec in Tests DB-5 and -6. This time also indicates the transition from bubbly flow to annular dispersed flow, as observed by the maximum in the transient pressure measurements.

The initial conditions for DB-7 were comparable with those for DB-1, except that the volume above the heated section was increased. However, the time of the blowdown transient in DB-7 was about 0.5 sec longer. The availability of more liquid coming down through the heated section caused rewetting and continued cooling in the TC2-TC9 locations, where DB-1 had experienced sustained thermal excursions. The time to delayed CHF is essentially about the same in both cases (~2.0 sec), as shown in Table 1.4.

More volume was added above the heated section when performing Tests DB-8 and -9. While all other parameters remained constant, Test DB-8 was conducted at a lower pressure. The thermocouple measurements indicated that essentially the entire heated section was in subcooled boiling in Test DB-8. In DB-9, however, single-phase flow was present over the entire heated zone initially. During blowdown, the wall temperatures all show three characteristics: early and stable CHF in the lower region, early CHF with rewet in the middle region, and continuous cooling and delayed CHF in the upper region. Essentially the delayed CHF occurred when the test section was depleted of liquid.

TABLE 1.4. Summary of Time to CHF during Blowdown in Test Section I

Test No.	Time at Power Trip, sec	Thermocouple No.													
		1	2	4	6	9	12	15	18	21	24	27	30	32	35
Time to CHF, ^a sec															
DB-1	6.5	1.2	1.1	1.1	1.0	1.0	1.2	1.9	2.3	2.0	2.0	2.0	2.1	2.1	b
DB-2	5.3	0.7	0.7	0.7	0.7	0.7	0.7	0.7	1.4	1.4	1.4	1.4	1.5	b	1.7
DB-5	3.1	0.4	0.4	0.4	0.4	0.5	0.5	0.4	0.4	0.5	0.5	0.9	1.3	1.4	1.5
DB-6	2.9	0.4	0.4	0.4	0.4	0.5	0.5	0.4	0.4	0.4	0.4	0.9	1.2	1.4	1.5
DB-7	7.2	1.1	1.1* 2.3 ^c	1.1* 2.2 ^c	2.1	2.1	2.1	2.1	2.4	2.0	2.0	2.0	2.1	2.2	2.3
DB-8	3.6	0.6	0.5	0.5	0.5	0.6	0.6	0.6* 2.2 ^c	2.5	2.2	2.2	2.3	2.4	2.5	2.5
DB-9	4.4	0.8	0.5	0.5	0.5	0.5	0.6	0.4	0.7	0.6* 2.2 ^c	2.3	2.1	2.2	2.2	2.2

^aTime was rounded off to nearest 0.1 sec.^bSignal was not recorded.^cEarly CHF was rewet, indicated by *, followed by a second CHF.

A comparison between these two tests shows that the early and sustained temperature-excursion region was extended 15 cm more in DB-9, the high-pressure test. The rewetting characteristic of TC15 in DB-8 was similar to that of TC21 in DB-9; their locations were 15 cm apart. This preliminary result shows the effect of initial pressure on transient CHF behavior. However, the present test section was very short and the blowdown transient was quite rapid, making the resolution between early and delayed CHF somewhat difficult.

An adiabatic blowdown was performed with identical inlet conditions as DB-7, but with no heat addition. The results are shown in Figs. I.42-I.47. The initial undershoot in pressure was much less in this test, AB-5. Both the test-section pressure drop and the pressure differential across the break orifice were lower than those for the diabatic case, and the fluid temperature followed saturation for most of the transient. The wall temperature, however, showed sluggish response. This may have been due to the large amount of stored heat in the wall and the relatively low thermal conductivity of Freon-11. When the liquid in the system was exhausted at about 2.5 sec, the wall could not be cooled anymore and therefore remained relatively constant beyond that point.

Fig. I.42. System Pressure during Test AB-5

Fig. I.43. Differential Pressure across Test Section during Test AB-5

Fig. I.44. Differential Pressure across Break Orifice during Test AB-5

Fig. I.45. Turbine-flowmeter Measurement during Test AB-5

Fig. I.46. Fluid Temperature during Test AB-5

Fig. I.47. Wall-thermocouple Measurements during Test AB-5

Fig. I.47 (Contd.)

Fig. I.47 (Contd.)

4. Description of Test Section II

A 3.05-m (10-ft) test section with a symmetric heat-flux profile is being fabricated. A schematic of this test section is shown in Fig. I.48. The heat-flux profile shown in Fig. I.49 is obtained by varying the wall thickness of the tube, as shown in Fig. I.50. The wall thickness will subsequently be checked using the ultrasonic technique. Due to the electrical noise and pickup often observed in the wall-temperature measurement of Test Section I, a different type of thermocouple⁴ was installed on Test Section II. First a 36-gauge, Type K thermocouple was constructed with a small disk-shaped junction. (The disk diameter was about 1 mm and its thickness about 0.1 mm.) Then a thin piece of mica was used to electrically isolate the junction, as shown in Fig. I.48. A view of the junction and the mica is shown in Fig. I.51.

SCHEMATIC OF TEST SECTION II

Fig. I.49

Heat-flux Profile for Test Section II

Fig. I.50. Dimensions of Test Section II. ANL Neg. No. 900-76-700.

Fig. I.51

Photograph of Thermocouple Bead and Mica. ANI Neg. No. 900-76-999.

A bench test was performed comparing the response of the above-mentioned thermocouple with one having no mica insulation. These thermocouples, together with an ungrounded sheathed thermocouple similar to those used on Test Section I, were mounted side by side circumferentially on the wall of a 15-cm-long test-section tube. The sheathed thermocouple was spot-welded onto the wall as was done in Test Section I, whereas the other two were secured by wrapping a piece of high-temperature glass tape around the tube several times. A sudden wall-temperature excursion was achieved by directing a flame down the center of the tube. The result in Fig. I.52 indicates that the bare junction with mica insulation (TC2) responded much faster than the sheathed thermocouple junction (TC1). The rate of temperature rise in TC2 was $15^{\circ}\text{C}/\text{sec}$, 20% slower than that for the bare thermocouple without the mica insulation. Because of the improved response, the disk-shaped junction thermocouples were therefore used in Test Section II.

Fig. I.52. Thermocouple Responses in a Jet Flame Test

The rig will undergo some slight changes in order to accommodate a longer test section. An upper plenum, which will be installed immediately above the exit-turbine flowmeter, is also in fabrication. This added volume will lengthen the blowdown transient significantly and provide an extended time interval between the transition from bubbly flow within the test section and depletion of liquid within the system.

B. Nitrogen-Water Blowdown Tests (Y. S. Cha and R. E. Henry, RAS)

Three test sections, shown in Fig. I.53, were fabricated for the nitrogen-water blowdown tests. The test results for discharge nozzle A were reported previously, and the test results for discharge geometries B and C are described in the following paragraphs.

Fig. I.53. Dimensions (mm) of the Three Discharge Geometries and Locations of Pressure Taps

Typical results for the vessel pressure and the pressures downstream from the nozzle, with and without the tube bundle inside the blowdown vessel, are shown in Figs. I.54-I.61. (Only P_1 and P_4 are shown in these figures, since P_2 and P_3 exhibited similar behaviors.) The designations in these figures

Fig. I.54. Measured Pressures for Test Section B without a Tube Bundle and with Negligible Dissolved Gas. ANL Neg. No. 900-76-1007.

Fig. I.55. Measured Pressures for Test Section B without a Tube Bundle and with 24-hr Gas Bubbling Time. ANL Neg. No. 900-76-1008.

Fig. I.56. Measured Pressures for Test Section B with a Tube Bundle and Negligible Dissolved Gas. ANL Neg. No. 900-76-1011.

Fig. I.57. Measured Pressures for Test Section B with a Tube Bundle and 24-hr Gas Bubbling Time. ANL Neg. No. 900-76-1010.

Fig. I.58. Measured Pressures for Test Section C without a Tube Bundle and with Negligible Dissolved Gas. ANL Neg. No. 900-76-1014.

Fig. I.59. Measured Pressures for Test Section C without a Tube Bundle and with 24-hr Gas Bubbling Time. ANL Neg. No. 900-76-1009.

Fig. I.60. Measured Pressures for Test Section C with a Tube Bundle and Negligible Dissolved Gas. ANL Neg. No. 900-76-1013.

Fig. I.61. Measured Pressures for Test Section C with a Tube Bundle and 24-hr Gas Bubbling Time. ANL Neg. No. 900-76-1012.

indicate the conditions under which the particular test was conducted. For example, R-B-TB-4.24-65-24 represents a test run with discharge geometry B, a tube bundle inside the vessel, initial vessel pressure of 4.24 MPa, water temperature of 65°C, and a system bubbling time of 24 hr. A designation such as R-C-NO-4.24-65-0 represents a test run with discharge geometry C, no tube bundle inside the vessel, initial vessel pressure of 4.24 MPa, water temperature of 65°C, and a zero system bubbling time. As shown in these figures, tests with discharge geometries B and C exhibited similar behaviors.

For tests with zero bubbling time, both the vessel pressure and the pressures downstream from the nozzle demonstrated a smoothly varying behavior (except at the end of the test, where the liquid above the nozzle was exhausted). For tests with a 24-hr bubbling time and without the tube bundle inside the vessel, nonperiodic, intermittent pressure oscillations (at relatively large time intervals) with relatively small amplitudes were measured in the constant-area portion of the test section. For tests with a 24-hr gas bubbling time and with the tube bundle inside the vessel, large-amplitude, nearly continuous oscillations occurred. Apparently, the available nucleation sites in the system for tests without the tube bundle were not sufficiently numerous to produce the highly compressible mixture responsible for the near-continuous oscillations observed in the tests with the tube bundle. This is similar to the test results obtained with discharge nozzle A and reported previously.

Tables I.5-I.8 show the measured blowdown time (defined as the time required to exhaust the liquid above the discharge nozzle) for these tests. Increased nucleation sites within the vessel enable gas to come out of solution on a time scale comparable to the blowdown time, i.e., many seconds. In this case, sufficient gaseous quantities can be produced to obtain a compressible mixture, which results in a reduced flow rate and increased blowdown time. However, when these vessel nucleation sites are absent, as in an ECCS accumulator, the noncondensable gas has a very small effect on the system flow rate.

TABLE I.5. Blowdown Time for Discharge Nozzle B
without Tube Bundle inside the Vessel

Run	t_B , sec	Δt_B , sec	% Difference
R-B-NO-4.24-65-24	26.9	0	
R-B-NO-4.24-65-0	24.2	2.7	11.1
R-B-NO-2.86-65-24	32.9		
R-B-NO-2.86-65-0	29.0	3.9	13.4
R-B-NO-1.48-65-24	43.6		
R-B-NO-1.48-65-0	42.0	1.6	3.81

TABLE I.6. Blowdown Time for Discharge Nozzle B
with Tube Bundle inside the Vessel

Run	t_B , sec	Δt_B , sec	% Difference
R-B-TB-4.24-65-24	27.0		
R-B-TB-4.24-65-0	21.9	5.1	23.3
R-B-TB-2.86-65-24	31.9		
R-B-TB-2.86-65-0	26.9	5.0	18.6
R-B-TB-1.48-65-24	44.5		
R-B-TB-1.48-65-0	40.3	4.2	10.4

TABLE I.7. Blowdown Time for Discharge Nozzle C
without Tube Bundle inside the Vessel

Run	t_B , sec	Δt_B , sec	% Difference
R-C-NO-4.24-65-24	13.9		
R-C-NO-4.24-65-0	12.3	1.6	13.0
R-C-NO-2.86-65-24	16.2		
R-C-NO-2.86-65-0	14.8	1.4	9.45
R-C-NO-1.48-65-24	21.9		
R-C-NO-1.48-65-0	21.0	0.9	4.28

TABLE I.8. Blowdown Time for Discharge Nozzle C
with Tube Bundle inside the Vessel

Run	t_B , sec	Δt_B , sec	% Difference
R-C-TB-4.24-65-24	13.7		
R-C-TB-4.24-65-0	10.3	3.4	33.0
R-C-TB-2.86-65-24	16.2		
R-C-TB-2.86-65-0	13.2	3.0	22.7
R-C-TB-1.48-65-24	21.0		
R-C-TB-1.48-65-0	18.1	2.9	16.0

References

1. J. C. Leung, R. E. Henry, and O. C. Jones, "Transient Critical Heat Flux, Light-water-reactor Safety Research Program: Quarterly Progress Report, April-June 1975, ANL-75-58, p. 30 (Sept 26, 1975)."
2. M. N. Hutcherson, "Contribution to the Theory of the Two-phase Blowdown Phenomenon, ANL-75-82 (Dec 1975)."
3. "Cox Flow Convectometer Installation, Operation, and Service Instructions Manual, Technical Bulletin CH-3606."
4. B. W. Sheron, "The Dryout and Rewetting of a Heated Surface During Flow Transients, Ph.D. thesis in nuclear engineering, Catholic University of America (Apr 1975)."

II. TRANSIENT FUEL RESPONSE AND FISSION-PRODUCT RELEASE PROGRAM

Responsible Group Leaders:

L. A. Neimark and R. B. Poeppel, MSD

Coordinated by:

L. R. Kelman, MSD

A physically realistic description of fuel swelling and fission-product release is needed to aid in the prediction of the behavior of fuel rods and fission products under certain hypothetical Light-Water Reactor (LWR) accident conditions. To satisfy this need, a comprehensive computer-base model, the Steady State and Transient (SST) fuel-element behavior code, is being developed at Argonne National Laboratory (ANL). This model is being incorporated into the Fuel-Rod Analysis Program (FRAP) code being developed by the Idaho National Engineering Laboratory (INEL). The analytical effort is supported by data developed from characterization of irradiated LWR fuel and from out-of-reactor transient heating tests on irradiated commercial LWR fuel under a range of thermal conditions. In the near term, emphasis in the experimental program is being placed on fuel thermal conditions that are applicable to anticipated hypothetical power-coolant-mismatch (PCM) accidents. Later experimental efforts will turn to conditions typical of other types of hypothetical accidents.

The technical status of the program was reviewed at ANL on August 11, 1976, by the Water Reactor Safety Research (WRSR) Fission Product Review Group.

J. Rest attended the Specialist Meeting on the Behavior of Water Reactor Fuel Elements under Accident Conditions, sponsored by the Committee on the Safety of Nuclear Installations (CSNI) of the Organization for Economic Cooperation and Development (OECD) and held in Norway, September 13-16, 1976. He presented the paper "Development and Experimental Verification of SST: A Steady-state and Transient Fuel Response and Fission-product Release Code," by J. Rest, M. G. Seitz, S. M. Gehl, and L. R. Kelman. He also visited the Berkeley Nuclear Laboratories in England and the European Institute for Transuranium Elements in Karlsruhe, Germany, for discussions of fission-gas and fission-product behavior.

Personnel from ANL participated in the Fuel Code Verification Workshop of the Fourth Water Reactor Safety Research Information Meeting at the National Bureau of Standards, Gaithersburg, Md., on September 28, 1976.

A request was made by NRC-RSR¹ for INEL and ANL to jointly propose a course of action for cross-checking the behavior of LWR fuel and the release of fission gas observed in the INEL in-reactor transient tests with that observed in the ANL out-of-reactor transient tests. The high fission-gas

release and extensive intergranular separation observed by ANL in commercial LWR fuel that has been transient-tested by direct-electrical heating (DEH), ANL-76-87, p. 16, is recognized as having important potential impact on the development of models for the transient behavior of LWR fuel rods and on the planning of experiments in the Power Burst Facility (PBF). The first transient tests by nuclear heating of experimental fuel in the PBF at INEL have shown apparent low fission-gas release and little intergranular separation. A cross-check program is envisaged that will involve "pre- and posttest examination of replicate sets of essentially identical fuel specimens that would be subjected to approximately the same thermal transient in the PBF and in ANL's DEH apparatus." Ground rules, feasibility, timing, and logistics of the proposed cross-check program were discussed at a meeting of representatives of NRC, INEL, and ANL at the National Bureau of Standards, Gaithersburg, Md., on September 28, 1976.

Recent significant analytical and experimental advances and the status of the program at the end of this quarter are summarized as follows:

1. GRASS sensitivity analyses, coupled with observations of DEH-tested fuel, indicate that a fuel-fracturing dependence on heating rate may be a key factor that influences release of fission gases under transient conditions.
2. GRASS(SST)-Mod 3 has been developed and will be transmitted to INEL for incorporation into FRAP. It differs from GRASS(SS)-Mod 2 in that it includes models for (a) the diffusion of fission gas from the lattice to grain boundaries and along grain edges, applicable under steady-state and transient conditions, (b) the instantaneous rate of growth of coalescing bubbles, and (c) the burst release of fission gas.
3. An LWR fuel-behavior code (LIFE-LWR) was generated based on SST and LIFE-III and includes thermal and mechanical properties and models.
4. The quantities of fission gases determined in the high-burnup region of the as-received Robinson fuel are $0.685 \pm 0.006 \text{ cm}^3 \text{ (STP) Xe/g fuel}$ and $0.0533 \pm 0.009 \text{ cm}^3 \text{ (STP) Kr/g fuel}$. The concentrations of some other fission products have also been determined.
5. The radial profile of fission gas in as-received Robinson fuel has been determined by a laser-sampling technique.
6. PCM-like transient tests of Robinson fuel have been run below the melting temperature of the fuel, and the released fission gases have been analyzed. The fuel is extensively cracked intergranularly, similar to fuel heated to central melting.
7. Evidence exists that volatile fission products accompany fission gases to grain boundaries during transient heating.

8. The DEH Transient Temperature Distribution (DEHTTD) code has been modified and used to calculate the radial temperature distribution in DEH fuel specimens during cooldown as well as heatup.

A. Modeling of Fuel-Fission-product Behavior (J. Rest, MSD)

1. Steady-state LWR Fuel-rod Performance Code Development
(J. Rest, M. C. Billone, and V. Z. Jankus, MSD)

Personnel at ANL were invited to contribute at the Fuel Code Verification Workshop to a presentation made by INEL that compared the performance predictions of GAPCON-THERMAN-1, LIFE-LWR, and FRAP-S for instrumented Halden fuel rods and one unpressurized fuel rod from a pressurized-water reactor (PWR). The designated responsibility of ANL was to run the comparison cases with SST. We received the necessary input data and limited experimental data from INEL on August 30, 1976. Because SST is based on an outdated version of the LIFE LMFBR fuel-element performance code and does not include any LWR-based modifications of the mechanical analysis, a new LWR fuel-rod behavior code (LIFE-LWR) was generated on the basis of the latest version of LIFE (LIFE-III), which includes mechanical as well as thermal LWR properties and models.

a. LIFE-LWR

In developing LIFE-LWR, we revised a number of LIFE models and added some models to reflect the operating conditions and phenomena particular to LWR fuel elements. These include:

(1) A thermal-hydraulics model to calculate the coolant-to-cladding heat-transfer coefficient for both boiling-water and pressurized-water coolants (based on SST code analysis).

(2) A radial flux-depression model that can be used for enriched PWR ^{235}U , BWR ^{235}U , PWR ^{239}Pu , and BWR ^{239}Pu fuel (based on SST code analysis).

(3) A provision for decreasing local fission rate and enrichment as a function of burnup or for changing these values on the history cards (based on SST code analysis).

(4) A model for the anisotropic (axial) growth of Zircaloy cladding as a function of fast fluence.

(5) A fuel densification model based on sintering.

The basis for LIFE-LWR is the LIFE-III² (specific master version is LIFE-3A) code, which has been developed, calibrated, and verified by the ERDA/RDD-sponsored LIFE Working Group, of which ANL is a member. LIFE-III was finalized in June 1976. It will be submitted to the Argonne Code Center in November 1976 upon completion of documentation.

Certain modifications were made to LIFE-III as an intermediate step in converting it to an LWR code. These include:

- (1) Removal of the anisotropic fuel swelling model, because the model is not applicable to LWR operating conditions and it requires excessive core storage.
- (2) An increase in the maximum number of axial sections from six to 10 to permit an accurate representation of axial power and flux gradients.
- (3) A bypass of the cladding wastage model because it is based on correlations for sodium corrosion and mixed-oxide fission-product attack of stainless steel.
- (4) Improvement of the solution procedure in the gap-conductance calculations.³

The Electric Power Research Institute (EPRI) is sponsoring an LWR Fuel Rod Modeling Code Evaluation Program (RD 397) in which a number of performance codes--including LIFE--are being examined. To minimize code differences due to variations in materials properties input, a standard set of properties for UO_2 fuel and Zircaloy cladding was adopted by EPRI.⁴ The EPRI-recommended properties incorporated into LIFE-LWR are (1) surface emissivity (UO_2 and Zircaloy), (2) thermal conductivity (UO_2 and Zircaloy), (3) thermal-expansion coefficient (UO_2), shear modulus and Poisson's ratio (UO_2 and Zircaloy), (5) steady-state creep (UO_2), (6) transient and steady-state creep (Zircaloy, assumed isotropic), (7) fracture stress (UO_2), and (8) anisotropic growth (Zircaloy).

In cases for which the EPRI properties list appeared incorrect or incomplete, data from MATPRO⁵ for densification due to sintering (UO_2) and coefficient of thermal expansion (Zircaloy, assumed isotropic) were used in LIFE-LWR. Major LIFE-III fuel models that are used in LIFE-LWR are (1) fuel swelling, (2) fuel cracking, (3) fission-gas retention and release, (4) gap conductance (based on the Ross-Stoute approach), and (5) porosity migration and grain growth (based on WARD PORMIG). Most of these models have undetermined physical constants that can be adjusted on the basis of LWR thermal- and mechanical-performance data.

b. Model Additions and Revisions

(1) Thermal Hydraulics. The thermal-hydraulic calculations in LIFE are performed in Subroutine GOLDN. The LIFE-LWR thermal-hydraulic model is obtained from the SST code,^{6,7} developed at ANL. It is applicable to both boiling-water and pressurized-water cooling systems. The water properties included in the code are thermal conductivity, k_w , specific heat, C_p , and viscosity, μ_w , which are approximated by polynomials of temperature (T_F , in $^{\circ}\text{F}$) over the temperature range 400-600 $^{\circ}\text{F}$.

$$k_w = 0.32521 + 5.287 \times 10^{-4} T_F - 9.6872 \times 10^{-7} T_F^2, \text{ Btu/h} \cdot \text{ft} \cdot ^\circ\text{F}; \quad (1)$$

$$C_p = 1.2895 - 2.1566 \times 10^{-3} T_F + 4.0543 \times 10^{-6} T_F^2, \text{ Btu/lb}_m \cdot ^\circ\text{F}; \quad (2)$$

$$\mu_w = 3.5021 \times 10^{-4} - 9.6859 \times 10^{-7} T_F + 8.0876 \times 10^{-10} T_F^2, \text{ lb}_m/\text{ft}^3. \quad (3)$$

The model for the cladding-to-coolant heat-transfer coefficient, H_c , is based on the Dittus-Boelter equation

$$H_c = \text{Nu} \cdot k_w \cdot \text{Pr}/D_e, \text{ Btu/h} \cdot \text{ft}^2 \cdot ^\circ\text{F}, \quad (4)$$

where

$$\text{Nu} = \text{Nusselt number} = 0.23 D_e V / (12 \mu_w)^{0.8}, \quad (5)$$

$$V = \text{water mass flow rate in lb}_m/\text{s} \cdot \text{ft}^2.$$

$$D_e = \text{equivalent diameter} = 4A_f/P, \text{ ft}, \quad (6)$$

$$A_f = \text{flow area associated with one rod, ft}^2,$$

$$P = \text{wetted perimeter, ft},$$

and

$$\text{Pr} = \text{Prandtl number} = (3600 C_p \mu_w / k_w)^{0.4}. \quad (7)$$

(2) Radial Flux Depression and Time Variation of Axial Fission-rate Profiles. The SST Subroutine FLXDEP was adapted to LIFE-LWR to calculate the radial flux depression. The equation used for this calculation is

$$q(r) = \left[\Gamma_1 (r/r_f)^{P_2} + P_3 \right] / r_f^2, \quad (8)$$

where $q(r)$ are nonnormalized shape factors used to determine the radial power density, r is the radial position, r_f is the fuel outer radius, and P_1 , P_2 , and P_3 are plant factors that are a function of enrichment and time. The input parameter, IPLANT, determines the appropriate P_1 , P_2 , and P_3 for PWR ^{235}U -enriched fuel (IPLANT = 1), BWR ^{235}U -enriched fuel (IPLANT = 2), PWR plutonium-enriched fuel (IPLANT = 3), or BWR plutonium-enriched fuel (IPLANT = 4).

As in SST, the initial axial fission-rate profile and enrichment in LIFE-LWR are established by input parameters. At any time

during the reactor operation, this shape can be changed by specifying the new shape factors on the appropriate history cards. An option also exists to decrease the local fission rate and enrichment as a function of burnup, thereby changing these values on a continual basis.

(3) Anisotropic Irradiation Growth of Zircaloy. LIFE-III contains an isotropic model (Subroutine HKNS) for the volumetric swelling of Type 316 stainless steel cladding. It has been observed that Zircaloy undergoes anisotropic growth described by the equations⁴

$$\epsilon_z^s = 10^{-3}(\phi t/10^{21})^{0.7} \quad (9)$$

and

$$\epsilon_r^s = \epsilon_\theta^s = -0.5\epsilon_z^s, \quad (10)$$

where ϕt is the fast fluence and the ϵ^s values are the strain components. Equations 9 and 10 have been programmed into LIFE-LWR Subroutine SOLVER, and the isotropic swelling calculated by HKNS has been set to zero.

(4) Fuel Densification. LIFE-III contains models for fuel densification due to hot pressing of as-fabricated pores and for pore migration that results from vapor transport. In generating LIFE-LWR, a sintering model based on MATPRO⁵ was included. According to the model, densification depends only on burnup and proceeds along the universal curve

$$\frac{\Delta L}{100L} = -3 + 0.93 \exp(-Bu) + 2.07 \exp(-35Bu), \quad (11)$$

where Bu is burnup in MWd/kg UO₂, L is the length of a fuel pellet, and ΔL is the change in length due to densification. The starting point depends on the sintering temperature T_s , which governs the maximum shrinkage

$$\left(-\frac{\Delta L}{100L}\right)_{\max} = 22.2 \frac{100 - D}{T_s - 1180}, \quad (12)$$

where D is initial fuel density in percent theoretical density (TD) and T_s is in °C. Sintering can be minimized by using a large value (e.g., 3000) for T_s . In LIFE-LWR Subroutine BEGIN, the initial Bu that corresponds to Eq. 12 is calculated. Then Eq. 11 is used to calculate the densification in SOLVER. This is added to the volume changes in SWELL.

c. Materials Properties for Zircaloy Cladding

The equations used in LIFE-LWR for fuel and cladding are presented below.

(1) Surface Emissivity⁴

$$\epsilon_{sc} = 0.751. \quad (13)$$

(2) Thermal Conductivity⁴

$$k_c = 8.08 + 3.17 \times 10^{-3} T_F, \text{ Btu/h} \cdot \text{ft} \cdot ^\circ\text{F}. \quad (14)$$

(3) Instantaneous Coefficient of Thermal Expansion (Assumed Isotropic)^{4,5}

$$\alpha_c = 1.096 \times 10^{-6} + 5.676 \times 10^{-9} T_F, ^\circ\text{F}^{-1}. \quad (15)$$

Equation 15 is based on data^{4,5} for diametral thermal expansion.

(4) Shear Modulus⁴

$$G_c = 5.376 \times 10^6 - 3.34 \times 10^{-3} T_F, \text{ psi}. \quad (16)$$

(5) Poisson's Ratio⁴

$$\nu_c = 0.298 - 7.025 \times 10^{-5} T_F. \quad (17)$$

(6) Creep Rate (Isotropic).⁴ The creep equation for cladding is used in Subroutine CREEC. All the creep constants are contained in that subroutine. The creep rate is

$$\begin{aligned} \dot{\epsilon}_c = & 9.07 \times 10^{-22} [1 + 0.111 \exp(-1.11 \times 10^{-6} \phi t / \phi)] [\exp(-8000 / RT_K)] \phi \sigma_e \\ & + [4.67 \times 10^{28} \exp(-138,000 / RT_K)] (\sigma_e / 1000)^n, \text{ h}^{-1}, \end{aligned} \quad (18)$$

where

$$n = 25.6 - 0.026 T_K, \quad (19)$$

ϕ is flux in $\text{n/cm}^2 \cdot \text{s}$, R is the universal gas constant, T_K is temperature in $^\circ\text{K}$, and σ_e is the equivalent stress in psi. The LIFE-III Subroutine CREEC was streamlined to accommodate this equation for Zircaloy creep.

d. UO₂ Fuel Properties(1) Surface Emissivity⁴

$$\epsilon_{sf} = 0.85. \quad (20)$$

(2) Thermal Conductivity⁴

$$k_f = \frac{41.26(1 - P)}{1 + 0.5P} \left(\frac{1}{129.4 + T_K} + 1.60173 \times 10^{-14} T_K^3 \right), \text{ W/cm} \cdot ^\circ\text{C}, \quad (21)$$

where P is fractional porosity.

(3) Instantaneous Coefficient of Linear Thermal Expansion⁴

$$\alpha_f = 6.8 \times 10^{-6} + 5.8 \times 10^{-9} T_C, ^\circ\text{C}^{-1}, \quad (22)$$

where T_C is temperature in $^\circ\text{C}$.

(4) Young's Modulus⁴

$$E_f = 25.5 \times 10^6 (1 - 2.28P)(1 - 5.532 \times 10^{-4} T_C), \text{ psi}, \quad (23)$$

where $25^\circ\text{C} < T_C < 1700^\circ\text{C}$.

(5) Poisson's Ratio⁴

$$\nu_f = 0.32[1 - 5.6 \times 10^{-5}(T_C - 25)]. \quad (24)$$

(6) Fuel Fracture Stress⁴ (Subroutine CRACK)

$$\sigma_f = 15,686 + 3.17 T_K, \text{ psi}. \quad (25)$$

(7) Fuel Creep⁴ The steady-state creep rate for UO_2 is

$$\begin{aligned} \dot{\epsilon}_c = & (9.73 \times 10^6 / G_d^2)(D - 87.7)^{-1} \exp(-90,000 / RT_K) \sigma_e \\ & + [1.38 \times 10^{-4} (D - 90.5)^{-1} \exp(-132,000 / RT_K)] \sigma_e^{4.5} \\ & + [7.5 \times 10^{-9} \exp(-90,000 / RT_K)] \dot{F} \sigma_e + 3.72 \times 10^{-23} \dot{F} \sigma_e, \text{ h}^{-1}, \end{aligned} \quad (26)$$

where $\dot{F}$ is the fission rate in fissions/($\text{cm}^3 \cdot \text{s}$), G_d is the grain diameter in μm , and the density D is in the range $92 \leq D \leq 97\% \text{ TD}$.

e. Uncertain Parameters

Some of the physical and semiempirical models in LIFE-LWR contain parameters for which no data exist, or the data have too much scatter to permit precise determination of numerical values. The general practice is to assign values to these parameters to achieve the best fit to low-burnup temperature and microstructure data and higher-burnup gas-release and cladding-strain data from irradiated rods. The major models that contain uncertain parameters and the intermediate values chosen for these parameters are described below.

(1) Gap Conductance. Uncertain parameters in the gap-conductance model include the helium accommodation coefficient (the factor used to describe the change in effective gap width due to the presence of solid fission products in the gap) and the coefficient in the solid-solid conductance term. The values used are 0.21, 500, and 0.26, respectively.

(2) Porosity Migration and Grain-growth Constants. The values of various parameters used in LIFE-LWR in the equations that describe the rate of porosity migration, which is due to vapor transport and surface-diffusion mechanisms, and the rate of grain growth are the same as those obtained for LIFE-III LMFBR irradiation conditions.

(3) Fission-gas Release. The LIFE-LWR equation for the rate of fractional gas release is

$$\Gamma = \exp(-0.13 - 21.450/T_R), h^{-1}, \quad (27)$$

where T_R is local temperature in °R. These values will probably be modified when a detailed verification of LIFE-LWR is performed.

(4) Fuel Creep. Because of variables that are not included in the model (Eq. 26), such as oxygen-to-metal ratio and impurity content, and scatter in the fuel-creep data, uncertainty exists in the fuel-creep-rate equation. Initially, the parameters in the creep equation are set to the present state-of-the-art values. As the data become more well-defined, they will be incorporated into the model.

(5) Fuel Swelling and Densification. The fuel-swelling model has three uncertain parameters. The pressure due to fission-gas-bubble surface tension is calculated from

$$p_g = 1.28 \exp(14,400/RT_K), \text{ psi}. \quad (28)$$

The rate at which a gas bubble swells to equilibrium, $\dot{\epsilon}_s$, as a result of the driving force of the pressure difference across the bubble surface is given by

$$\dot{\epsilon}_s = 2.25 \cdot 100 \epsilon_s \dot{\epsilon}_{ssc} (p_a - p_f - p_g), \quad (29)$$

where

$$\begin{aligned} \dot{\epsilon}_{ssc} = & (1.05 \times 10^6 / G_d^2) \exp(-90,000/RT_K) + [7.5 \times 10^{-9} \exp(-90,000/RT_K) \\ & + 3.72 \times 10^{-23}] \Gamma, \end{aligned} \quad (30)$$

p_a is the internal bubble pressure calculated from the van der Waals gas law, and p_f is the hydrostatic pressure in the fuel matrix.

f. Description of Standard Fuel-rod Cases

Personnel from INEL recommended that ANL only run Halden rods IFA-226 AA and AK and the EPRI Case D (unpressurized PWR fuel rod).

(1) Halden IFA-226 Rod AA. Rod AA initially contained 92.1% TD fuel, an 8.1-mil helium-filled diametral gap, and 23-mil Zircaloy-4 cladding. It was positioned in the Halden reactor so that the bottom of the fuel column operated at peak power. For code comparison, fuel centerline temperature versus local power was calculated near the top of the fuel column for a decreasing power ramp near the beginning of life with 11-kW/ft peak power and an increasing power ramp with 13-kW/ft peak power, which occurred at 1200 MWd/MT.

(2) Halden IFA-226 Rod AK. Rod AK initially contained 96% TD fuel, an 8.1-mil helium-filled diametral gap, and 23-mil Zircaloy-4 cladding. It was positioned in the Halden reactor so that the top of the fuel column operated at peak power. For Rod AK, the cladding axial strain, $\Delta\epsilon/\epsilon$, versus peak power was calculated for a decreasing power ramp near the beginning of life with 15-kW/ft peak power. For this case, the strains were mainly due to the cladding thermal expansion.

(3) EPRI Case D. Case D was used by EPRI in Phase II of their LWR Fuel Rod Modeling Code Evaluation. This case consists of an unpressurized PWR fuel rod for which cold diametral gap, internal pressure, fission-gas release, and cladding strain data are available after 13,200 MWd/MT (~10,000 full-power hours). The Case D rod initially consisted of 95% TD fuel, an 8.5-mil diametral gap that contained 88% helium and 12% nitrogen, 26-mil Zircaloy cladding, and 1800-psi coolant pressure. The peak power occurred at about two-thirds of the axial length up from the bottom of the fuel rod, where the code comparisons are to be made. These comparisons are for fuel centerline temperature, gap conductance, gap size, fission-gas release, and plenum pressure as a function of time or burnup and end-of-life (EOL) (~13,200 MWd/MT) values for cladding strain, cold diametral gap, fission-gas release, and plenum pressure.

g. Results

(1) Halden IFA-226 Rod AA. Table II.1 shows calculated Rod AA fuel centerline temperatures (T_c) versus peak power for the two ramps. LIFE-LWR tends to overpredict fuel centerline temperatures by as much as 400°F on the first ramp. Past experience with SST and LIFE-LWR indicates that this is a general trend when calculations are compared with beginning-of-life Halden data. However, the agreement between calculations and data is quite good for the second ramp. Calculated temperatures are within 100°F of measured temperatures.

TABLE II.1. LIFE-LWR Calculated Fuel Centerline Temperature vs Peak Power for Halden IFA-226 Rod AA

Ramp	Peak Linear Power, kW/ft	T _g , °F	
		Calculated	Measured ^a
Near Beginning of Life	11.0	3080	-
	10.0	2760	-
	8.0	2250	1850
	6.0	1760	1450
	4.0	1290	1200
	2.0	860	800
1200 MWd/MT	3.0	1020	1020
	4.9	1460	1400
	6.9	1840	1820
	8.8	2200	2100
	10.8	2550	2650
	12.8	3010	-

^aData supplied to ANL in graphical form by INEL.

(2) Halden IFA-226 Rod AK. The change in cladding length for Halden IFA-226 Rod AK is listed in Table II.2 as a function of peak linear power for the first ramp. The length of the cladding at zero power is used as the reference. On the average, the

TABLE II.2. LIFE-LWR Calculated Change in Cladding Length (Δl) for Halden IFA-226 Rod AK as a Function of Peak Linear Power for the First Ramp^a

Peak Power, kW/ft	Δl , mm	
	Calculated	Measured ^b
15.0	0.66	1.0
12.0	0.51	0.56
10.0	0.41	0.32
8.0	0.33	0.16
6.0	0.23	0.07

^aReference cladding length is 785.62 mm at zero power.

^bGraphical data available in Ref. 8.

LIFE-LWR results agree with the measurements. However, discrepancies exist at low and high power. Three factors should be considered when evaluating the comparison in Table II.2: (1) The cladding strains are quite small, ~0.1%, (2) no error bars are available for the measurements, and (3) the thermal-expansion coefficient programmed into LIFE-LWR is for diametral expansion. For simplicity, thermal expansion was treated as isotropic and was therefore used in the axial elongation

calculations. Because Zircaloy is anisotropic, the thermal-expansion coefficient is different in the axial than in the diametral direction. Some of the differences between calculated and measured values could be due to this anisotropy.

(3) EPRI Case D. Case D was run twice. The first time the problem was set up assuming a 7000-h full-power history (Tables II.3 and II.4). However, the Case D rod actually was irradiated for ~10,000 h at full power. Subsequently, Case D was rerun for the correct power history (Tables II.5 and II.6). The results for the correct history differ somewhat from those presented at the Water Reactor Safety Research Information Meeting⁹ for cladding creep down, plenum pressure, and maximum fuel temperature. However, the trends in the results are similar.

The variations of centerline temperature, gap conductance, radial gap width, fission-gas release, and plenum pressure with power level and burnup are reasonable (Table II.5). Note that LIFE-LWR predicts that the fuel-cladding gap narrows as a function of burnup. This is partly due to cladding creep down and fuel swelling. Also, the code predicts that the maximum fuel temperature occurs at 7.5 kW/ft (8700 MWd/MT) instead of the peak power of 7.97 kW/ft (10,800 MWd/MT). The reason for this is the calculated increase in gap conductance with burnup.

TABLE II.3. LIFE-LWR Results for Case D Assuming a 7000-h History

Time, h	Peak Power, kW/ft	T_c , °F	Gap Conductance, Btu/h·°F·ft ²	Radial Gap Width, mil	Fission-gas Release, %	Plenum Pressure, psi
2	6.34	1880	542	2.95	0.0	25.6
1845	6.34	1860	551	2.79	0.9	26.6
2655	5.28	1630	533	2.76	1.0	27.4
3248	2.50	1040	448	3.14	0.9	26.1
4581	6.91	2030	583	1.79	2.5	35.0
4846	7.50	2160	612	1.64	3.0	38.0
5467	6.91	2000	599	1.25	3.7	41.5
5731	7.49	2110	657	1.00	4.1	45.1
5814	7.97	2210	700	0.87	4.3	46.9
7000	6.62	1800	765	0.59	4.7	51.9

TABLE II.4. Comparison between LIFE-LWR Results and Experimental Data for Case D Assuming a 7000-h History

Parameter	Value	
	LIFE-LWR Calculated	Measured
Maximum T_c , °F	2210	2970 ^a
EOL Fission-gas Release, %	4.7	12.7
EOL Cold Plenum Pressure, psi	23.5	64.4
EOL Cold Diametral Gap, mil	2.9	4.2
EOL Cladding Creep, %	-0.54	-0.59

^aInferred from microstructure.

TABLE II.5. LIFE-LWR Results for Case D Assuming the Correct 10,555-h History

Time, h	Burnup, MWd/MT	Peak Power, kW/Et	T_c , °F	Gap Conductance, Btu/h·°F·ft ²	Radial Gap Width, mil	Fission-gas Release, %	Plenum Pressure, psi
2	0	6.34	1880	542	2.95	0.0	25.6
2,650	3,550	6.34	1860	546	2.53	1.4	28.1
3,800	4,850	5.28	1630	536	2.36	1.5	29.7
4,600	5,300	2.50	1040	439	2.77	1.4	28.2
6,600	8,100	6.91	1950	661	0.97	3.7	44.7
7,000	8,700	7.50	2020	773	0.66	4.3	49.8
7,800	10,000	6.91	1810	902	0.42	4.6	54.5
8,220	10,600	7.49	1850	1190	0.22	4.8	58.9
8,340	10,800	7.97	1910	1410	0.13	4.9	60.9
10,055	13,200	6.62	1560	1760	0.14	4.7	65.0

TABLE II.6. Comparison between LIFE-LWR Results and Experimental Data for Case D Assuming the Correct 10,055-h History

Parameter	Value	
	LIFE-LWR Calculated	Measured
Maximum T_c , °F	2020	2970 ^a
EOL Fission-gas Release, %	4.7	12.7
EOL Cold Plenum Pressure, psi	29.9	64.4
EOL Cold Diametral Gap, mil	1.5	4.2
EOL Cladding Creep, %	-0.73	-0.59

^aInferred from microstructure.

Table II.6 compares LIFE-LWR calculations and experimental data for Case D. The code underpredicts the maximum temperature, cold gap size, fission-gas release, and plenum pressure. These results are consistent with the predictions of FRAP-S and GAPCON-THERMAL. Physically, an underprediction of fuel densification results in a smaller gap and lower temperatures. Unfortunately, not enough time was available for a rerun of the problem with higher densification rates and lower fuel-swelling rates to determine whether these mechanisms are responsible for the high estimated centerline temperature. Note that the temperature and gas-release results for Case D are higher than results for comparable rods irradiated in the Halden reactor.

(4) Conclusions. Some of the conclusions that result from the code comparison⁹ are:

(a) Significant modeling differences exist between FRAP-S1, GAPCON-THERMAL, and LIFE-LWR.

(b) Good agreement is shown between the code predictions and the IFA-226 thermal data.

(c) For the EPRI standard problem, relatively good agreement exists between the predictions from GAPCON-THERMAL (without densification), FRAP-S1, and LIFE-LWR.

(d) GAPCON-THERMAL (without densification), FRAP-S1, and LIFE-LWR underpredicted peak fuel centerline temperature and cold internal pressure for the high-fission-gas-release fuel rod, but qualitatively the predictions are in good agreement with the limited data available for the low-fission-gas-release fuel rods from the EPRI standard problem.

(e) GAPCON-THERMAL, with densification, overpredicts the fuel temperature, internal pressure, and percent fission-gas release for the EPRI standard problem.

2. Transient Fission-gas Release (J. Rest, MSD)

a. Rate of Growth of Coalescing Bubbles

The steady-state GRASS code assumes that, when two fission-gas bubbles coalesce, the noninstantaneous rate of growth of the resultant bubble to equilibrium size can be treated as instantaneous. This approximation is reasonable because of the greater length of the code time steps for the thermal and mechanical analysis compared with the time required for coalescing bubbles to reach equilibrium size. However, in a transient analysis this assumption breaks down and the noninstantaneous rate of growth of coalescing bubbles must be included. The need for a GRASS model that describes the limited rate of growth of coalescing bubbles became apparent when GRASS-calculated bubble-size distributions for DEH test conditions were compared with qualitative experimental observations. The code predicted similar end-of-test bubble distributions in the lattice and on the grain boundaries. However, no bubbles were observed intragranularly, whereas relatively large bubbles were quite numerous on the grain faces.

In general, when two bubbles initiate coalescence, a strain field is generated in the lattice around the growing bubble. Initially, bubble coalescence is a volume-conserving process; vacancies moving under the influence of the strain field migrate to the bubble, which results in an increase in the bubble volume. As vacancies enter the growing bubble, the strain field gradually relaxes until it vanishes (to first order) when the bubble has reached its equilibrium size. A lattice-bubble relaxation time, τ_B^l , can be defined such that, at a time $t = \tau_B^l$ after the initiation of bubble coalescence, the resultant bubble has approached a stable configuration by an amount $1/e$. We can estimate τ_B^l from a consideration of lattice-vacancy thermodynamics:¹⁰

$$\tau_B^l = R_{ij}^2 / D_V C_V^e, \quad (31)$$

where R_{ij} is the sum of the radii of the coalescing bubbles, D_V is the vacancy-diffusion coefficient, and C_V^e is the fractional vacancy-equilibrium concentration. The terms D_V and C_V^e are both functions of temperatures; i.e.,

$$D_V = D_V^0 \exp(-E_V^m / kT_K) \quad (32)$$

and

$$C_V^e = \exp(-E_V^f / kT_K), \quad (33)$$

where E_V^m is the vacancy-migration energy, E_V^f is the vacancy-formation energy, D_V^0 is a preexponential constant, and k is Boltzmann's constant. For example, using $R_{ij} = 30 \mu\text{m}$, $E_V^m = 2.0 \text{ eV}$, $E_V^f = 2.0 \text{ eV}$, and $D_V^0 = 0.01 \text{ cm}^2/\text{s}$, the values for τ_B^l at 1000 and 3000 K are 1.3×10^9 and $4.7 \times 10^{-5} \text{ s}$, respectively.

One would expect that, because of enhanced grain-boundary-vacancy diffusion, the grain-boundary-bubble relaxation time, τ_B^g , would be smaller than the lattice-bubble relaxation time. Assuming that E_V^m and D_V^0 for vacancies on grain boundaries are one-half and one-tenth, respectively, of those for vacancies in the lattice, then at K, $\tau_B^g = 1.1 \times 10^5$ s, and at 3000 K, $\tau_B^g = 9.8 \times 10^{-6}$ s.

Instead of directly calculating the individual fission-gas bubble behavior, GRASS groups the continuum of bubble sizes into a relatively small number of bubble-size classes with each size class, i , characterized by an average bubble radius, R_i . The basic GRASS calculation is that of the rate at which bubbles grow from the i to the $i + 1$ size class. The motivation for treating the problem from this level is the much greater facility in performing the calculation (much less computer time required). The effect of a limited rate of growth of coalescing bubbles on the GRASS calculation is a reduction in the rate at which bubbles grow from the i into the $i + 1$ size class by the amount

$$[1 - (1 - \gamma)\exp(-\Delta t/\tau_B)], \quad (34)$$

where τ_B is τ_B^l or τ_B^g , depending on whether the bubbles are intra- or inter-granular, Δt is the code time step (i.e., the fuel temperatures and stresses are assumed constant during Δt s), and α ($0 \leq \gamma \leq 1$) corresponds to the bubble-class transfer rate that would exist in the total absence of point-defect motion.

Equations 31-34, which describe the limited rate of growth of coalescing bubbles, have been included in the GRASS analysis of fission-gas behavior. Consequently, the predicted bubble-size distributions that result from the DEH tests agree qualitatively with the experimental observations. As more quantitative information becomes available, the limits of applicability of the above approach will be better established.

b. Burst Release of Fission Gas

Fission gas trapped on grain faces and along grain edges is susceptible to venting through cracks caused by fuel fracturing. The gas vented during DEH tests where extensive fuel fracturing was observed is an extreme example of this mode of fission-gas release. Few base-line data on fuel fracturing and gas release during fuel fracturing are available to date. However, most of the gas release associated with fuel fracturing is correlated primarily with reactor shutdown.

A preliminary model that describes the burst release of fission gas due to fuel fracturing has been developed and included in the GRASS code. The input to this model is the number of cracks that occur during a time interval in a particular fuel annulus. The LIFE-II cracking model² used in the SST code supplies this information currently for steady-state irradiations. During a power increase (i.e., the fuel temperature decreases at a rate $>0.01^\circ\text{C/s}$), GRASS calculates the rate at which fission gas located in a

closed network of interconnected porosity (i.e., channels initially not connected to the fuel surface) that intercepts a crack vents to the fuel surface. The rate of fission-gas release via fuel cracking is assumed to be proportional to the excess pressure within the channel; i.e.,

$$\frac{dN(t)}{dt} = \alpha[P_0 - P(t)], \quad (35)$$

where $N(t)$ is the moles of gas vented after t seconds, $P(t)$ is the pressure (dynes/cm²) in the channel at time t , P_0 is the initial pressure in the channel, and α is a proportionality constant (moles/g). If the gas within the channel is assumed to be described by the ideal gas law, Eq. 35 becomes

$$\frac{dN(t)}{dt} = \frac{\alpha R T_K}{V} [N_0 - N(t)], \quad (36)$$

where N_0 is the initial number of moles of gas in the channel, V is the volume of the channel (cm³), and R is the gas constant. In Eq. 36, T_K has been assumed constant over the time interval t . The boundary conditions for Eq. 36 are

$$\text{and } \left. \begin{array}{l} N(0) = 0 \\ N(\infty) = N_0 \end{array} \right\} \quad (37)$$

Solving Eqs. 36 and 37 yields

$$N(t) = N_0[1 - \exp(-\alpha_1 T_K t)], \quad (38)$$

where

$$\alpha_1 = \alpha R / V. \quad (39)$$

The constant α_1 will be determined on the basis of a comparison of code predictions with experimental results, when available. Initial results indicate that the relative fractional fission-gas release during power cycling will be greater for fuel rods irradiated at a low rather than a high power level. This can be explained by the fact that for the high-power-level (high fractional fission-gas release) rods a smaller percentage of the retained gas exists along the grain edges; thus, a smaller percentage is available for venting during fuel cracking.

c. GRASS Results for Fission-gas Release under DEH Test Conditions

The GRASS code was run for typical DEH transient-test heating rates using GRASS results for a steady-state irradiation in the H. B. Robinson

nuclear reactor as initial conditions. The DEHTTD-computed fuel-temperature profiles, based on measured DEH fuel surface temperatures, were not used; thus, this GRASS analysis is essentially a qualitative scoping study. Table II.7 compares the measured gas release with the results of the analysis for two heating rates. The experimentally observed ratio of the gas release during the 200°C/s ramp to that for the 25°C/s ramp is between 1:36 and 1:7. The GRASS-estimated gas release (using the standard empirical diffusivities for bubble mobility) for these heating rates yields a ratio of ~1:5.8.

TABLE II.7. Preliminary GRASS Sensitivity Analyses for DEH Test Conditions

Heating Rate, °C/s	Measured Gas Release, %	GRASS-estimated Gas Release, % (No Venting of Channels by Fuel Fracturing)	GRASS-estimated Gas Release, % (Assuming Experimentally Observed Fuel Fracturing Dependence on Heating Rate)
200	1-5	0.37	2.7
25	36	2.14	19.2

In these GRASS calculations for transient fission-gas release, no venting (by fuel fracturing) of the fission gas retained in channels along the grain edges occurred; i.e., GRASS does not contain a model for fuel fracturing. Extensive fuel fracturing was observed (ANL-76-87, pp. 23-32) as a result of the DEH testing, and, furthermore, the volume fraction of fuel that contained the cracks was greater for the slower than for the faster heating rate. The last column in Table II.7 lists the GRASS-calculated fission-gas release assuming channel venting by fuel cracking and using the observed fuel fracturing dependence on heating rate: ~80% of the fuel volume was occupied by cracks for the 25°C/s heating rate compared with 25% for the 200°C/s ramp. The calculated ratio of the release rates for this case is ~1:7.1.

Thus, the DEH experimental results for fission-gas release can be qualitatively understood if it is assumed that the retained fission gas in channels along the grain edges is released by the fracturing of the UO₂. The apparent quantitative discrepancy between core predictions and experiment may in part be due to the release of gas directly from grain boundaries that lie along grain-boundary separations formed at high temperature (ANL-76-87, p. 22). This mechanism of transient gas release was not included in the above calculations. It remains to be determined whether the DEH test conditions that resulted in the extensive fuel cracking occur during hypothetical in-reactor transients. This question is being pursued and will be discussed in future quarterly reports.

d. Model Development

Scanning-electron-microscope (SEM) examinations (ANL-76-87, pp. 23-32) of DEH-tested fuel have revealed the development of sinuous channels on the grain faces after a saturation density of grain-boundary fission gas has

been attained. These face channels link up and extend to the grain-edge channels, thus enhancing the release of the gas from the grains. Models that characterize this phenomenon will be developed and included in GRASS during the next quarter. The effects of grain-boundary fission-gas saturation and grain-boundary fracture on the burnup dependence of steady-state and transient fission-gas release will also be analyzed.

3. FRAP Code Development

Personnel from INEL spent about one week at ANL, during which FRAP-GRASS interfacing was discussed and FRAP-S with GRASS(SS)-Mod 2 was modified to run on the ANL IBM 370/195 computer. GRASS(SST)-Mod 3 is under preparation for transmittal to INEL for inclusion in the FRAP codes. This version of the GRASS code differs from GRASS(SS)-Mod 2, which was sent to INEL on March 1, 1976, in that it includes models for (a) the diffusion of fission gas from the lattice to grain boundaries and along the boundaries to grain edges (applicable under steady-state and transient conditions), (b) the (noninstantaneous) rate of growth of coalescing bubbles, and (c) the burst release of fission gas due to fuel fracturing. A realistic description of fuel swelling requires that local swelling be a function of the local state of stress in the material. An analytical model (ANL-75-58, pp. 35-36) that relates fission-gas-induced swelling to applied stress can be described by

$$\frac{\Delta V_{\text{gas}}}{V} = \frac{RP(J,K) \cdot GS(J,K) \cdot RT}{PG(J,K) + PRSO(J,K)} + GS(J,K) \cdot BVI(J,K), \quad (40)$$

where

$$\frac{\Delta V_{\text{gas}}}{V} = \text{swelling strain due to fission gas,}$$

V = volume of annular region (J,K); J refers to the axial section (maximum of 10) and K refers to the radial region (maximum of 15),

$RP(J,K)$ = fraction of retained gas that resides in bubbles in region (J,K),

$GS(J,K)$ = amount of retained fission gas in zone (J,K),

R = gas constant,

T = average temperature in region (J,K),

$PG(J,K)$ = effective surface-tension-induced pressure in region (J,K)

$PRSO(J,K)$ = hydrostatic pressure in zone (J,K), and

$BVI(J,K)$ = volume per mole of dissolved fission gas.

On the basis of local values of temperature and hydrostatic stress (among others), GRASS computes the volume strain due to the retained fission gas for a specified time interval. This component of fuel swelling can then be included in expressions for total fuel swelling, the solution of which results in new values for the total strains and hydrostatic pressure.

A model that relates fission-gas-induced swelling to applied stress, such as described by Eq. 40, is essential for the FRAP-code utilization of GRASS (or any realistic model for fission-gas release and fuel swelling). In addition, a sophisticated stress analysis is required to predict fuel fracturing, which is essential input to models that describe the burst release of fission gas (e.g., GRASS).

The inclusion of Eq. 40 in the FRAP swelling analysis will facilitate the effective use of GRASS in FRAP.

B. Experimental Program (M. G. Seitz and L. R. Kelman, MSD)

Transient-heating tests of irradiated Robinson fuel were performed this quarter, using the DEH apparatus, to determine the fission-gas release and fuel microstructural changes that may be produced in hypothetical in-reactor transient-heating events. For three tests of equivalent heating rate that were interrupted at various times prior to fuel melting, from 0.8 to 14% of the total amount of xenon present in the fuel was released.

Characterization of the fuel is conducted as part of the effort to determine fuel behavior that affects the motion and release of fission products from LWR fuels under transient-heating conditions. In previous examinations of pellets in which central melting was produced, interconnected separations at fuel grain boundaries were observed that could directly vent fission gases from the fuel. In some cases these separations formed long, wide tears visible at low magnification. The separations appear to be a manifestation of changes in fuel mechanical properties caused by exsolution of fission products and migration to grain boundaries. This quarter the posttest examinations were extended to the fuel specimens that were transient-heated to temperatures below the fuel melting temperature. Therefore, the test results are of direct consideration to hypothetical LWR transient-heating accidents in which fuel melting is in general not expected to occur.

Knowledge of the fission-product concentration in irradiated LWR fuel and the radial distribution of fission gases is important to characterization of fission-product release during transient experiments. Fission-product concentrations were determined by fuel dissolution and were used to characterize volatile fission-product release from transient-tested fuel. Measurements of the total gas content and results of laser sampling of fission gas within a pellet are reported.

1. Transient-heating Tests (M. G. Seitz and D. R. Pepalis, MSD)

a. Test Conditions

Three DEH transient tests using H. B. Robinson fuel were conducted at a slow heating rate, as specified in the initial DEH transient-test plan (ANL-76-15, p. 45). The conditions of the tests, all of which were terminated prior to melting of the fuel, are listed in Table II.8.

Fuel preparation was the same in all three tests. The fuel columns consisted of an irradiated fuel pellet with unirradiated UO_2 pellets above and below the fuel. The pellets were stacked in a quartz container of 1-mm wall thickness and ~10-mm inside diameter. With the as-fabricated diameter of the fuel pellets of 9.30 mm and a thermal expansion from room temperature of <5% in unmolten fuel,⁵ quartz-tube containers from 9.98 to 10.29 mm were chosen to avoid constraint of the fuel during heating. Containers that constrain the fuel during heating will be considered in future tests.

TABLE II.8. Conditions of Interrupted DEH Transient Tests on H. B. Robinson Fuel

Test Number	Surface Temperature, ^a °C		Transient-heating Time, s	Fuel Burnup, ^b %	Fuel Weight, g
	Initial	Maximum			
22	807	1510	36	2.82	10.7
23	695	1290	28	3.00	10.5
24	774	1625	52	3.13	10.3

^aInitial surface temperature was determined by thermocouple measurement, maximum surface temperature by optical pyrometer measurement.

^bFuel burnup estimated from gamma-scan intensity normalized using neodymium mass-spectrometric burnup analyses (ANL-76-15, p. 50).

Fig. II.1. Fuel Surface and Centerline (Calculated) Temperatures in Three Interrupted Transient Tests. Neg. No. MSD-63581.

The use of molybdenum powder to improve electrical continuity of the fuel column was continued. An accurate knowledge of the temperature distribution in the fuel during heatup and cooldown is important when considering fission-product migration and release. Temperature profiles within the fuel were calculated using the DEHTTD code (ANL-75-58, p. 40), which was modified to predict temperatures after the electrical power was terminated. The input parameters for the calculations were the fuel surface temperature, measured by thermocouples and pyrometer, and current, voltage, and power input, measured by root-mean-square converters (ANL-76-15, p. 40). Centerline-temperature prediction and measured surface temperatures for the three tests are shown in Fig. II.1.

The electrical continuity of the fuel columns was measured before each test and was similar to that reported in ANL-76-15 (p. 49). In addition, the electrical conductivity of two of the fuel columns was measured after the transient-heating tests. The column that was transient-heated for a short time (Test 22) showed little change in electrical conductivity, even though the depleted-UO₂ pellets were cracked from the heating. The column that was transient-heated for a longer period (Test 24) showed a decrease

in electrical conductivity. This decrease in the fuel-column conductivity may be related to restructuring of the fuel.

Changes in electrical conductivity affect code prediction of temperature profiles during heatup of the fuel column. The predictions, however, are not expected to be sensitive to changes in material properties, because they are derived from matched solutions to redundant heat-flow equations. Even so, consideration will be given to the changes in electrical properties, with possible refinements to the prediction of temperature profiles.

b. Fission-gas Release during Transient Heating (M. G. Seitz and D. R. Pepalis, MSD)

Fission gases were collected and analyzed in the three transient-heating tests that were terminated prior to melting of the fuel, and the results are listed in Table II.9. The percentage of gas released was calculated from the collected fission gas, fuel weight and burnup provided in Table II.8, and total retained gas in sibling fuel, obtained by a dissolution method.

TABLE II.9. Fission Gas Collected from Interrupted DEH Transient Tests

Test Number	Fission Gas Collected, cm ³ (STP)		Percent Released ^a	
	Xe	Kr	Xe	Kr
22	0.91	0.13	14	26
23	0.058	0.011	0.8	2.0
24 ^b	0.67	0.16	9.5	30

^aBased on the total gas measurements of the present report and corrected for fuel burnup provided in Table II.8.

^bGas recovered from Test 24 represents only a portion of the collected gas. Analysis of the remaining gas samples is in progress.

Figure II.2 is a plot of the gas release data for xenon as a function of the length of time the transient power was applied. The results are for four tests run at the slow heating rate: the three that were terminated before central melting of the fuel and one, Test 21, that was run last quarter (ANL-76-87, p. 16) to central melting of the fuel. The gas released from unmolten fuel was obtained by subtracting from the total gas released all the gas in the fuel that melted. In addition, the total gas release from Test 21 is plotted, with a dashed line projected back to the estimated time of fuel melting.

Fig. II.2

Gas Release vs Time of Transient Power Application at the Slowest Heating Rate. Numbers refer to test numbers. Neg. No. MSD-63585.

Incomplete sampling of fission gas from the charcoal trap is believed to have occurred in Test 24 as a result of too low a temperature during gas extraction. Thus, the measured values shown in Table II.9 and Fig. II.2 for Test 24 are believed to be low. Subsequent extraction has been performed at higher temperature to recover any residual fission gases, and this gas is being analyzed.

In Fig. II.2, a threshold for initial fission-gas release appears to exist at a heating time of 28 s. Gas release in the tests with longer heating times increases rapidly to ~30% release at 60 s.

As observed in Table II.9, considerably less krypton is released from the fuel than xenon, such that the importance of krypton motion and release is less than that for xenon. The percentage of krypton released, however, is higher than for xenon; this indicates that the relatively lighter krypton may be more mobile than xenon. This is consistent with the high krypton gas release relative to xenon from fuel during steady-state operation.¹² The measured xenon/krypton release ratios during transient heating, steady-state operation,¹² and in the as-received fuel (Sec. II.B.2.c of the present report) are 4-7, 8, and 13, respectively.

Figure II.3 is a plot of gas release versus heating rate for DEH transient-heating tests reported in ANL-76-87 (p. 16). Fuel melt fractions

Fig. II.3

Gas Release vs Heating Rate for Tests to Center Melting. Two isomelt curves (0 and 11% melt fractions) are shown. Neg. No. MSD-63583.

were measured from polished sections and used to calculate gas release at incipient melting (assuming release of 100% of gas in the fuel that melted).

2. Fuel Characterization (M. G. Seitz, MSD)

a. Posttest Characterization (M. G. Seitz and J. J. Puro, MSD)

The microstructure of fuel from transient Test 22 is shown in Fig. II.4. No evidence of melting was observed. However, the microstructure was profoundly affected by the heating and shows the same features observed previously in fuel that had undergone central melting. The heat-affected zone has low optical reflectivity, the result of porosity from gas-bubble precipitation, fuel separation at grain boundaries, and loss of grains during polishing. Large tears within the fuel are also visible at this magnification. The grain-boundary separations can serve as an avenue for the release of gas that has precipitated on grain boundaries.

2.0 mm

Fig. II.4. Fuel from Transient Test 22 Showing Grain-boundary Separations and Tearing without Fuel Melting. Neg. No. MSD-188772.

As discussed under Sec. II.B.1.a, the DEH test specimen was designed so that the fuel pellet does not expand to the quartz container during the tests interrupted, short of fuel melting. Therefore, no lateral constraint was present. The fuel apparently responded to forces such as fission-gas pressure. Constraint is an important parameter when considering gas release from transient-heated fuel. With total constraint producing large hydrostatic forces on the fuel and gases, precipitated gas bubbles would remain small

and very little fuel motion, such as that which would cause separations or tears, would occur. Also, no gas would be lost outside of the constraining wall. During hypothetical accident conditions, an overall diametral restraint may occur, with little constraint in some regions, i.e., at pellet ends and between fuel particles that have moved apart. Therefore, the effect of cladding constraint on gas release will be explored in future experiments.

Although the degree of constraint may affect the capability of the fuel to expand and subsequently release fission gas, it is not expected to affect the underlying changes in fuel mechanical properties produced upon transient heating. These changes are controlled by diffusion rates, melting temperatures, and other physical-chemical parameters that are not sensitive to pressure.

Evidence presented last quarter (ANL-76-87, p. 20) suggests that intergranular separations result from stresses in fuel in which fission gases have moved to and weakened grain boundaries. Verification that other fission products occur at the grain boundaries is complicated because the material at boundaries is thin, possibly no greater than 1000 Å thick, and distributed over large areas of grain surfaces ($\sim 1 \times 10^5 \text{ cm}^2$ of surface per cm^3 of fuel). The fuel was not heated sufficiently long to permit fission products to form large masses. Detection methods such as electron-beam excitation and X-ray identification are not practical because of the deep penetration of keV-energy electrons.

Consequently, a technique was used based on the chemical treatment of cesium to make it visible in an optical microscope. The technique used a spot test specific for cesium, in which potassium and bismuth iodide are treated with acetic acid and applied to the fuel.¹³ The cesium in the fuel forms a bright red precipitate of Cs_2BiI_5 and, in small quantities, is visible as a white translucent precipitate. A procedure for application of the technique to irradiated fuels is given by Clifton and Jepson.¹⁴

The technique was applied to both as-received and transient-heated fuel. The fuel was polished using kerosene, a nonpolar lubricant and wash. Alcohol was specifically excluded to prevent the loss of cesium during polishing. The as-received material appeared not to be affected by the chemical treatment. This result is interpreted as an indication that the cesium remains dispersed within the fuel grain and nowhere achieves concentrations sufficiently high to produce a visible precipitate. The technique is purported to be sensitive to $1 \times 10^{-12} \text{ g}$ cesium per $1 \times 10^{-6} \text{ m-dia}$ spot.¹⁴

The fuel from Test 21, however, showed a red coloration of the metal ingots within the molten fuel region, which indicates a positive test for cesium. Furthermore, a translucent precipitate was observed at the grain boundaries in the heat-affected zone of the fuel, where gas-bubble precipitation and grain-boundary separations were observed. Figure II.5 shows

0.05 mm

Fig. II.5. Polarized-light Photograph of Cesium Precipitate at Grain Boundaries of Transient-heated Fuel. Neg. No. MSD-188768.

Test 21, an optical photograph of which was shown in the last quarterly report (ANL-76-87, p. 20). Except for a redistribution in the molten fuel region, no large-scale migration of fission products is observed. Specifically, no fission-product accumulation is observed that could correspond to the chemically sensitive band revealed by etching, which was reported last quarter. The lack of large-scale migration is expected, on the basis of the relatively small releases of fission products (~1% for cesium) observed in some tests.

the precipitate in polarized light. The region of fuel in this photograph corresponds approximately to the reflected-light photograph presented last quarter (ANL-76-87, p. 20). This observation is evidence that cesium accompanies fission gases to grain boundaries during transient heating.

Fission products other than cesium, e.g., molybdenum and ruthenium, are known to have low solubilities in UO_2 and would also be expected to separate from the grains during transient heating and concentrate at grain boundaries.

Autoradiographs were taken of some of the transient-tested fuel pellets. Figure II.6 is a beta-gamma autoradiograph of the cross section of fuel from

Fig. II.6

Autoradiograph of a Polished Cross Section of Transient-heated Fuel. Central region was molten. Neg. No. MSD-188767.

b. Radial Distribution of Fission Gas (M. G. Seitz, MSD)

Laser sampling of the as-received irradiated fuel was performed to determine the radial profile of fission gas within the fuel pellets. The technique and the sample used in the work were described last quarter (ANL-76-87, p. 24).

The radioactivity of the ^{85}Kr was measured in each sample from 12 radial positions in the fuel and was converted to relative gas concentrations using the measured volume of the craters formed by laser heating. The relative gas concentrations were normalized to the total gas measurements obtained by fuel dissolution. Figure II.7 shows the measured radial distribution of fission gas. The error bars in the figure are based on errors in the measurement of the relative gas concentrations and the crater volumes.

Fig. II.7. Measurements of Radial Fission-gas Distribution by Laser Sampling.
Neg. No. MSD-63582.

The statistical error that results from the number of disintegrations counted is $<1\%$. The major source of error is from the measurement of the crater volumes. The craters in this sampling were $\sim 10\ \mu\text{m}$ in radius and $\sim 7\ \mu\text{m}$ deep. It is estimated that the dimensions can be measured to $\pm 1\text{-}\mu\text{m}$ accuracy, which leads to an expected error of about one-third in crater volume. The error at each radial position is the average for the five craters. As shown in Fig. II.7, the error is large and does not permit a detailed description of the gas concentration with radial position.

In general, the gas concentrations, as indicated by the ^{85}Kr activities, do not show significant variations over the radius of the pellet. An increase in concentration on the edge of the pellet near the cladding is evident, however. This increase is probably due to increased burnup at the surface of the pellet relative to the center. Extremely little fission-gas redistribution is expected, on the basis of the small gas release ($\sim 0.2\%$) that occurred from the fuel during power operation. Craters of different dimensions can be produced by, for example, changes in focusing and time between laser shots. In future laser sampling work, larger craters will be produced to minimize error associated with thin measurement.

c. Pretest Total Gas and Fission-product Analyses (M. G. Seitz and D. R. Pepalis, MSD)

To determine the percentage of gas released during a DEH test, the total gas content of the pretested fuel must be known. The quantity

of gas in a high-burnup portion of the H. B. Robinson fuel was determined to be $0.685 \pm 0.006 \text{ cm}^3 \text{ (STP) Xe/g fuel}$ and $0.0533 \pm 0.009 \text{ cm}^3 \text{ (STP) Kr/g fuel}$ with the use of the total fission-gas analysis system described in the previous quarterly report (ANL-76-87, p. 32).

For the analyses, a length of fuel approximately equivalent to one pellet was cut from two adjacent fuel pellets. The remaining portions of the pellets were polished and determined to have the structure of most of the Robinson fuel pellets (ANL-76-87, p. 18). Chemical analyses and the quantities of fission gases collected from five gas samples are presented in Tables II.10 and II.11, respectively. Gas sample 2 contains the fission gases released upon fuel dissolution. The first sample was obtained before fuel dissolution and represents a background measurement; the final three samples were obtained to verify that all the fission gases were recovered from the system.

TABLE II.10. Chemical Analyses of Gases Recovered by Fuel Dissolution

Gas Sample	mole %								
	He	H ₂ O	N ₂	NO _x	O ₂	Ar	CO ₂	Kr	Xe
1	0.7	8.1	75.0	0.1	14.3	0.9	0.9	<0.05	<0.05
2	0.3	0.2	51.9	37.1	0.1	0.2	3.2	0.50	6.4
3	1.1	28	51.0	7.0	1.4	0.9	10.6	<0.01	<0.03
4	0.03	13	43.5	1.0	1.1	0.5	40.8	0.01	0.02
5	<0.01	0.6	78.3	0.2	18.8	1.1	1.0	<0.01	<0.02

TABLE II.11. Quantities of Gas Collected from Dissolutions of 8.6 g of Fuel

Gas Sample	Volume, cm ³ at STP		
	Total	Xe	Kr
1	11.86	$<5.9 \times 10^{-3}$	$<5.9 \times 10^{-3}$
2	91.61	5.86	0.458
3	36.01	$<1.08 \times 10^{-2}$	$<3.6 \times 10^{-3}$
4	37.65	$<7.5 \times 10^{-3}$	$<3.7 \times 10^{-3}$
5	93.69	$<1.87 \times 10^{-2}$	$<9.4 \times 10^{-3}$
Total		5.89 ± 0.02	0.458 ± 0.005

The tables indicate that virtually all the fission gases were removed immediately after dissolution of the fuel. The fission gases were accompanied by significant quantities of nitrogen, oxides of nitrogen, and water vapor that were presumably generated during dissolution of the fuel and were not retained by the molecular sieve. Experimental uncertainties were determined from the detectability limit of gas in the samples and a possible error of 0.05 g in the measurement of fuel weight.

Isotopic analyses of the fission gas in the H. B. Robinson fuel are presented in Table II.12. Except for variations within the accuracy of the measurements, the compositions of all fission-gas samples obtained from the transient-heating tests (e.g., ANL-76-49, p. 25) are the same as the composition of the total gas sample. For a given gas, the partial gas releases produced during transient testing show no evidence of mass-fractionation effects.

TABLE II.12. Isotopic Analysis of Fission Gas in H. B. Robinson Fuel

Xenon			Krypton		
Isotope ^a	Concentration, %	Accuracy	Isotope	Concentration, %	Accuracy
128	0.03	±0.01	83	11.4	±0.3
130	0.14	±0.02	84	31.2	±0.4
131	8.0	±0.2	85	6.0	±0.3
132	20.5	±0.3	86	51.3	±0.3
134	28.1	±0.3			
136	43.3	±0.4			

^aA detectable amount of an isotope of mass 129, which is possibly ¹²⁹I, was also present.

The concentrations of some fission products within the fuel were determined using standard radiochemical counting techniques on an aliquot of the fuel solution. Table II.13 lists the quantities of radionuclides in the Robinson fuel, determined from the measurements.

TABLE II.13. Quantities of Radionuclides in High-burnup Portion of H. B. Robinson Fuel

Nuclide	Concentrations, atoms/g fuel		Counting Error, %
	Atoms at Count Time ^a	Atoms at EOL ^b	
¹³⁴ Cs	6.62 x 10 ¹⁵	1.45 x 10 ¹⁶	2.7
¹³⁷ Cs	1.47 x 10 ¹⁷	1.55 x 10 ¹⁷	0.1
¹⁰⁶ Ru	7.23 x 10 ¹⁵	3.42 x 10 ¹⁶	2.0
¹⁴⁴ Ce	1.19 x 10 ¹⁶	9.21 x 10 ¹⁶	0.3

^aSample was counted August 22, 1976.

^bEOL was May 6, 1974.

To examine whether fission products other than the rare gases were lost from the fuel solution, the radioactivity of the basic wash was measured. Fractional transfer of detected isotopes (Table II.14) is small. Since the basic wash is the major trap for fission products that leave the fuel solution, the loss of fission products from the solution is small. The variability in the different isotope fractions in the basic wash suggests that transfer by vapor as well as aerosols may have occurred. The transfer of ^{106}Ru is less than that for the cesium isotopes. This is unexpected in view of the likelihood of the formation of volatile ruthenium oxides during reactions with HNO_3 .

TABLE II.14. Loss of Radioactive Isotopes from Fuel Solution during Fission-gas Extraction

Isotope	Fractional Transfer to NaOH Wash	Atoms in NaOH Wash ^a
^{134}Cs	9.0×10^{-5}	5.14×10^{12}
^{137}Cs	8.9×10^{-5}	1.13×10^{14}
^{106}Ru	3.8×10^{-5}	2.34×10^{12}
^{144}Ce	1.9×10^{-5}	1.98×10^{12}
^{125}Sb	b	2.75×10^{12}

^aSample was counted August 14, 1976.

^bAntimony was not measured in the fuel solution.

The concentrations of fission products within the fuel may be used to compute the amount of these fission products that are released during DEH tests. Table II.15 lists the fractional release of cesium, ruthenium, and

TABLE II.15. Fractional Fission-product Release to Quartz Container during a DEH Test

Isotope	Fission-product Release, ^a %
^{134}Cs	1.0
^{137}Cs	0.96
^{106}Ru	0.008
^{144}Ce	0.009

^aTest was run to center melting of fuel in 20 s (ANL-76-49, p. 27).

cerium isotopes to the quartz container during a transient test with center melting (ANL-76-49, p. 27). Since most of the activity released during the transient is deposited on the quartz container, these numbers provide an estimate of the total fission-product release in the test.

3. Experimental Technique (M. G. Seitz and D. R. Pepalis, MSD)

Calibration of Chromel/Alumel Type K thermocouples used to measure surface temperatures in DEH tests against tungsten/tungsten-26% rhenium thermocouples indicate that the Type K thermocouples are accurate to the 1040°C temperature achieved in the calibration. When surface temperatures higher than 1150°C are achieved in DEH tests, as indicated by optical pyrometer readings, the thermocouple and optical pyrometer readings are not in agreement. Because the optical pyrometer was calibrated and is used as described in ANL-75-72 (p. 38), it is relied upon for high-temperature measurements. To eliminate the discordance in measurements at high temperature, platinum/platinum-10% rhodium thermocouples will replace the Type K thermocouples and should be useful at the maximum surface temperatures achieved in the tests.

The conductive molybdenum paint described in the previous quarterly report (ANL-76-87, p. 30) was used in the preparation of the fuel columns for the three tests run during the present quarter. No electrical arcs, thermal hot spots, or other indications of discontinuities in electrical conductivity were observed. From posttest examinations, the molybdenum does not appear to react with the fuel, and its use will be continued.

References

1. W. Y. Johnston, NRC, private communication (Aug 12, 1976).
2. M. C. Billone, B. L. Harbourne, V. Z. Jankus, R. B. Poeppel, T. S. Roth, W. J. Shack, J. D. Stephen, B. Sundquist, F. R. Taraba, J. E. Turner, and S. Zawadzki, The LIFE-III Fuel-element Performance Code User's Manual (ANL report; to be published).
3. L. S. H. Chow, Improved Model for Heat Transfer across the Fuel-cladding Gap in LMFBR Fuel Elements (ANL report; to be published).
4. M. G. Andrews, H. R. Freeburn, and S. R. Pati, Light Water Reactor Fuel Rod Modeling Code Evaluation, Phase II Topical Report, CENPD-218, Combustion Engineering, Inc., Appendix A (Apr 1976).
5. P. E. MacDonald, ed., MATPRO: A Handbook of Materials Properties for Use in the Analysis of Light-Water-Reactor Fuel Rod Behavior, ANCR-1263, NRC-5, Aerojet Nuclear Company (Feb 1976).
6. J. Rest, unpublished work.

7. J. Rest, SST: A Computer Code to Predict Fuel Response and Fission Product Release from Light-Water-Reactor Fuels During Steady-state and Transient Conditions, Trans, Am. Nucl. Soc. 22(1), 462-463 (Nov 1975).
 8. E. T. Laats, P. E. MacDonald, and W. J. Quapp, USNRC--OECD Halden Project Fuel Behavior Test Program--Experiment Data Report for Test Assemblies IFA-226 and IFA-239, ANCR-1270, NRC-3, Aerojet Nuclear Company (Dec 1975).
 9. J. M. Broughton, D. H. Malin, C. R. Hann, S. R. Wagoner, J. Rest, and M. Billone, Comparison of Results from the Steady-state Fuel Behavior Codes GAPCON-THERMAL, LIFE-LWR and FRAP-S, presented at workshop on Code Verification--Fuel Behavior Research, Fourth Water Reactor Safety Research Information meeting, Washington, D. C. (Sept 1976).
 10. M. W. Finnis, M. R. Hayns, and R. Bullough, The Response of Fission Gas Bubbles to Rapid Heating, AERE-R-7920, Atomic Energy Research Establishment, Harwell (1975).
 11. F. R. Campbell, R. Deshaies, and M. J. F. Notley, Transient Fission Gas Release Rates within UO₂ Fuel Elements Following Power Increases, AECL-4912, Chalk River Nuclear Laboratories (1974).
 12. R. Lorenz, J. Collins, and S. R. Manning, Quarterly Progress Report on Fission-product Release from LWR Fuel for the Period October-December 1975, ORNL/TM-5290, Oak Ridge National Laboratory.
 13. F. Feigl and V. Anger, Spot Tests in Inorganic Analysis, 6th Edition, Elsevier Publishing Company (1972).
 14. M. Clifton and M. D. Jepson, "A Technique for the Observation of Micro-segregation of Caesium in Irradiated Fuel Pins," Post-Irradiation Examination Techniques, Proc. Eur. Conf. on Post-Irradiation Examination Techniques, British Nuclear Energy Society, London (1972).
-

III. MECHANICAL PROPERTIES OF ZIRCALOY CONTAINING OXYGEN

H. M. Chung, A. M. Garde, and T. F. Kassner, MSD

A. Summary

Uniaxial and biaxial mechanical-property data on Zircaloy-4 are being obtained over a wide range of temperature and strain rate to establish a more quantitative cladding-embrittlement criterion applicable to postulated loss-of-coolant-accident (LOCA) situations in light-water reactors (LWR's). The mechanical-property information will be incorporated into fuel-element modeling codes that will provide a quantitative basis for evaluating cladding deformation over a wide range of LOCA and power-coolant-mismatch (PCM) conditions.

The uniaxial tensile properties of as-received Zircaloy-4 cladding have been determined between room temperature and 1000°C. At temperatures $\leq 800^\circ\text{C}$, the ultimate tensile and yield strengths of the cladding are greater than for the sheet material, primarily because of the fine grain size of the tubing. Above $\sim 800^\circ\text{C}$, the tubing recrystallizes rapidly and the properties of the Zircaloy cladding and sheet are essentially the same. The dependence of the total strain on temperature is similar, although the tubing is somewhat less ductile at temperatures $\leq 400^\circ\text{C}$ and more ductile at higher temperatures.

More complete results have been obtained for the strain-rate dependence of the 0.2% yield stress of 5- and 11- μm grain-size Zircaloy-4 sheet at several temperatures between 700 and 1000°C. These data were used to determine the activation energy for the Stage III deformation process for Zircaloy. The activation energies between 270 and 352 kJ/mole are in reasonable agreement with the activation energy for self-diffusion in α -Zircaloy. Relationships between the strain rate and the flow stress for a dislocation creep process (at strain rates $\geq 10^{-3} \text{ s}^{-1}$ for Stage III deformation) and grain-boundary sliding mechanism with diffusional accommodation (at lower strain rates for Stage II deformation) were used to calculate the dependence of the yield stress on the strain rate at 850 and 900°C for the 5- and 11- μm grain-size materials. The calculated curves are in good agreement with the experimental data for the range of strain rates in which the transition from Stage III to Stage II behavior occurs as the strain rate decreases. Microstructural observations are also in general agreement with the deformation mechanisms in the respective regions.

Additional information has been obtained on the diametral expansion and rupture characteristics of Zircaloy-4 cladding in vacuum and steam environments. To evaluate the effect of axial constraint on the relationship between the maximum circumferential strain and the burst temperature at a low heating rate, a number of tube-burst experiments were performed at a heating rate of 5°C/s in vacuo and steam. A maximum in the circumferential strain was observed at $\sim 810^\circ\text{C}$ for the axially unconstrained and constrained tubing in vacuo. The circumferential strains were $\sim 40\%$ higher for the unconstrained tubing at

temperatures between ~650 and 900°C. Similar to the behavior of constrained cladding at this heating rate, the circumferential expansion of the unconstrained tubing was greater in steam than in vacuo between ~700 and 800°C.

The effect of initial pressure on the maximum circumferential strain of axially constrained Zircaloy-4 cladding in steam at heating rates of 5, 55, and 115°C/s was reported in the previous quarter. The data from these experiments have been analyzed to establish the diametral strain as a function of temperature at the three heating rates. A comparison of these results with similar information obtained for tubes ruptured in vacuo revealed that oxidation decreased the diametral strain at failure more than the strain at the onset of plastic instability for burst temperatures $\geq 850^\circ\text{C}$.

To explore the possibility that thermal shunting and/or in-situ oxidation of the thermocouples in steam could result in greater uncertainties in the burst-temperature measurements, nonpressurized specimens were preoxidized in steam under transient-heating conditions, rapidly cooled to room temperature, and then burst in vacuo at the same heating rate. The maximum circumferential strains for the preoxidized specimens ruptured in vacuo were in good agreement with the in-situ burst results in steam at temperatures between ~780 and 830°C. The results confirm that errors in the temperature measurements due to thermal shunting and oxidation are negligible in our experiments.

An analysis of the additional tangential stress introduced by the oxide layer and its effect on the biaxiality condition during transient-heating (5°C/s) burst experiments was conducted to rationalize the enhanced circumferential strain in a steam environment for rupture temperatures in the α -phase region.

Several short-term isothermal stress-rupture tests were performed on axially constrained cladding in steam and vacuo at temperatures between ~720 and 840°C. The circumferential strains at rupture were in good agreement with values from transient-heating experiments (115°C/s) in the respective environments. In these experiments, the rupture circumferential strain in steam was considerably greater than in vacuo at burst temperatures near 825°C.

B. Mechanical Properties of As-received Zircaloy

1. Comparison of the Tensile Properties of Stress-relieved Tubing and Recrystallized Sheet

The tensile properties of Zircaloy-4 sheet with a recrystallized equiaxed grain size of 11 μm have been reported in previous quarterly reports.¹⁻⁴ For comparison with these results, uniaxial-tensile tests have been performed on the Zircaloy-4 tubing⁵ used in the biaxial tube-burst tests and other Nuclear Regulatory Commission research programs. The microstructure of the material in the as-received stress-relieved condition consists of fine (~2- μm -wide) elongated grains. The wall thickness of the central 10-mm gauge length of the

70-mm-long specimens was reduced from 0.635 to 0.432 mm to ensure that fracture occurred away from the Swageloks that gripped the samples.

Figure III.1 shows the temperature dependence of the ultimate tensile strength and yield stress of the tube material. The properties of the recrystallized sheet are shown for comparison. At temperatures below $\sim 800^{\circ}\text{C}$, the fine grain size and elongated grain shape of the cold-worked material are primarily responsible for the higher strength of the tubing. At higher temperatures, complete recrystallization of the tubing occurs during the tensile test, and the flow stress of the sheet and tube materials is essentially the same.

The total strain of the Zircaloy tube and sheet is plotted in Fig. III.2 as a function of the deformation temperature. The materials exhibit similar behavior, although the tubing is somewhat less ductile below $\sim 400^{\circ}\text{C}$ and more ductile at higher temperatures. The temperature dependence of the uniform strain of the two materials is shown in Fig. III.3. Below $\sim 850^{\circ}\text{C}$, the uniform strains are comparable; at 900°C , however, the tube material exhibits a large strain (~ 0.7) that was confirmed by duplicate tests. This behavior can be related to the microstructural changes that occur during deformation. Whereas the sheet material has an equiaxed grain structure before and during deformation and grain growth is not significant, the tube recrystallizes and the small

Fig. III.1. Ultimate Tensile Strength and 0.2% Yield Stress of Zircaloy-4 Stress-relieved Tube and Recrystallized Sheet Material as a Function of Temperature. Neg. No. MSD-63395.

Fig. III.2. Total Strain of Zircaloy-4 Tube and Recrystallized Sheet as a Function of Temperature. Neg. No. MSD-63396.

Fig. III.3

Uniform Strain of Zircaloy-4 Tube and Recrystallized Sheet as a Function of Temperature. Neg. No. MSD-63401.

equiaxed grains grow considerably during the tensile test. The higher work-hardening rate, associated with grain growth, and the grain-boundary sliding deformation mechanism result in a larger uniform strain in the tube material at $\sim 900^\circ\text{C}$.

2. Activation Energy for the Deformation of Zircaloy-4 at High Temperatures

If the deformation of Zircaloy is thermally activated at high temperatures, the strain rate $\dot{\epsilon}$ during steady-state deformation can be expressed in terms of a constant A , shear modulus G , Boltzmann constant k , Burgers' Vector b , stress σ , grain size d , absolute temperature T , grain-size exponent p , stress exponent n (i.e., $n = 1/m$, where m is the strain-rate sensitivity parameter), activation energy for deformation Q , and the gas constant R ; i.e.,

$$\dot{\epsilon} = \frac{AG(b/d)^p}{kT} \left(\frac{\sigma}{G} \right)^n \exp\left(-\frac{Q}{RT}\right). \quad (1)$$

A plot of $\log \dot{\epsilon} kT/G$ versus reciprocal temperature at a constant value of σ/G and grain size can be used to evaluate Q . As pointed out by Gifkins,² two precautions should be taken to obtain a meaningful value for Q ; i.e., the temperature range should be selected such that the stress exponent (and hence the strain-rate sensitivity) is approximately constant, and no change in the deformation mechanism should occur for the particular value of σ/G .

More complete results for the strain-rate dependence of the 0.2% yield stress of 5- and 11- μm grain-size Zircaloy-4 at several temperatures between 700 and 1000°C are shown in Figs. III.4 and III.5, respectively. These data suggest a tendency toward an S-shape curve, as observed in other superplastic materials, although a constant threshold stress⁷ at low strain rates is not evident. The comparatively large slope of the curves at temperatures

between 850 and 950°C and stress levels ≤ 6 MPa is indicative of a high value of m (Stage II deformation); at higher stress levels, however, the m value is lower (Stage III deformation). As the temperature increases, the transition from Stage II to Stage III behavior occurs at higher strain rates.

Fig. III.4. Strain-rate Dependence of the 0.2% Yield Stress of 5- μ m Grain-size Zircaloy-4 at Several Temperatures between 700 and 950°C. ANL Neg. No. 306-76-256.

Fig. III.5

Strain-rate Dependence of the 0.2% Yield Stress of 11- μ m Grain-size Zircaloy-4 at Several Temperatures between 700 and 1000°C. ANL Neg. No. 306-76-242.

The temperature dependence for the Stage III deformation process for the 5- and 11- μ m grain-size material is shown in Figs. III.6 and III.7, respectively, in which the strain rate and compensated strain rate, at constant stress σ or σ/G , are plotted as a function of reciprocal temperature. The activation energies for Stage III deformation, defined by either

$$Q = - \left. \frac{Rd \ln(\dot{\epsilon})}{d(1/T)} \right|_{\sigma} \quad (2)$$

or

$$Q = - \left. \frac{Rd \ln(\dot{\epsilon}kT/G)}{d(1/T)} \right|_{\sigma/G}, \quad (3)$$

were computed from the slopes of the respective curves. The shear modulus of zirconium, calculated from the relationship given by Hearmon⁸ and the

elastic-constant data of Fisher and Renken,⁹ was used to obtain the compensate strain-rate curves. The activation energies in the range of 270-352 kJ/mole obtained from Eqs. 2 and 3 are stress-dependent; i.e., the Q values in Figs. III.6 and III.7 decrease with an increase in stress. This behavior is consistent with the increase in the m value with temperature over the 700-900°C

Fig. III.6

Temperature Dependence of the Strain Rate for Stage III Deformation of 5- μ m Grain-size Zircaloy-4. ANL Neg. No. 306-76-250.

Fig. III.7

Temperature Dependence of the Strain Rate for Stage III Deformation of 11- μ m Grain-size Zircaloy-4. Neg. No. MSD-63431.

range (Figs. III.5 in ANL-76-87⁴ and III.2 in ANL-76-49³ for the 5- and 11- μm grain-size material, respectively). The values of the activation energy are in good agreement with those obtained by Kearns et al.¹⁰ for the deformation of Zircaloy-hydrogen alloys (304-365 kJ/mole) and in reasonable agreement with the activation energies for self-diffusion in α -Zircaloy (218,¹¹ 276,¹² and 282,¹³ kJ/mole).

An experimental determination of the stress and grain-size exponents, n and p in Eq. 1, is frequently used to evaluate the relative contributions of different mechanisms of superplastic deformation. Figure III.8 is a logarithmic plot of the strain rate versus grain size for Stage II deformation of Zircaloy at 850°C. The value of p , in Eq. 1, determined from the slope of the curve is -1.

Fig. III.8

Logarithmic Plot of the Strain Rate as a Function of Grain Size for Stage II Deformation of Zircaloy-4 at 850°C. ANL Neg. No. 306-76-248.

To evaluate the deformation process in the two-phase material at 850 and 900°C, values of the total strain rate $\dot{\epsilon}_T$ were calculated from the contributions from dislocation creep, in Eq. 1, and from the grain-boundary sliding mechanism with diffusional accommodation,⁷ in Eq. 4:

$$\dot{\epsilon}_{D-A} = \frac{100\Omega D_V}{d^2 kT} \left(\sigma - \frac{0.72\Gamma}{d} \right) \left(1 + \frac{3.3WD_B}{dD_V} \right); \quad (4)$$

that is,

$$\dot{\epsilon}_T = \dot{\epsilon} + \dot{\epsilon}_{D-A}. \quad (5)$$

The definition of the symbols in Eq. 4 and their values are as follows: atomic volume of zirconium $\Omega = 2.37 \times 10^{-29} \text{ m}^3$; ¹⁴ volume diffusion coefficient* for zirconium $D_V = 5.9 \times 10^{-6} \exp(-Q_V/RT) \text{ m}^2/\text{s}$, where $Q_V = 218 \text{ kJ/mole}$; ¹¹ grain-boundary diffusion coefficient of α -zirconium $D_B = 5.9 \times 10^{-6} \exp(-Q_B/RT) \text{ m}^2/\text{s}$, where $Q_B = 0.60 Q_V$; grain size $d = (5 \text{ or } 11) \times 10^{-6} \text{ m}$; grain-boundary free energy $\Gamma = 0$ (an experimental value of Γ will be obtained in the future); and grain-boundary layer thickness $W = 1 \times 10^{-9} \text{ m}$. With regard to additional parameters in Eq. 1, the following values were used for both grain sizes: $n = 4.35$, $p = -1$, $Q = 316 \text{ kJ/mole}$, and $G = (1.94 \text{ and } 1.82) \times 10^{10} \text{ J/m}^3$ at 850 and 900°C, respectively. The constant A was adjusted so that the computed and measured yield stresses were the same at strain rates $\gtrsim 10^{-3} \text{ s}^{-1}$. (The strain-rate contribution $\dot{\epsilon}_{D-A}$ from Eq. 4 was negligible in this range.) The experimental data and calculated curves for the dependence of yield stress on the strain rate at 850 and 900°C are shown in Fig. III.9 for the 5- and 11- μm grain-size materials. The curves, based upon Eqs. 1, 4, and 5 and the values for the parameters listed above, are in good agreement with the experimental data at strain rates $\lesssim 10^{-3} \text{ s}^{-1}$, at which the transition to Stage II behavior occurs.

Fig. III.9

Comparison of the Calculated and Experimental Strain-rate Dependence of the 0.2% Yield Stress for 5- and 11- μm Grain-size Zircaloy-4 at 850 and 900°C. ANI. Neg. No. 306-76-253.

Although the Ashby-Verrall model⁷ was originally proposed for single-phase material, the information in Fig. III.9 and microstructural observations indicate that this model is also applicable for two-phase materials. Figure III.10 is a micrograph of the surface of a 5- μm specimen deformed at

Fig. III.10

Micrograph of the Surface of a 5- μm Grain-size Zircaloy-4 Specimen Deformed at a Strain Rate of $3.3 \times 10^{-4} \text{ s}^{-1}$ at 900°C That Shows Offsets of the Inscribed Scratches at the Grain Boundaries. Mag. $\sim 595\times$. Neg. No. MSD-63402.

*The diffusion coefficient of zirconium in α -zirconium and zirconium-tin alloys is sensitive to the dislocation density in the material.¹² The lower activation energy for zirconium diffusion¹¹ is more appropriate for a material undergoing plastic deformation.

a strain rate of $3 \times 10^{-4} \text{ s}^{-1}$ at 900°C (Stage II). Before the test, several light scratches were inscribed on the gauge-length region, perpendicular to the tensile axis. The micrograph shows that the grains do not elongate during deformation and that new grains (free of scratches) appear on the surface. In addition, a tendency for grain rotation and grain-boundary sliding is observed.

A microstructure that is characteristic of Stage III behavior, i.e., elongated grains that result from dislocation activity, is shown in Fig. III.11 for Zircaloy-4 deformed at a strain rate of $3 \times 10^{-1} \text{ s}^{-1}$ at 850°C . These observations, in addition to the large value of strain-rate sensitivity for the two-phase material ($m \approx 0.8$ at low strain rates) and the dependence of strain rate on grain size ($-3 < p < 0$), are consistent with the Ashby-Verrall model. The shift in the stress-versus-strain-rate curves, in Fig. III.9, to higher strain rates as the temperature increases is also predicted by the model.

Fig. III.11

Microstructure of an 11- μm Grain-size Specimen Deformed to 0.7% Strain at a Strain Rate of $3.0 \times 10^{-1} \text{ s}^{-1}$ at 850°C . Mag. $\sim 188\times$. Neg. No. MSD-63405.

C. Biaxial Deformation and Rupture Characteristics of As-received Zircaloy-4 Cladding In Vacuo

1. Rupture Characteristics of Unconstrained Cladding at a Low Heating Rate

The biaxial-deformation behavior of unconstrained Zircaloy-4 cladding has been determined in vacuo at a heating rate of 5°C/s. The circumferential strain at failure is plotted as a function of burst temperature in

Fig. III.12

Maximum Circumferential Strain vs Burst Temperature for Axially Unconstrained Zircaloy-4 Cladding In Vacuo at Heating Rates of 5 and 115°C/s. ANL Neg. No. 306-76-255.

Fig. III.12. Compared with the 115°C/s results, the burst temperature at the strain maximum is lower (810 versus 825°C) and the circumferential strain is higher (1.25 versus 1.5) for the 5°C/s heating rate. The temperature of the strain maxima at the lower heating rate corresponds to the α -phase boundary of Zircaloy-4 with ~0.1 wt % oxygen.³ The increase in the temperature of the strain maxima as the heating rate increases can be attributed to the kinetics of the $\alpha \rightarrow \beta$ phase transformation during the transient-heating experiments. The circumferential-strain data at the lower heating rate are in good agreement with the isothermal results reported by Busby and Marsh¹⁵ for recrystallized Zircaloy tubes.

Figure III.13 shows the axial contraction of the tubes as a function of burst temperature for both heating rates. The strain values represent the average axial strain in the central 75-mm-long, constant-temperature region in the tube. The axial contraction results from the anisotropy of the α -phase material during plastic deformation. The maxima in the axial and

circumferential strains occur at the same temperature for the respective heating rates. The axial strain is somewhat larger at the higher heating rate, because less time is available for recovery and accommodation in the material.

2. Effect of Axial Constraint on the Deformation Behavior of Zircaloy Cladding

Since the deformation behavior of constrained and unconstrained tubes is essentially the same up to the onset of plastic instability,⁴ the large difference in the circumferential strain at failure for temperatures $\leq 850^\circ\text{C}$

Fig. III.13

Average Axial Strain vs Burst Temperature for Unconstrained Zircaloy-4 Cladding In Vacuo at Heating Rates of 5 and 115°C/s. ANL Neg. No. 306-76-258.

occurs during the ballooning stage. To quantitatively relate the circumferential strain to the degree of constraint, an axial-constraint parameter γ was defined as³

$$\gamma = \frac{1}{\epsilon'_Z} \left(\frac{L}{L_0} - 1 \right), \quad (6)$$

where L_0 is the original length of the cladding, L is the length of the insert rod or fuel-pellet column, and ϵ'_Z is the axial strain at rupture for an unconstrained tube at a given heating rate and internal pressure, e.g., the data from the lower curves in Fig. III.13. Figure III.14 shows the diametral strain as a function of time for two 115°C/s burst tests with different axial-constraint parameters. The γ values of 0.2 and 0.49 correspond to axial gap distances of 2.5 and 5.9 mm, respectively. In both experiments, the contraction of the cladding corresponded to the axial gap distance.

Fig. III.14

Effect of the Degree of Axial Constraint on the Maximum Diametral Strain during the Ballooning Deformation of Zircaloy-4 Cladding In Vacuo at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-162.

Figure III.15 shows the maximum circumferential strain as a function of burst temperature for unconstrained tubes and tubes with axial gap distances of 2.5 and 5.9 mm at a heating rate of 115°C/s . Similar results for a heating rate of 5°C/s are shown in Fig. III.16. The alumina insert rod, in addition to establishing a known degree of constraint, will produce a small temperature gradient across the tube wall because the rod acts as a heat sink during transient heating of the tubes. At temperatures $<900^{\circ}\text{C}$, this effect will be small and, thus, the decrease in the circumferential strain and the shift in the strain maxima can be attributed to the degree of axial constraint.

Fig. III.15

Maximum Circumferential Strain vs Burst Temperature for Three Axial-constraint Conditions at a Heating Rate of 115°C/s In Vacuo. ANL Neg. No. 306-76-161.

Fig. III.16

Maximum Circumferential Strain vs Burst Temperature for Axially Constrained and Unconstrained Zircaloy-4 Cladding at a Heating Rate of 5°C/s In Vacuo. ANL Neg. No. 306-76-251.

The small strain maximum at $\sim 800^{\circ}\text{C}$ and the strain minimum at $\sim 815^{\circ}\text{C}$ for the 2.5-mm gap distance at the 115°C/s heating rate in Fig. III.15 was not observed at other heating rates and axial-constraint conditions in either vacuum or steam environments. Up to six thermocouples were spark-welded to each of the cladding tubes to minimize the uncertainty in the measurement of the burst temperatures. Results of isothermal burst tests that confirm this anomalous behavior will be discussed in Sec. D in conjunction with the effects of steam oxidation on the deformation and rupture characteristics.

D. Effect of Steam Oxidation on the Biaxial Deformation of Zircaloy-4 Cladding

1. Diametral Expansion Characteristics of Zircaloy-4 Cladding in Steam at Different Heating Rates

The relationships between maximum circumferential strain, initial pressure, and burst temperature for axially constrained (2.5-mm gap) Zircaloy-4 cladding in steam were reported in the previous quarterly report⁴ for heating rates of 5, 55, and 115°C/s. For burst temperatures predominantly in the β -phase region ($>950^\circ\text{C}$), oxidation of the tube wall resulted in a large decrease in the circumferential strain, compared with the values obtained in vacuo. For burst temperatures in the α -phase region, oxidation enhanced the circumferential strain at the three heating rates.

Figures III.17-III.19 show the diametral strain as a function of temperature for the axially constrained cladding at heating rates of 115, 55, and 5°C/s, respectively. The diametral strain at the onset of plastic instability and the circumferential strain at failure are also indicated on the figures. Compared with the results obtained in vacuo (Figs. III.17 and III.18 of Ref. 4), the extent of ballooning deformation after the onset of plastic instability decreases significantly at temperatures in the β -phase region.

Fig. III.17

Diametral Strain vs Temperature for Axially Constrained Zircaloy-4 Cladding at Different Internal Pressures at a Heating Rate of 115°C/s in Steam. ANL Neg. No. 306-76-246.

Fig. III.18

Diametral Strain vs Temperature for Axially Constrained Zircaloy-4 Cladding at Different Internal Pressures at a Heating Rate of 55°C/s in Steam. ANL Neg. No. 306-76-245.

Fig. III.19

Diametral Strain vs Temperature for Axially Constrained Zircaloy-4 Cladding at Different Internal Pressures at a Heating Rate of 5°C/s in Steam. ANL Neg. No. 306-76-252.

formation behavior of α -phase Zircaloy, a number of unconstrained tubes were ruptured in steam at a heating rate of 115°C/s. Figure III.20 indicates that the burst temperatures for constrained and unconstrained tubes in steam are the same but somewhat greater than in vacuo.

Fig. III.20

Burst Temperature as a Function of Initial Internal Pressure for Axially Constrained Zircaloy-4 Cladding in Steam and Vacuo and for Axially Unconstrained Cladding in Steam at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-127.

Although steam oxidation results in strengthening of the cladding, the maximum circumferential strain for unconstrained tubes in steam and vacuo (Fig. III.21) is the same for a heating rate of 115°C/s. This is in direct

The difference between the diametral and circumferential strains at failure for burst temperatures in the α -phase region results from a greater degree of asymmetric ballooning at the lower temperatures. A comparison of the diametral strain-time curves for similar burst temperatures at the same heating rate in steam and vacuo reveals that oxidation decreases the diametral strain at failure more than at instability.

2. Effect of Steam Oxidation on the Deformation Behavior of Constrained and Unconstrained Zircaloy-4 Cladding at a High Heating Rate

The maximum circumferential strain for axially constrained Zircaloy-4 cladding (Figs. III.28 and III.29 of Ref. 4), was greater in steam than in vacuo for burst temperatures in the α -phase region at heating rates of 5, 55, and 115°C/s. To determine the effect of axial constraint on the de-

Fig. III.21

Maximum Circumferential Strain vs Burst Temperature for Unconstrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-249.

contrast to behavior of axially constrained cladding at the high heating rate, in that oxidation resulted in a significant increase in the circumferential strain for burst temperatures between ~800 and 850°C.

Additional tube-rupture experiments on axially constrained cladding have been performed at the 115°C/s heating rate to confirm the observations that concern the effect of steam oxidation on the deformation behavior at temperatures in the α -phase region. To explore the possibility that thermal shunting and/or in situ oxidation of the thermocouples in steam can result in uncertainties in the burst-temperature measurements, specimens were pre-oxidized in steam under transient-heating conditions (i.e., 115°C/s of heating rate to ~800°C and rapidly cool to room temperature) and then burst in vacuo at the same heating rate. Furthermore, the Pt-Pt 10% Rh thermocouples were spark-welded to the specimens before and after the pre-oxidation treatment.

Figure III.22 shows the maximum circumferential strain for the preoxidized specimens burst in vacuo along with data for as-received specimens burst in vacuo and in steam. The results indicate that there is no difference in circumferential-strain characteristics for the specimens burst in the two environments at temperatures $\leq 800^\circ\text{C}$ and that the values for the pre-oxidized specimens ruptured in vacuo are in good agreement with the in situ burst data in steam at temperatures between ~780 and 830°C. These results indicate that errors in the temperature measurement due to thermal shunting and oxidation of the thermocouples are negligible.

Microstructural examinations revealed significant differences in the rupture region of specimens burst in steam and vacuo at temperatures between ~800 and 850°C. As discussed in Ref. 4, the fracture edge of the cladding burst in vacuo is sharp, whereas comparatively blunt edges are produced in steam. Figure III.23 shows typical cross sections of the failure location of tubes burst in vacuo and steam at temperatures between ~800 and 850°C. The large reduction in the wall thickness of the specimen burst in vacuo (Fig. III.23A) is characteristic of a rupture-type failure, whereas the fracture edge of the specimen burst in steam (Fig. III.23B) is similar to that produced by a shear process. Elongated as well as equiaxed (recrystallized) α -grains are evident in both specimens. Figure III.24 shows the corresponding scanning-electron

Fig. III.22

Maximum Circumferential Strain vs Burst Temperature for Axially Constrained As-received Cladding in Steam and Vacuo and Preoxidized Cladding Ruptured In Vacuo at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-264.

Fig. III.23. Cross Section of the Fracture Location of Axially Constrained Zircaloy-4 Cladding That Burst at 825°C In Vacuo (A) and in Steam (B) at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-239.

Fig. III.24. Scanning-electron Micrographs of the Fracture Edge of the Zircaloy-4 Cladding Shown in Fig. III.23 That Burst In Vacuo (A) and Steam (B) at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-238.

micrographs of the failure locations of the specimens burst in vacuo and steam. The single row of coalesced voids in Fig. III.24A is characteristic of a tensile failure, whereas typical shear dimples and ridges that result from the coalescence of microvoids are evident in Fig. III.24B. The shear direction is from the lower right to the upper left in the latter figure.

The photomicrographs in Figs. III.23 and III.24 suggest that the deformation-slip systems operative during the ballooning stage of tightly constrained cladding are different in the two environments at temperatures near 825°C. From an examination of the burst location on the tubes, the failure mode for the specimens in Fig. III.22 has been classified in terms of either a rupture- or fracture-type failure in Fig. III.25. Below ~800°C, fracture occurs by a shear process for the specimens burst in vacuo and steam. Fracture-type failures also occur in the pre-oxidized specimens burst in vacuo and for specimens burst in steam at temperatures up to ~830°C. Rupture failures occur in vacuo and steam above ~810 and 830°C, respectively.

In contrast to the behavior of constrained cladding, a fracture-type failure mode was observed for unconstrained tubing near 825°C (maximum circumferential strain 1.24) in both environments. These observations indicate that a fundamental transition in the deformation mechanism of α -phase Zircaloy occurs in the constrained cladding that is responsible for the enhanced ductility in steam at temperatures between ~800 and 850°C. The diametral strain-time curves for axially constrained cladding at a heating rate of 115°C/s in Figs. III.26 and III.27 indicate that this transition occurs after the onset of plastic instability in the material just before rupture. As discussed in Ref. 4 ($\vec{c} + \vec{a}$) pyramidal slip is suppressed as a result of oxidation in the steam environment and localized thinning of the tube wall becomes more difficult; this leads to bending of the tube and a larger circumferential strain at failure.

3. Effect of Steam Oxidation on the Deformation Behavior of Constrained and Unconstrained Cladding at a Low Heating Rate

Relationships between the initial pressure, maximum circumferential strain, and burst temperature for axially constrained Zircaloy-4 cladding in steam for a heating rate of 5°C/s were given in Figs. III.27, III.29, and III.32 of Ref. 4. Similar data for unconstrained cladding in vacuo and steam are shown in Figs. III.28 and III.29. The burst temperatures for unconstrained

Fig. III.25

"Rupture" and "Fracture" Failure-mode Classifications for the Deformation Data in Fig. III.22 on Axially Constrained Zircaloy-4 Cladding. ANL Neg. No. 306-76-261.

Fig. III.26

Maximum Diametral Strain as a Function of Time before Rupture for Axially Constrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-262.

Fig. III.27

Maximum Diametral Strain as a Function of Time before Rupture for Axially Constrained As-received Zircaloy-4 Cladding in Steam and Vacuo and Preoxidized Cladding In Vacuo at a Heating Rate of 115°C/s. ANL Neg. No. 306-76-244.

Fig. III.28

Burst Temperature as a Function of Initial Internal Pressure for Axially Constrained and Unconstrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 5°C/s. ANL Neg. No. 306-76-257.

Fig. III.29

Maximum Circumferential Strain vs Burst Temperature for Axially Unconstrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 5°C/s. ANL Neg. No. 306-76-259.

cladding in Fig. III.29 are somewhat lower than those for the constrained case in the respective environments. Also, the burst temperature in steam is lower than that in vacuo for temperatures below ~810°C. If the strengthening effect due to oxygen dissolution into the α -phase material during the low-heating-rate tests was the only factor, the burst temperature in steam would be higher than in vacuo for a given initial pressure. Apparently, the additional tangential stress introduced by the oxide film more than compensates for the solute strengthening of the underlying metal and decreases the rupture temperature.

Similar to the behavior of the constrained cladding at the 5°C/s heating rate, the circumferential expansion of the unconstrained tubes, shown in Fig. III.29, is greater in steam than in vacuo at temperatures between 700 and 800°C. This is in contrast to the results at a heating rate of 115°C/s, in which only the constrained cladding (2.5-mm gap) exhibited larger circumferential strains in the steam environment.

The diametral-strain-versus-time curves in Fig. III.30 for two runs in steam and one in vacuo, with approximately the same burst temperature, show that the diametral strains are nearly the same for the initial stages of deformation and that the enhanced ballooning in steam occurs immediately before rupture. The additional tangential stress introduced by the oxide layer can decrease the burst temperature, as mentioned above, and increase the diametral strain as a result of a change in the biaxiality.³ During simultaneous oxidation and deformation, the tangential stress σ_θ can be written as

$$\sigma_\theta(t) = \frac{P(t)D(t)}{2h(t)} + \sigma_s(t), \quad (7)$$

where P , D , h , and σ_s are the time-dependent pressure differential across the tube wall, tube diameter, tube thickness, and stress induced by the oxide layer,

respectively. The biaxiality $b = \sigma_{\theta}(t)/\sigma_z(t)$, where $\sigma_z(t)$ is the axial stress component, can be written as

$$b = \frac{P(t)D(t)}{2h(t)\sigma_z(t)} + \frac{\sigma_s(t)}{\sigma_z(t)} = b_V(t) + \frac{\sigma_s(t)}{\sigma_z(t)}, \quad (8)$$

where $b_V(t)$ is the biaxiality condition in a vacuum environment.

Fig. III.30

Maximum Diametral Strain as a Function of Time before Rupture for Axially Constrained Zircaloy-4 Cladding in Steam and Vacuo at a Heating Rate of 5°C/s. ANL Neg. No. 306-76-263.

The variation of the biaxiality and tangential strain with time for constrained and unconstrained tubes in vacuo and steam is shown schematically in Fig. III.31. The effect of axial constraint on these parameters was discussed in Ref. 3 for tubes that were burst in vacuo. In this case, the larger tangential strain for an unconstrained tube was associated with a smaller decrease in the biaxiality after the onset of plastic instability in the material. A similar situation exists for deformation in steam; however, an increase in biaxiality (i.e., $b > 2$), as a result of the oxidation, occurs before the onset of plastic instability; consequently, the biaxiality and the tangential strain at rupture are larger in steam than in vacuo.

Fig. III.31

Schematic Representation of the Effect of Axial Constraint on the Biaxiality and Tangential Strain in the Plastic-instability Region of a Zircaloy Tube Burst in Vacuum and Steam Environments. ANL Neg. No. 306-76-260.

The stress induced by the oxide layer σ_s can be estimated from the expression given by Southwell¹⁶

$$\sigma_s = \frac{E(R - 1)\zeta_0}{RD}, \quad (9)$$

where E , R , and ζ_0 are Young's modulus, the Pilling-Bedworth ratio, and the thickness of the oxide layer, respectively. The tangential stress that results from the oxide layer produced on a tube that ruptured at 760°C in a 5°C/s heating-rate experiment was computed from Eq. 9. A value of 4.9 MPa was obtained from the measured oxide-layer thickness of 1.05×10^{-3} mm, a tube diameter of 13.4 mm at the onset of plastic instability, and a Pilling-Bedworth ratio of 1.56. A value of Young's modulus of 1.8×10^5 MPa for an oxide layer of zero porosity at 700°C, obtained from the data of Smith and Crandall,¹⁷ was used. The axial and tangential stresses due to an internal pressure of 5.6 MPa

at the onset of instability are ~46.5 and 93 MPa, respectively, for the unconstrained tube. Therefore, the biaxiality at the onset of instability increases ~5% (i.e., from 2.0 to 2.1) as a result of oxidation under the above conditions.

Fig. III.32

Average Axial Strain vs Burst Temperature for Unconstrained Zircaloy-4 Cladding In Vacuo and Steam at a Heating Rate of 5°C/s. ANL Neg. No. 306-76-247.

Figure III.32 compares the axial strain as a function of burst temperature for the unconstrained cladding in steam and vacuo at a heating rate of 5°C/s. As with the experiments at higher heating rates, maximum axial contraction is coincident with the circumferential strain maximum in both environments.

4. Stress-rupture Characteristics of Zircaloy-4 Cladding in Steam and Vacuum Environments

A limited number of isothermal stress-rupture experiments have been conducted on as-received and preoxidized Zircaloy-4 cladding in vacuo and steam to verify the observations, in Figs. III.22 and III.29, that concern enhanced circumferential strain during transient-heating experiments in steam. Four thermocouples (one control and three monitoring) were

spark-welded to thin tantalum foils (0.178 mm thick) that had been spot-welded to the cladding tube. The thermocouple locations did not result in preferential burst sites. The specimens were heated to the test temperature in vacuo,

steam was admitted to the chamber and the temperature readjusted, and the specimen was rapidly pressurized by opening a needle valve. For the rupture tests in vacuo, the specimens were pressurized after the test temperature was attained. The pressure and temperature were recorded by the Visicorder, and the diametral strain was monitored by the high-speed camera and laser-light system used in the transient-heating experiments.

Figure III.33 shows the diametral strain as a function of time for an unconstrained as-received and a preoxidized (100°C/s in steam to 800°C and quench) specimen ruptured in vacuo at 750°C at an internal pressure of 5.2 MPa. The calculated oxygen distribution in the ZrO_2/α composite specimen is shown in the inset of the figure. The larger diametral strain associated with the preoxidized specimen is attributed to the increase in the biaxiality of the specimen due to the oxide layer.

Fig. III.33

Maximum Diametral Strain as a Function of Time in Isothermal Stress-rupture Tests on Unconstrained As-received and Preoxidized Zircaloy-4 Cladding In Vacuo at 750°C and an Internal Pressure of 5.2 MPa. ANL Neg. No. 306-76-241.

Figure III.34 shows the diametral and axial strains and the localized ballooning characteristics of another preoxidized specimen ruptured in vacuo under the same conditions. Figure III.35 shows similar results for an as-received and a preoxidized specimen ruptured in vacuo at 750°C and an internal pressure of 4.2 MPa along with a strain-time curve for an as-received specimen burst in steam. The larger degree of oxidation and oxygen dissolution in the α -phase Zircaloy during the isothermal test in steam resulted in a smaller strain at rupture compared with that of either the as-received or the preoxidized specimen tested in vacuo. This result indicates that, under these conditions, oxygen dissolution in the material had a greater effect on the ductility than the increase in biaxiality due to the oxide layer.

Several stress-rupture tests were performed on constrained as-received cladding in steam and vacuo at temperatures between ~ 720 and 840°C . The internal pressure in these tests was adjusted such that the deformation time (viz, 5-8 s) was much shorter than at the 4.2- and 5.2-MPa stress levels at 750°C described above. Consequently, as shown in Fig. III.36, the circumferential-strain data are in good agreement with the results from the 115°C/s heating-rate experiments on constrained cladding in steam and vacuo (i.e., Fig. III.22). Figure III.37 shows photographs of the as-received and

preoxidized unconstrained specimens ruptured in vacuum at 750°C (i.e., those represented in Fig. III.35) and the as-received constrained specimens ruptured in steam and vacuo at 820°C (i.e., those represented in Fig. III.36). The degree of oxidation and the strain rate in the two sets of experiments illustrate the effects of increased biaxiality (Fig. III.37A) and pyramidal-slip suppression (Fig. III.37B) on circumferential-strain enhancement in the steam environment.

CAMERA SPEED 20 FRAMES/s
FRAMES BEFORE BURST 4460, 66, 20, 8, 2, BURST

OD SURFACE PREOXIDIZED

Fig. III.34

Localized Ballooning Characteristics and Diametral and Axial Strains during an Isothermal Stress-rupture Test on Preoxidized Zircaloy-4 Cladding In Vacuo at 751°C and an Internal Pressure of 5.2 MPa. ANL Neg. No. 306-76-254.

Fig. III.35

Maximum Diametral Strain as a Function of Time in Isothermal Stress-rupture Tests on Unconstrained As-received Zircaloy-4 Cladding in Steam and Vacuo and Preoxidized Cladding In Vacuo at 750°C and an Internal Pressure of 4.2 MPa. ANL Neg. No. 306-76-237.

Fig. III.36

Comparison of Maximum Circumferential Strain vs Burst-temperature Data Obtained from Short-term Isothermal Stress-rupture Tests on Constrained Zircaloy-4 Cladding in Steam and Vacuo with Similar Data (i.e., the curves) from Transient-heating Experiments at 115°C/s in the Two Environments. ANL Neg. No. 306-76-243.

Fig. III.37

Photographs of Zircaloy-4 Cladding from the Isothermal Stress-rupture Tests. (A) Unconstrained preoxidized ZrO_2/α composite and as-received cladding ruptured in vacuo at 750°C at an internal pressure of 4.2 MPa. (B) Axially constrained as-received cladding ruptured in vacuo and steam at 825°C at an internal pressure of 5.2 MPa. ANL Neg. No. 306-76-240.

References

1. Light-water-reactor Safety Research Program: Quarterly Progress Report, July-September 1975, Sec. III, "Mechanical Properties of Zircaloy Containing Oxygen," ANL-75-72.
2. Light-water-reactor Safety Research Program: Quarterly Progress Report, October-December 1975, Sec. III, "Mechanical Properties of Zircaloy Containing Oxygen," ANL-76-15.
3. Light-water-reactor Safety Research Program: Quarterly Progress Report, January-March 1976, Sec. III, "Mechanical Properties of Zircaloy Containing Oxygen," ANL-76-49.
4. Light-water-reactor Safety Research Program: Quarterly Progress Report, April-June 1976, Sec. III, "Mechanical Properties of Zircaloy Containing Oxygen," ANL-76-87.
5. R. H. Chapman, Compiler, Characterization of Zircaloy-4 Tubing Procured for Fuel Cladding Research Programs, ORNL/NUREG/TM-29 (July 1976).
6. R. C. Gifkins, Grain-boundary Sliding and Its Accommodation during Creep and Superplasticity, Met. Trans. 7A, 1225 (1976).
7. M. F. Ashby and R. A. Verrall, Diffusional-accommodated Flow and Superplasticity, Acta Met. 21, 149 (1973).
8. R. F. S. Hearmon, The Elastic Constants of Anisotropic Materials, Adv. Phys. 5, 323 (1956).
9. E. S. Fisher and C. J. Renken, Single Crystal Elastic Moduli and the hcp→bcc Transformation in Ti, Zr, and Hf, Phys. Rev. 135, A482 (1964).
10. J. J. Kearns, J. E. McCauley, and F. A. Nichols, Effect of Alpha/Beta Phase Constitution on Superplasticity and Strength of Zircaloy-4, J. Nucl. Mater. 61, 169 (1976).
11. V. S. Lyashenko, V. N. Bykov, and L. V. Pavlinov, A Study of Self-Diffusion in Zirconium and Its Alloys with Tin, Fiz. Metall. Metalloved. 8, 362 (1959), English Translation 8, 40 (1960).
12. G. M. Hood and R. J. Schultz, Tracer Diffusion in α -Zr, Acta Met. 22, 459 (1974).
13. M. C. Naik and R. P. Agarwala, Self- and Impurity Diffusion in Alpha-Zirconium, Acta Met. 15, 1521 (1971).
14. L. T. Lloyd, Thermal Expansion of Alpha-Zirconium Single Crystals, ANL-6591 (Jan 1963).
15. C. C. Busby and K. B. Marsh, High Temperature Deformation and Burst Characteristics of Recrystallized Zircaloy-4 Tubing, WAPD-TM-900, Bettis Atomic Power Laboratory, Westinghouse Electric Corp., Pittsburgh, Pa. (Jan 1970).

16. R. V. Southwell, An Introduction to the Theory of Elasticity for Engineers and Physicists, Clarendon Press, Oxford, p. 406 (1936).
17. C. F. Smith and W. B. Crandall, Calculated High Temperature Elastic Constants for Zero Porosity Monoclinic Zirconia, J. Am. Ceram. Soc. 47, 624 (1964).

IV. STEAM-EXPLOSION STUDIES

Responsible Section Manager:

R. E. Henry, RAS

A. Vapor Explosions at Elevated Pressures (R. E. Henry and L. M. McUmbert, RAS)

Analyses performed on the energetics of vapor explosions have indicated that the bubble-growth characteristics during the vaporization process play a significant role in determining the progression of the explosive interaction. Details of bubble-growth characteristics, which can be obtained in Ref. 1, show that many considerations must be made in understanding the details relating to the formation and growth of a vapor space. However, from a first-order point of view, two basic regimes are of major interest. These are the inertially dominated growth regime and the thermally dominated regime.

Figure IV.1 is a simplified illustration of these bubble-growth behaviors in terms of the bubble radius first time. In the inertially limited behavior, the pressure inside the bubble is higher than that of the surrounding liquid and the temperature within the bubble is essentially equal to that of the liquid. Therefore, the growth of the vapor cavity is limited by the rate at which it can move liquid away, i.e., the inertia of the liquid. Under these conditions, the radius r of the spherical bubble increases linearly in time t and can be represented by

$$r = \sqrt{\frac{2}{3} \frac{P_v - P_l}{\rho_l} t}, \quad (1)$$

Fig. IV.1. Inertially and Thermally Dominated Bubble-growth Behavior. ANL Neg. No. 900-5175.

where P_v is the vapor pressure corresponding to the liquid temperature, P_ℓ is the liquid pressure far removed from the bubble, and ρ_ℓ is the liquid density.

The thermally dominated regime is characterized by the liquid and vapor having the same pressure but different temperatures. In this case, the growth is regulated by the rate at which energy can be supplied to the liquid-vapor interface via thermal conduction from the liquid surrounding the bubble. Under these conditions, the radius of the vapor bubble increases as a square root of time and can be approximated by

$$r = 2(T_\ell - T_{SAT}) \frac{\rho_\ell c_\ell}{\rho_v H_{fg}} \sqrt{\alpha_\ell t}, \quad (2)$$

where T_ℓ is the liquid temperature far removed from the bubble, T_{SAT} is the saturation temperature corresponding to the pressure, c_ℓ is the liquid specific heat, ρ_v is the vapor density, H_{fg} is the latent heat of vaporization, and α_ℓ is the liquid thermal diffusivity.

The model presented in Ref. 2 suggests that explosive interactions themselves should terminate in the regime where the bubble growth is dominated by thermal considerations beginning at the critical size. The critical size is determined by a force balance between surface tension and pressure difference between the inside and outside of a spherical bubble:

$$P_v - P_\ell = \frac{2\sigma}{r}. \quad (3)$$

This indeed seems to be the case based on past experiments in which the maximum interaction pressures were measured. Experiments reported in Refs. 3 and 4 show that, with Freon and mineral oil and Freon and water, the maximum interaction pressure is 22-25 atm with a rise time of about 1 ms. This maximum pressure represents a reduced pressure of about 0.6 for the exploding liquid. If it represents a valid correlating parameter, this is an extremely important finding for light-water reactor safety when considering the failure of fuel at nominal operating pressures that correspond to reduced pressure of about 0.66. Consequently, explosions under these conditions in pressurized, light-water reactors may be impossible to achieve. To investigate this phenomena and the role that the initial environmental pressure has upon the onset of explosive interactions, a series of experiments was carried out for Freon 22/water and Freon 22/mineral-oil systems within a contained geometry.

Separate systems were constructed to investigate Freon/mineral-oil and Freon/water explosive interactions under elevated ambient pressures. This was done to simplify the progression of the experiments as well as to prevent system contamination that would result in switching between water and mineral oil as the hot liquids in the same system. Figure IV.2 illustrates the system configuration for the experiments. The vessels were fabricated from 10- and 15-cm pipes with electrically heated, insulated cups suspended

Fig. IV.2. High-pressure Apparatus. ANL Neg. No. 900-76-571.

were mounted in this same locale. The vacuum line was outfitted with a solenoid valve, which was energized simultaneously with the diaphragm-punch transport cylinder to isolate the vacuum system once the test was initiated.

To determine that the system configuration itself did not affect the explosive characteristics of the given fluids, the initial set of tests for both systems were run at an ambient pressure of 1 atm with argon as the cover gas. The results are illustrated in Figs. IV.3 and IV.4 for the Freon/oil and Freon/water systems, respectively. These results are in excellent agreement with the previously published data of Refs. 3 and 4, the maximum interaction pressure being 23 atm, which is close to the value deduced from the bubble-growth considerations. Consequently, the apparatus itself did not affect the explosive behavior of the liquid pairs.

Following the 1-atm ambient-pressure tests, similar experiments were conducted at ambient pressures of 2.2 and 8 atm, and as shown in Figs. IV.3 and IV.4, no explosive interactions were observed in either liquid

from the upper flange. Thermocouples were provided within this cup to measure the liquid level before the test as well as the initial fluid temperature, and the solenoid-punch system was used to rupture the diaphragm at the base of the cup and initiate the test. The lower portion of the vessel in which the second fluid was maintained was fitted with thermocouples to measure the temperature distribution within the bath and high-response, piezoelectric pressure and force transducers to monitor the interaction behavior of the two liquids. This portion of the vessel was also outfitted with trace heaters and insulation for control of the liquid temperature.

In addition, the gas space above the interaction zone was equipped with a pressure transducer to monitor the overall system pressure, and a rupture disk and vacuum line

pair at these elevated ambient pressures. These experimental results not only demonstrate that the maximum explosive interaction pressure is limited by bubble-growth considerations, but more importantly, that explosive interactions cannot be initiated at elevated ambient pressures. Also, these ambient pressures that prohibit the initiation of an explosive event are considerably less than the pressure at which the bubble growth is thermally dominated from its critical size.

Fig. IV.3. High-ambient-pressure Freon/Mineral-oil Data. ANL Neg. No. 900-76-826.

Fig. IV.4. High-ambient-pressure Freon/Water Data. ANL Neg. No. 900-76-824.

B. Combined Physical-Chemical Explosions (R. P. Anderson, D. R. Armstrong, and N. E. Parker, RAS)

The occurrence of combined physical-chemical explosions has been reported for aluminum-water systems. The criteria for the initiation of the chemical portion of these explosions and its contribution to the energetics of the interaction are unknown and the subject of this program.

The first requirement for the program is a method of generating reproducible explosions in a system capable of recording temperature, pressure, and velocity histories. Early investigators were unable to produce aluminum-water explosions with less than 20 kg of aluminum, except in small shock tubes.

The difficulty in generating explosions in small-scale systems was tentatively assumed to be caused by a stable vapor film separating the two liquids. The ease with which explosions have been produced in large-scale

experiments was attributed to the collapse of this vapor film in the presence of large-scale mixing forces. An experimental program, set up on the basis of this physical picture, attempted to generate explosions in small-scale systems by penetrating or collapsing this vapor film. Experiments using the penetration technique were described in earlier reports.^{5,6} Current effort has been directed toward two methods of collapsing the vapor film.

Fig. IV.5. Impact Apparatus. ANL Neg. No. 900-76-76.

ciencies for the two explosions are 5.2 and 8.2%, respectively, based on the maximum work that could be produced in the system by a constant-temperature expansion of the water at the aluminum temperature.⁶

During the analysis of the above experiments a discrepancy was noted between the calculated impact impulse and that measured by the force transducer. A series of runs was made impacting the weighted cylinder on various objects placed on the force transducer. Although this discrepancy has not yet been resolved, some interesting information has resulted from these runs. The molten-aluminum system is much softer than the water system. The aluminum impulse shows a lower peak and longer duration. The peak force measured for the impact on molten aluminum was about one-third of that for a water-filled crucible. It is reasonable to assume that the reduction in peak pressure is of the same order, so that the impact experiments described in Ref. 7 did not produce pressures of 25 MPa, as reported, but only about 8 MPa,

The first method uses the apparatus shown in Fig. IV.5 to inject a volume of water into the aluminum, then compress the system with a dropping steel cylinder, which impacts on a graphite toroid immediately above the molten aluminum. This apparatus has been modified by placing an exploding wire inside the water-filled glass sphere. The wire is triggered by a signal from a force transducer beneath the aluminum so that the dispersion occurs while the system is compressed. This procedure has produced an explosion.

Two runs have been made. Both used 1 g of water at 20°C and 900 g of aluminum, one test at 780°C, the other at 880°C. The measured energy output was 125 J for the 880°C run and 181 J for the 780°C run. These measurements were based solely on the motion of the piston and do not include the energy of the aluminum dispersed by the explosion. Relative effi-

well below the critical pressure of water. During the next quarter, impact tests will be run with a time delay between exploding the wire and the piston impact.

The second method of collapsing the vapor film is shown in Fig. IV.6. This is a modification of the experiment (described in Ref. 7) that produced

Fig. IV.6. Explosion-initiated Apparatus. ANL Neg. No. 900-76-849.

a vapor explosion. A graphite crucible with a plug valve in the bottom was used to melt and drop the aluminum. This gave a more reproducible geometry than the pouring method used earlier. The water tank was a Lucite cylinder with 1.2-cm-thick walls and inside dimensions of 10-cm depth and 7.5-cm diameter. A pressure transducer was flush-mounted in the wall. One experiment was run with this equipment; 28 g of 800°C aluminum was dropped into 250 g of 20°C water and subjected to a wire explosion. The resulting vapor explosion shattered the water tank and broke the graphite crucible and heater. No pressure data were obtained because the electrical noise from the exploding wire turned off the instrument recorder. Motion pictures of the event showed that, within one frame (2500 frames per second) of the wire explosion, the water tank was cracked and the aluminum and water were dispersing rapidly.

A stronger water tank and heater are being fabricated, and work is progressing to control the electrical noise associated with the exploding wire. This experiment should become operational during the next quarter.

References

1. T. Theofanous, L. Biasi, H. S. Isbin, and H. K. Fauske, Theoretical Study of Bubble Growth in Constant and Time-Dependent Pressure Fields, Chem. Engr. Sci. 24, 885 (1969).
2. R. E. Henry and H. K. Fauske, "Nucleation Characteristics and Physical Explosions," Proc. Third Specialists Meet. Sodium/Fuel Interaction in Fast Reactors, Tokyo, Japan, PNC N251 76-12 (1976).
3. R. E. Henry et al., "Large Scale Vapor Explosions," Proc. Fast Reactor Safety Meet., Beverly Hills, CA., CONF-740401-P2, 922 (Apr 1974).
4. R. E. Henry, H. K. Fauske, and L. M. McUmber, "Vapor Explosions with Subcooled Freon," Proc. Third Specialists Meet. Sodium/Fuel Interaction in Fast Reactors, Tokyo, Japan, PNC N251 76-12 (1976).
5. Light-water-reactor Safety Research Program: Quarterly Progress Report, October-December 1975, ANL-76-15, p. 1.
6. Light-water-reactor Safety Research Program: Quarterly Progress Report, January-March 1976, ANL-76-49, p. 60.
7. Light-water-reactor Safety Research Program: Quarterly Progress Report, April-June 1976, ANL-76-87, p. 62.