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

April—June 1977

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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ARGONNE NATIONAL LABORATORY
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April-June 1977

Robert G. Sachs, Laboratory Director
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Previous reports in this series:

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ABSTRACT

This progress report summarizes the Argonne National Laboratory work performed during April, May, and June 1977 on water-reactor-safety 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:

H. K. Fauske, R. E. Henry, and P. A. Lottes, RAS

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

1. Objective

A crucial phenomenon to be modeled in the analysis of a hypothetical loss-of-coolant accident (LOCA) in a light-water reactor is the time at which efficient cooling ceases on the fuel-rod surface. This loss of cooling during depressurization has been referred to in the literature as a transient critical-heat-flux (CHF) condition. In a pressurized water reactor, the case of principal interest is an instantaneous guillotine rupture of an inlet coolant pipe, which not only results in depressurization of the system, but also causes the flow through the core to change from vertical upflow to downflow. Numerous experiments have indicated that CHF does not occur as flow reverses, but is delayed for some time. Several seconds delay were observed in the early semiscale results,¹ B&W tests,² and the FCTF tests at Oak Ridge.³

Quite the contrary, recent semiscale experiments^{4,5} and the THTF facility at Oak Ridge⁶ have experienced very early CHF, less than 800 ms. This spectrum of experimental results warrants more basic study of the mechanisms, and this is the objective of the present study. Without understanding and discerning among the basic mechanisms, it is extremely difficult to predict the performance of the full-size reactor systems.

2. Status Summary

a. Previous Work

Freon blowdown experiments performed in a single heated tube (shown in Figs. I.1 and I.2) have successfully demonstrated the occurrence of very rapid CHF,^{7,8} as observed in some water experiments. These tests also show the strong influence of initial steady-state pressure on the early CHF zone. By lowering the initial pressure while maintaining other parameters the same, we extend the subcooled boiling region, but the bulk-temperature profile remained unchanged for a subcooled outlet. Results indicated more cooling in the low-pressure tests, with the shorter region in the bottom zone of the test section experiencing early CHF during depressurization. These observed phenomena were explained to be related to the nucleation characteristics at the heater surface during steady-state operation and during the blowdown transient.

SCHEMATIC OF TEST SECTION II

Fig. I.1. Schematic of Test Section II. Conversion factor:
1 in. = 2.54 cm. ANL Neg. No. 900-76-699.

Fig. I.2

Test Section II Power Profile.

ANL Neg. No. 900-77-149.

b. Current Work

During this quarter, about 20 tests were conducted at break diameters ranging from 0.25 to 0.61 cm, and power input up to 7.1 kW total. Results of 11 tests are reported here; data reduction are still underway with other tests. Initial conditions of these tests are given in Table I.1; other important parameters and blowdown results are listed in Table I.2. Figures I.3-I.13 illustrate the steady-state wall and bulk-fluid temperature. Also shown are the regions of the heated section that experienced early CHF (less than 1 s). Transient data are presented in Figs. I.14-I.79. Location of instrumentation and its symbol used in the figures are given in Table I.3.

TABLE I.1. Steady-state Test Conditions in Test Section II

Test No.	Break-orifice Diameter, cm	Pressure, MPa	Fluid Temperature, °C		Inlet Velocity, m/s	Mass Flow Rate, kg/m ² ·s	Total Power, kW
			Inlet	Outlet			
DB-120	0.508	2.24	115	150	1.50	1831	7.13
DB-121	0.508	2.81	116	151	1.54	1871	7.13
DB-126	0.508	2.23	114	148	0.54	670	2.70
DB-127	0.508	2.81	114	149	0.53	660	2.70
DB-128	0.508	2.19	114	147	0.88	1070	4.02
DB-130	0.381	2.23	115	149	0.84	1028	4.03
DB-131	0.381	2.23	114	148	0.99	1211	4.59
DB-132	0.381	2.76	114	148	0.97	1189	4.60
DB-133	0.381	2.76	114	148	0.85	1166	3.98
DB-134	0.254	2.21	114	149	0.99	1215	4.60
DB-135	0.254	2.21	112	146	0.83	1020	3.92

TABLE I.2. Other Blowdown Parameters

Test No.	Volume above Heated Zone, cm ³	Temperature-trip Setting, °C	Time at Power Trip, s	Location of Overheating Thermocouple, cm	Region in Early CHF, cm
DB-120	2620	221	4.8	48	130
DB-121	2620	221	3.4	168	191
DB-126	2620	221	17.6	48	Nil
DB-127	2620	221	17.1	48	Nil
DB-128	2620	221	16.2	48	Nil
DB-130	2620	221	24.0	48	Nil
DB-131	2620	221	12.0	48	Nil
DB-132	2620	221	13.0	48	Nil
DB-133	2620	221	22.4	48	Nil
DB-134	2620	221	7.9	48	38
DB-135	2620	221	9.9	48	38

Fig. I.3. Steady-state Wall-temperature Profile in Test DB-120.
ANL Neg. No. 900-77-1341.

Fig. I.4. Steady-state Wall-temperature Profile in Test DB-121.
ANL Neg. No. 900-77-1339.

Fig. I.5. Steady-state Wall-temperature Profile in Test DB-126.
ANL Neg. No. 900-77-1338.

Fig. I.6. Steady-state Wall-temperature Profile in Test DB-127.
ANL Neg. No. 900-77-1349.

Fig. I.7. Steady-state Wall-temperature Profile in Test DB-128.
ANL Neg. No. 900-77-1343.

Fig. I.8. Steady-state Wall-temperature Profile in Test DB-130.
ANL Neg. No. 900-77-1348.

Fig. I.9. Steady-state Wall-temperature Profile in Test DB-131.
ANL Neg. No. 900-77-1344.

Fig. I.10. Steady-state Wall-temperature Profile in Test DB-132.
ANL Neg. No. 900-77-1342.

Fig. I.11. Steady-state Wall-temperature Profile in Test DB-133.
ANL Neg. No. 900-77-1340.

Fig. I.12. Steady-state Wall-temperature Profile in Test DB-134.
ANL Neg. No. 900-77-1345.

Fig. I.13. Steady-state Wall-temperature Profile in Test DB-135.
ANL Neg. No. 900-77-1347.

Fig. I.14. System Pressure during Test DB-120.
 ANL Neg. No. 900-77-1359.

Fig. I.15. Differential Pressure across Test Section during
 Test DB-120. ANL Neg. No. 900-77-1363.

Fig. I.16. Differential Pressure across Break Orifice during Test DB-120. ANL Neg. No. 900-77-1358

Fig. I.17. Turbine-flowmeter Measurement during Test DB-120. ANL Neg. No. 900-77-1355.

Fig. I.18. Fluid Temperature during Test DB-120.
ANL Neg. No. 900-77-1356.

Fig. I.19. Wall-thermocouple Measurement during Test DB-120.
ANL Neg. Nos. 900-77-1351, -1325, -1362, and -1357.

Fig. I.19 (Contd.)

Fig. I.19 (Contd.)

Fig. I.19 (Contd.)

Fig. I.20. System Pressure during Test DB-121.
ANL Neg. No. 900-77-1303.

Fig. I.21. Differential Pressure across Test Section during Test DB-121. ANL Neg. No. 900-77-1319.

Fig. I.22. Differential Pressure across Break Orifice during Test DB-121. ANL Neg. No. 900-77-1314.

Fig. I.23. Turbine-flowmeter Measurement during Test DB-121.
ANL Neg. No. 900-77-1317.

Fig. I.24. Fluid Temperature during Test DB-121.
ANL Neg. No. 900-77-1322.

Fig. I.25. Wall-thermocouple Measurement during Test DB-121.
ANL Neg. Nos. 900-77-1321, -1302, -1311, and -1307.

Fig. I.25 (Contd.)

Fig. I.25 (Contd.)

Fig. I.25 (Contd.)

Fig. I.26. System Pressure during Test DB-126.
 ANL Neg. No. 900-77-1323.

Fig. I.27. Differential Pressure across Test Section during
 Test DB-126. ANL Neg. No. 900-77-1367.

Fig. I.28. Differential Pressure across Break Orifice during Test DB-126. ANL Neg. No. 900-77-1365.

Fig. I.29. Turbine-flowmeter Measurement during Test DB-126. ANL Neg. No. 900-77-1366.

Fig. I.30. Fluid Temperature during Test DB-126.
ANL Neg. No. 900-77-1352.

Fig. I.31. Wall-thermocouple Measurement during Test DB-126.
ANL Neg. Nos. 900-77-1328, -1350, -1360, and -1330.

Fig. I.31 (Contd.)

Fig. I.31 (Contd.)

Fig. I.31 (Contd.)

Fig. I.32. System Pressure during Test DB-127.
ANL Neg. No. 900-77-1336.

Fig. I.33. Differential Pressure across Test Section during Test DB-127. ANL Neg. No. 900-77-1337.

Fig. I.34. Differential Pressure across Break Orifice during Test DB-127. ANL Neg. No. 900-77-1312.

Fig. I.35. Turbine-flowmeter Measurement during Test DB-127.
 ANL Neg. No. 900-77-1310.

Fig. I.36. Fluid Temperature during Test DB-127.
 ANL Neg. No. 900-77-1353.

Fig. I.37. Wall-thermocouple Measurements during Test DB-127.
ANL Neg. Nos. 900-77-1327, -1361, -1308, and -1326.

Fig. I.37 (Contd.)

Fig. I.37 (Contd.)

Fig. I.37 (Contd.)

Fig. I.38. System Pressure during Test DB-128.
 ANL Neg. No. 900-77-1309.

Fig. I.39. Differential Pressure across Test Section during
 Test DB-128. ANL Neg. No. 900-77-1332.

Fig. I.40. Differential Pressure across Break Orifice during Test DB-128. ANL Neg. No. 900-77-1334.

Fig. I.41. Turbine-flowmeter Measurement during Test DB-128. ANL Neg. No. 900-77-1335.

Fig. I.42. Fluid Temperature during Test DB-128.
ANL Neg. No. 900-77-1329.

Fig. I.43. Wall-thermocouple Measurement during Test DB-128.
ANL Neg. Nos. 900-77-1320, -1318, -1316, and -1313.

Fig. I.43 (Contd.)

Fig. I.43 (Contd.)

Fig. I.43 (Contd.)

Fig. I.44. System Pressure during Test DB-130.
ANL Neg. No. 900-77-1409.

Fig. I.45. Differential Pressure across Test Section during Test DB-130. ANL Neg. No. 90C-77-1374.

Fig. I.46. Differential Pressure across Break Orifice during Test DB-130. ANL Neg. No. 900-77-1403.

Fig. I.47. Turbine-flowmeter Measurements during Test LB-130.
ANL Neg. No. 900-77-1402.

Fig. I.48. Fluid Temperature during Test DB-130.
ANL Neg. No. 900-77-1397.

Fig. I.49. Wall-thermocouple Measurement during Test DB-130.
ANL Neg.Nos. 900-77-1399, -1411, -1404, and -1385.

Fig. I.49 (Contd.)

Fig. I.49 (Contd.)

Fig. I.49 (Contd.)

Fig. I.50. System Pressure during Test DB-131.
 ANL Neg. No. 900-77-1416.

Fig. I.51. Differential Pressure across Test Section during
 Test DB-131. ANL Neg. No. 900-77-1421.

Fig. I.52. Differential Pressure across Break Orifice during Test DB-131. ANL Neg. No. 900-77-1432.

Fig. I.53. Turbine-flowmeter Measurement during Test DB-131. ANL Neg. No. 900-77-1414.

Fig. I.54. Fluid Temperature during Test DB-131.
ANL Neg. No. 900-77-1418.

Fig. I.55. Wall-thermocouple Measurement during Test DB-131.
ANL Neg. Nos. 900-77-1415, -1372, -1412, and -1371.

Fig. I.55 (Contd.)

Fig. I.55 (Contd.)

Fig. I.55 (Contd.)

Fig. I.56. System Pressure during Test DB-132.
ANL Neg. No. 900-77-1405.

Fig. I.57. Differential Pressure across Test Section during Test DB-132. ANL Neg. No. 900-77-1405.

Fig. I.58. Differential Pressure across Break Orifice during Test DB-132. ANL Neg. No. 900-77-1395.

Fig. I.59. Turbine-Flowmeter Measurement during Test DB-132.
 ANL Neg. No. 900-77-1396.

Fig. I.60. Fluid Temperature during Test DB-132.
 ANL Neg. No. 900-77-1393 Rev. 1.

Fig. I.61. Wall-thermocouple Measurement during Test EB-132.
 ANL Neg. Nos. 900-77-1394, -1400, -1392, and -1384.

Fig. I.61 (Contd.)

Fig. I.61 (Contd.)

Fig. I.61 (Contd.)

Fig. I.62. System Pressure during Test DB-133.
ANL Neg. No. 900-77-1407.

Fig. I.63. Differential Pressure across Test Section during
Test D3-133. ANL Neg. No. 900-77-1410.

Fig. I.64. Differential Pressure across Break Orifice during Test DB-133. ANL Neg. No. 900-77-1424.

Fig. I.65. Turbine-flowmeter Measurement during Test DB-133. ANL Neg. No. 900-77-1429.

Fig. I.66. Fluid Temperature during Test DB-133.
ANL Neg. No. 900-77-1423.

Fig. I.67. Wall-thermocouple Measurement during Test DB-133.
ANL Neg. Nos. 900-77-1373, -1417, -1401, and -1425.

Fig. I.67 (Contd.)

Fig. I.67 (Contd.)

Fig. I.67 (Contd.)

Fig. I.68. System Pressure during Test DB-134.
ANL Neg. No. 900-77-1419.

Fig. I.69. Differential Pressure across Test Section during Test DB-134. ANL Neg. No. 900-77-1387.

Fig. I.70. Differential Pressure across Break Orifice during Test DB-134. ANL Neg. No. 900-77-1430.

Fig. I.71. Turbine-flowmeter Measurement during Test CB-134.
 ANL Neg. No. 900-77-1376.

Fig. I.72. Fluid Temperature during Test DB-134.
 ANL Neg. No. 900-77-1377 Rev. 1.

Fig. I.73. Wall-thermocouple Measurement during Test DB-134.
ANL Neg. Nos. 900-77-1380, -1381, -1382, and -1431.

Fig. I.73 (Contd.)

Fig. I.73 (Contd.)

Fig. I.73 (Contd.)

Fig. I.74. System Pressure during Test DB-135.
 ANL Neg. No. 900-77-1375.

Fig. I.75. Differential Pressure across Test Section during
 Test DB-135. ANL Neg. No. 900-77-1391.

Fig. I.76. Differential Pressure across Break Orifice during Test DB-135. ANL Neg. No. 900-77-1423.

Fig. I.77. Turbine-flowmeter Measurement during Test DB-135. ANL Neg. No. 900-77-1422.

Fig. I.78. Fluid Temperature during Test DB-135.
ANL Neg. No. 900-77-1433.

Fig. I.79. Wall-thermocouple Measurement during Test DB-135.
ANL Neg. Nos. 900-77-1434, -1413, -1435, and -1426.

Fig. I.79 (Contd.)

Fig. I.79 (Contd.)

Fig. I.79 (Contd.)

TABLE I.3. Location of Instrumentation

Instrument	Symbol	Location, cm ^a
Lower pressure transducer	PT1	-5.1
Upper pressure transducer	PT2	+279.4
Test-section DP cell	DP1	Between -5.1 and +279.4
Break-orifice DB cell	DP2	Horizontal break assembly
Lower turbine flowmeter	TV1	-23.0
Upper turbine flowmeter	TV2	+297.0
Inlet fluid thermocouple	TF1	-5.1
Outlet fluid thermocouple	TF2	+279.4
Wall thermocouples	TC'X'	'X' stands for +x inches

^a+ means above the $z = 0$ line (see Fig. I.1); - means below the $z = 0$ line.

Tests DB-120 and -121 had the same inlet temperature, mass velocity, and test-section power, but the initial steady-state pressure was different; DB-120 was conducted at 2.24 MPa and DB-121 at 2.81 MPa. As a result, the DB-120 wall-temperature profiles exhibit a longer subcooled boiling zone. During the blowdown, the lower 130 cm in DB-120 experienced early CHF as compared to 191 cm in DB-121. Since no wall-thermocouple measurements between 191 and 236 cm were recorded during the transient, early CHF could have been precipitating in part of this length in the high-pressure test, DB-121. Measurements of flowmeters, test-section pressure drop, and pressure differential across the break were comparable in both tests; hence, the thermal hydraulics were similar. The early CHF appears to extend some distance into the subcooled boiling zone. However, in this early portion of subcooled boiling, only comparatively few nucleation sites are operating,⁹ so that a fraction of the wall heat flux is transferred by normal single-phase forced convection, a phenomenon called partial subcooled boiling.

Fully developed subcooled boiling occurs when the single-phase component reduces to zero, and here both velocity and subcooling have relatively little effect on the surface temperature. One of the partial-subcooled boiling models is due to Rohsenow,¹⁰ who suggests that

$$\phi = \phi_{\text{SPL}} + \phi_{\text{SCB}},$$

where ϕ is the total surface heat flux, ϕ_{SPL} is the heat flux transferred by single-phase convection, and ϕ_{SCB} is the heat flux transferred by bubble nucleation. In this method, the single-phase component ϕ_{SPL} of the total heat flux is given by

$$\phi_{\text{SPL}} = h_{\text{SPL}}[T_W - T_f(z)],$$

where h_{SPL} is the single-phase heat-transfer coefficient. With this simple model, the ratio of ϕ_{SPL}/ϕ at the point of transition from early CHF to sustained cooling was found to be 0.70 for DB-120 and 0.8 for DB-121. Hence, upstream of this point, the boiling heat flux accounts for less than 30% of the total heat flux.

The essential point is that not enough nucleation sites are active in steady-state operation and remain active to transfer the full flux during the transient in part of the partial-subcooled boiling region. This experimental comparison and qualitative discussion clearly show the importance of site activation during the blowdown, and this behavior was observed earlier in the water experiment.^{5,11}

Another parameter found to be crucial at the end of subcooled blowdown is the ratio of volumetric generation rate Q_v to break discharge flow rate Q_b . In the case, where the break flow rate is smaller, so that the ratio Q_v/Q_b is greater than one, the system pressure would most likely rise.

It is therefore not difficult to envision that the coolant above the heated channel would be mostly levitated. The resulting limited flow down through the heated zone therefore precipitates dryout conditions quite rapidly.

To a first approximation, the volumetric generation rate can be given by the net amount of vapor generated due to heat input alone. Hence,

$$Q_v = \frac{q}{h_{fg}} (v_g - v_f),$$

where q is the total heat input, h_{fg} is the latent heat, and v_g and v_f are the specific volume of vapor and liquid, respectively. Here the sensible heat required to raise the subcooled liquid to saturation has been ignored and the vapor generation due to flashing is neglected.

The break discharge flow rate through a sharp-edge orifice (in the present system) can be estimated by the conventional incompressible relationship

$$Q_b = 0.61 A_b \sqrt{2 v_f \Delta P},$$

where A_b is the area of the orifice, and the discharge coefficient is taken to be 0.61. This is a reasonable approximation, since the fluid at the discharge is subcooled.

Given the power level and the break size, the ratio Q_v/Q_b at the end of subcooled blowdown can be estimated as shown in Table I.4. Here ΔP is taken to be the difference between the saturation pressure corresponding to the upper plenum and the ambient pressure of 0.1 MPa. The experimental results can be discussed with the help of this table.

TABLE I.4. Comparison of Volumetric Generation Rate (Q_v) with Break Flow Rate (Q_b)

		q , kW	2.7	4.0	4.6	5.0	7.1
		Q_v , m ³ /h	0.65	0.98	1.12	1.23	1.74
d_b , cm	Q_b , m ³ /h	Q_v/Q_b	1.03	1.55	1.77	1.94	2.74
0.254	0.63		0.52	0.78	0.90	0.98	1.38
0.381	1.26		0.26	0.39	0.45	0.49	0.70
0.508	2.52						

Test DB-135 was conducted at 4.0 kW with a break diameter of 0.254 cm. The pressure at the end of subcooled blowdown was observed to rise for almost 1 s, since the Q_v/Q_b ratio was estimated to be 1.55. Again,

because more volume was generated in the heated channel than could be exhausted, essentially all the liquid in the upper plenum was being levitated. This is also evident in the flowmeter measurements, shown in Fig. I.77. The lower portion of the heated tube was caused to experience CHF early in time.

Test DB-130 was performed at the same power, but with a larger break, resulting in a Q_v/Q_b ratio of 0.78. In this case, efficient cooling lasted for 15 s. Test DB-133 was conducted at a higher initial pressure, and practically the same cooling capability was observed. This is understandable, since the pressure effect only becomes apparent in tests exhibiting early CHF. Test DB-128 had a break diameter of 0.508 cm. Again efficient cooling resulted. In this case, it only lasted for about 10 s because, with a larger break, the coolant was exhausted at a faster rate.

The same trend with increasing break size was observed in Tests DB-131, -132, and -134, in which the total heat input was increased to 4.6 kW. However, degradation in blowdown heat transfer was obvious from the wall-temperature plots as compared with tests using 4.0-kW power. This observation is summarized in Table I.5.

TABLE I.5. Summary of Blowdown Heat-transfer Results^a

Break Diameter, cm	Total Power, kW				
	2.7	4.0	4.6	5.0	7.1
0.254		DB-135, 38 cm in early CHF	DB-134, 38 cm in early CHF		
0.381		DB-130, 11-s delay	DB-131, 8 cm in early CHF	DB-107 and -111, 54 cm in early CHF	
0.508	DB-126, 9-s delay	DB-128, 10-s delay			DB-120, 145 cm in early CHF

^aNominal parameters: $P = 2.2$ MPa, $T_{in} = 114^\circ\text{C}$, $T_{out} = 149^\circ\text{C}$.

c. Future Work

The experimental facility will be modified to allow double-ended breaks to be simulated. More steady-state CHF tests will be conducted at equivalent conditions of the PWR and over a range of system pressures.

B. One-dimensional Drift-flux Modeling: Comparison of Local Drift-velocity Correlation to Experimental Data (M. Ishii, RAS)

The present drag-similarity criterion $C_D(N_{Re})$ and the mixture-viscosity concept $\mu_m(\alpha_d)$ have been used to develop a relative velocity relation. This correlation can predict the relative motion between phases for various two-phase flows such as bubble, droplet, and particulate flows. The results are summarized in Table I.6. The comparisons of the theoretical predictions with more than 1000 experimental data showed that satisfactory agreements were obtained at all ranges of particle concentration and Reynolds number.

TABLE I.6. Summary of Drift Velocity (V_{dj}) in Infinite Media

	Bubble in Liquid	Droplet in Liquid	Droplet in Gas	Solid Particle
μ_c/μ_m	$(1 - \alpha_d)$	$-(1 - \alpha_d)^2$	$-(1 - \alpha_d)^{2.6}$	$-(1 - \alpha_d)^{2.6}$
Stokes Regime	$\frac{2}{9} \frac{g \Delta \rho r_d^2}{\mu_f} (1 - \alpha_d)^3$		$\frac{2}{9} \frac{g(\rho_c - \rho_d)}{\mu_c} r_d^2 (1 - \alpha_d)^2 \frac{\mu_c}{\mu_m}$	
Undistorted-particle Regime	$\frac{10.8 \mu_c}{\rho_c r_d} \frac{\mu_c}{\mu_m} (1 - \alpha_d)^2 \frac{\psi^{4/3}(1 + \psi)}{1 + \left[\frac{\mu_c}{\mu_m} (1 - \alpha_d)^{0.5} \right]^{6/7}} \frac{\rho_c - \rho_d}{\Delta \rho}$			For Newton's Regime ($r_d^* \geq 34.67$),
$N_\mu \leq 0.11 \frac{1 + \psi}{\psi^{8/3}}$	where $\psi = 0.55[(1 + 0.08 r_d^{*3})^{4/7} - 1]^{0.75}$			$\psi = 17.67$
Distorted-particle Regime	$\sqrt{2} \left(\frac{\sigma g \Delta \rho}{\rho_c^2} \right) (1 - \alpha_d)^n \frac{\rho_c - \rho_d}{\Delta \rho}$			-
	$n = 1.75$	$n = 2.0$	$n = 2.25$	
Churn-turbulent Regime	$\sqrt{2} \left(\frac{\sigma g \Delta \rho}{\rho_f^2} \right)^{1/4}$	$\sqrt{2} \left(\frac{\sigma g \Delta \rho}{\rho_c^2} \right)^{1/4} \frac{\rho_c - \rho_d}{\Delta \rho}$		-

The range of the volumetric concentration considered in the comparison extends from $\alpha_d = 0$ up to $\alpha_d = 0.55$ for solid-particle systems. For fluid-particle systems, even higher concentration data were included. In particular, the dense packing regime of α_d up to 0.95 was examined for the droplet suspension in liquid.

In terms of the particle Reynolds number, the data used in the comparison spanned from the Stokes regime all the way to the Newton's regime for solid-particle systems. For fluid-particle systems, data for the distorted-particle regime and the churn-turbulent regime were used extensively to study the difference between the solid- and fluid-particle systems.

These results showed that the drag law in various dispersed two-phase flows could be put on a general and consistent base through the use of the mixture viscosity.

Figure I.80 compares the results for a solid-particle system with the empirical correlation of Richardson and Zaki.¹² An agreement at relatively low volumetric concentrations is excellent at all Reynolds-number regions. At very high values of α_d , the present theory predicts much lower drift velocities than the Richardson-Zaki correlation. However, the original experimental data of Richardson and Zaki also indicate this trend, which is predicted by the present theory, as shown in Fig. I.81. The data in Fig. I.81 were taken at relatively high Reynolds numbers; however, the similar behavior also occurred at the lower-Reynolds-number regions. For example, the experimental data of Hanratty and Bandukwala¹³ obtained in the Stokes regime agree with the present theory at all concentrations (see Fig. I.82).

Fig. I.80

Comparison of Predicted Results with Richardson and Zaki's Empirical Correlations for Solid-particle-flow System. ANL Neg. No. 900-76-442.

Fig. I.81

Comparison of Theory with Experimental Data of Richardson and Zaki for Solid-particle Flow at High Reynolds Number. ANL Neg. No. 900-76-444.

Fig. I.82

Comparison of Theory with Experimental Data of Hanratty and Bandukwala for Solid-particulate Flow at Low Reynolds Number. ANL Neg. No. 900-77-1100.

particles. Furthermore, the maximum packing α_{dm} is significantly larger for bubbly or droplet flows than for particulate flows. Consequently, a proper mixture viscosity μ_m and maximum packing α_{dm} should be used.

Of particular interest in this undistorted-fluid-particle regime is the higher-concentration region, i.e., the foam or the dense-packing region, where a pronounced difference between fluid- and solid-particle systems can be observed due to the difference in the maximum packing. Figure I.83 compares

At maximum packing the particles will interlock each other, and therefore they cannot be suspended as individual particles in fluid. Because of this limiting condition, the drift velocity should become very small as the particle concentration approaches the maximum packing α_{dm} . Therefore, the present model properly predicted the relative motion in solid-particulate flows at all Reynolds-number ranges and up to the highest concentration. This can be considered as an indirect proof that the assumed similarity criterion for the drag coefficient based on the multiparticle Reynolds number has been correct.

For a fluid-particle system in the viscous regime, i.e., in the undistorted-particle regime, the particle behaviors should be similar to those of solid-particle systems. The main difference is that, in the fluid-particle systems, the properties of both phases have significant contributions on the size of

Fig. I.83

Comparison of Undistorted-bubble-regime Drift Velocity with Data of Miles et al. at Very High Void Fraction. ANL Neg. No. 900-77-1099.

experimental data of Miles et al.¹⁴ with the present theory. Because of the direct particle-to-particle interactions in the foam region, some discrepancies between the present theory and the data were expected. However, in spite of these effects, the agreement appeared suprisingly satisfactory.

In the distorted-fluid-particle regime, the single-particle drag coefficient depends only on the particle radius and fluid properties and not on the velocity or the viscosity; i.e., $C_{D\infty} = (4/3)r_d\sqrt{g\Delta\rho/\sigma}$. Thus, for a particle of a fixed diameter, $C_{D\infty}$ becomes constant. In considering the drag coefficient for a multiparticle system with the same radius, we must take into account the restrictions imposed by the existence of other particles on the flow field. Because of the random characteristic of the turbulent eddies and particle oscillations, a particle sees the increased drag due to other particles in essentially a similar way as in the Newton's regime for a solid-particle system, where $C_{D\infty}$ is also constant under a turbulent-flow condition.

The result for the distorted-particle regime has been compared to various experimental data for bubbly flows¹⁵⁻¹⁸ and for droplet-liquid flows.^{15,19-21} Figure I.84 shows relative velocities both in the bubbly and the droplet-liquid flow regimes. The data of Lackme¹⁵ clearly indicate the difference in the concentration dependence of the relative velocity between a bubbly flow and a droplet flow. These characteristics have been correctly predicted by the correlation. Figures I.85-I.87 show further comparisons between the theoretical predictions and experimental data, in both the batch and countercurrent bubbly flows. In Fig. I.85, the departure of the experimental data from the prediction around void fraction 0.3 can be observed. This indicates the flow-regime transition due to increased turbulence and bubble coalescences. The new flow regime is either the churn-turbulent or slug-flow regime and it contains considerably larger bubbles.

Fig. I.84

Difference between Bubble System and Droplet Dispersion System in Distorted-particle Regime. ANL Neg. No. 900-77-1101.

Fig. I.85. Comparison of Predicted Gas Flux with Air-Water Experimental Data of Novosad and Siemes. ANL Neg. No. 900-77-1104.

Fig. I.86. Comparison of Predicted Gas Flux with Air-Water Experimental Data of Bridge et al. ANL Neg. No. 900-77-1103.

Fig. I.87

Comparison of Predicted Gas Flux with Air-Water Experimental Data of Bridge et al. in Forced-convection System. ANL Neg. No. 900-77-1102.

Figures I.88-I.90 compare the present theory and the results of various liquid-liquid dispersion experiments. For bubbling water through mercury,¹⁹ the flow-regime transition occurs at $\alpha_d \approx 0.4$. However, as can be seen from

Figs. I.84 and I.90, the distorted-particle regime can exist in much higher void-fraction ranges in the absence of significant amounts of coalescences among fluid particles. Except for the data of Weaver et al.,²⁰ the agreements of the theoretical predictions with the data for various fluid combinations are satisfactory.

Fig. I.88. Comparison of Predicted Liquid Flux with Experimental Data. ANL Neg. No. 900-77-1108.

Fig. I.89. Comparison of Theory with Experimental Data of Weaver et al. for Liquid-Liquid System. ANL Neg. No. 900-77-1107.

Fig. I.90
Comparison of Theory with Experimental Data of Letan and Kehat for Liquid-Liquid System. ANL Neg. No. 900-77-1106.

As the radius of the fluid particle is further increased, the wake and bubble-boundary layer can overlap due to the formation of large wake regions. In other words, a particle can directly influence both the surrounding fluid and other particles. Hence the entrainment of a particle in a wake of other particles becomes possible. This flow regime is known as the churn-turbulent-flow regime and is commonly observed in bubbly flows. In the existence of sufficient turbulent motions in the continuous phase, the transition from the distorted-particle regime to the churn-flow regime occurs at the particle concentration around 0.3. This criterion for the transition can be applied to most

forced-convection two-phase flows. However, for a batch process, detailed coalescence mechanisms and surface contaminations become important in determining the transition.

Fig. I.91. Comparison of Experimental Data with Predicted Gas Flux Based on Churn-flow Model. ANL Neg. No. 900-77-1105.

Figure I.91 compares the prediction for the churn turbulent flow and the experimental data of Yoshida and Akita.²² The data were taken for an air-aqueous sodium sulfite solution system with various column diameters ranging from 7.7 to 60 cm. As the figure indicates, the theory underpredicts the gas flux for smaller-column-diameter experiments. However, for larger-column diameters, the agreement between the prediction and data becomes increasingly satisfactory. This tendency can be easily explained by the two-dimensional effect due to the void and velocity profiles.

higher-flux regions, this will give a higher dispersed phase flux than the case with uniform profiles. Then the mean gas volumetric flux should be somewhat higher than the prediction. For more detailed discussions on this point, see Ref. 23.

The dispersed phase is locally transported with the local drift velocity relative to local volumetric flux. Therefore, if more particles are concentrated in

C. Reflood Tests (Y. S. Cha, CT; G. G. Dewey and J. G. Warner, RAS)

A series of tests with relatively high inlet-liquid velocities were performed. Variables in these tests are listed in Table I.7. Also shown in Table I.7 are the testing conditions for Runs No. 8 and 9 (conducted with relatively low inlet velocities) reported previously. The inlet-liquid temperature was 65°C (150°F) and the initial temperature of the upper plenum was 121°C (250°F) for all the test runs. During these tests, the upper plenum was open to the surroundings at atmospheric pressure.

TABLE I.7. Descriptions of Test Conditions

Run No.	Average Inlet Velocity, cm/s	Initial Temperature of T_{18} , °C	Power, W
8	4.83	724	1.539
9	4.19	774	0
10	25.8	816	0
11	26.1	665	0
12	25.9	535	0
13	26.0	821	1.644
14	26.0	665	1.293
15	26.2	535	0.792
19	12.7	802	0
20	23.0	802	0
21	20.8	802	0

Typical temperature profiles at some distance from the lower electrode are shown in Figs. I.92 and I.93. The slope of the temperature-versus-time curve increases with inlet-liquid velocity, as shown in Figs. I.92 and I.93.

Typical lower-plenum pressures are shown in Figs. I.94-I.96. In these figures, t_{q1} represents the instant when thermocouple No. 1 was quenched. All the tests (except Runs No. 8 and 9) showed that the lower-plenum pressure increased sharply during the beginning of the test and then decreased to some minimum value before it starts to rise again.

The initial sharp rise in lower-plenum pressure was probably due to pressure buildup in the test section as a result of the large rate of vapor generation. The pressure buildup in the test section decreased with time, since the portion of the test section above the quenching liquid front was being

Fig. I.92
Temperature of Thermocouple
No. 18 vs Time for Runs No. 9-12

Fig. I.93

Temperature of Thermocouple No. 18
vs Time for Runs No. 10, 19, and 21

Fig. I.94
Lower-plenum Pressure
vs Time for Run No. 10

Fig. I.95

Lower-plenum Pressure
vs Time for Run No. 12

Fig. I.96

Lower-plenum Pressure
vs Time for Run No. 19

precooled by the upward flow of steam and entrained liquid droplets, as shown in Figs. I.92 and I.93. The hydrostatic pressure increases with time, since the liquid front is moving up continuously. The combined effects of increasing in hydrostatic pressure and decreasing in pressure buildup in the test section form the valley in the lower-plenum pressure profile in Figs. I.94-I.96. The pressure buildup in the test section decreases with decreasing inlet-liquid flow velocity and initial test-section temperature.

The location of the quenching-liquid front can be determined approximately by the temperature profiles shown in Figs. I.92 and I.93. The hydrostatic head (P_h) associated with the quenching front is

$$P_h = \rho gh,$$

where ρ is the density of the water at 65°C, g is the magnitude of gravitational acceleration, and h is the distance between the quenching front and the pressure transducer at the lower plenum. Let ΔP denote the difference between the measured average lower-plenum pressure (P_m) and hydrostatic pressure associated with the quenching liquid front,

$$\Delta P = P_m - P_h.$$

Figures I.97 and I.98 show the variation of ΔP with time. Obviously ΔP decreases with time and approaches zero during later stage of the tests. If the flow in the test section was the so-called inverted annular flow with liquid in the center and a thin vapor film adjacent to the hot surface which had been reported for forced convective boiling inside vertical tubes,²⁴ then ΔP would be equal to the sum of the pressure build-up and the hydrostatic pressure associated with the liquid core ahead of the quenching front.

Fig. I.97. ΔP vs Time for
Runs No. 10-12

Fig. I.98. ΔP vs Time for Runs No. 19-21

Figures I.99-I.102 show the quenching time versus axial distance for various test runs. In these figures, the axial distance was measured from the the bottom electrode and the time zero was chosen to be the instant when thermocouple No. 1 was quenched. These figures indicate that the time required for the quenching front to reach a given location in the test section increases with the initial temperature in the test section and the power input during the test, and decreases with the inlet-liquid velocity.

Fig. I.99

Quenching Time vs Axial Dis-
tance for Runs No. 8 and 9

Fig. I.100
Quenching Time vs Axial
Distance for Runs No. 10-12

Fig. I.101. Quenching Time vs Axial Distance for Runs No. 13-15.

Fig. I.102. Quenching Time vs Axial Distance for Runs No. 19-21.

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II. TRANSIENT FUEL RESPONSE AND FISSION-PRODUCT RELEASE PROGRAM

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A physically realistic description of fuel swelling and fission-product release is needed to aid in predicting the behavior of fuel rods and fission products under certain hypothetical light-water-reactor (LWR) accident conditions. To satisfy the near-term need, a comprehensive computer-based model, the Steady State and Transient Gas-release and Swelling Subroutine (SST-GRASS) 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 EG&G Idaho, Inc., at the Idaho National Engineering Laboratory (INEL). Also being developed, but at a lower priority, is a model to predict the behavior of volatile fission products in and release from LWR fuel under hypothetical accident conditions. The volatile fission-product results will also serve as input to NRC-sponsored programs that are developing a radiological source term for hypothetical accidents.

The analytical effort is supported by a data base and correlations developed from characterization of irradiated LWR fuel and from out-of-reactor transient heating tests of irradiated commercial and experimental LWR fuel under a range of thermal conditions.

Emphasis in the early stages of the program has been on thermal conditions in pressurized-water-reactor (PWR) fuel that are applicable to anticipated hypothetical power-cooling-mismatch (PCM) accidents. Recent efforts include conditions typical of other types of hypothetical accidents. The program is also developing information on fission-gas release during steady-state and load-following operations.

Dr. J. C. Janvier, from the CEA Radiometallurgy Laboratory at Grenoble, visited us on April 6 to exchange information within the CEA-NRC agreement on LWR safety. Also, we made presentations at, and participated in the following meetings: American Ceramic Society Annual Meeting, Chicago, IL, April 23-28; MATPRO (Materials Properties) Workshop on models for fuel and cladding properties between NRC contractors and representatives of PNS-GFK, Idaho Falls, ID, May 16-18; and the ANS Meeting, New York, NY, June 12-17.

Recent significant analytical and experimental advances and the status of the program at the end of this quarter are summarized below.

1. GRASS fission-gas release predictions for transient heating conditions are in good agreement with results of direct-electrical-heating (DEH) simulation tests of power-cooling-mismatch (PCM) accidents.
2. Results of the GRASS analyses for PCM-type DEH tests suggest that at high temperatures, fission-gas bubbles migrate primarily by a surface-diffusion-based mechanism.
3. Based on data from the initial series of PCM-type tests, relationships have been developed between released fission gas and transient temperature history and between released fission gas and microstructural change. Preliminary conclusions about the mechanisms of fission-gas release have been developed on these empirical correlations.
4. Improvements have been made in our ability to calculate the radial temperature profile of DEH test specimens by incorporating more accurate UO_2 physical-property data and a transient cracking model into the DEHTTD (DEH Transient Temperature Distribution) code.
5. A topical report on the initial series of PCM-type tests has been completed.
6. Metallographic and SEM investigation of PBF-tested Saxton fuel has shown that there are intergranular separations similar to those seen in DEH-tested Robinson fuel and that the separations result from the same fission-gas bubble growth and coalescence mechanisms.

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

1. Modeling of Fission-gas Behavior during Transient Heating Conditions

During this quarter, verification of the transient GRASS analysis was initiated using available data from the initial series of PCM-type DEH tests. The GRASS simulation of the DEH tests consisted of three distinct phases:

- a. The DEHTTD code was modified to obtain reasonable agreement between calculated and observed incipient fuel melting.
- b. GRASS was coupled to the DEHTTD code.
- c. The effect of changing uncertain materials properties used in the GRASS code on the agreement between calculated and measured gas release from the DEH-tested pellets was examined.

Item a is covered in detail in Sec. B.5. The interfacing of GRASS with the DEHTTD code consisted mainly of writing an interpolating routine to obtain fuel temperatures applicable to the SST-GRASS nodal spacing (equal-mass rings) from the DEHTTD-code radial nodes (rings with equal thickness) and temperature arrays.

The SST-GRASS code was run for the steady-state irradiation of the DEH test fuel in the H. B. Robinson reactor, and the end-of-life fuel and fission-gas history was put into a disk storage file to provide the necessary initial conditions to the transient analysis. Thus, the first step in a GRASS-DEHTTD run is to read in this storage file and set all the GRASS arrays to the end-of-life steady-state conditions.

The results of an initial comparison of GRASS-calculated gas-release values with experimental results from the PCM-type DEH tests show that, in general, GRASS underpredicts DEH transient gas release. Although GRASS predictions agree reasonably well with DEH test results characterized by very low to moderate gas release ($<10\%$), the code predictions do not agree with the results of DEH tests having high values of fractional fission-gas release ($>10\%$). Results from the DEH tests show that there is a strong correlation between the degree of grain-boundary separation (i.e., increase in pore-solid surface area) and the amount of fission-gas release. (Gas release increases as pore-solid surface area increases; e.g., see Sec. B.3.c.) The GRASS code is assumed to have the potential to predict the gas release for a DEH test if enough gas is calculated to be trapped in the grain-edge tunnels to account for the difference between the calculated and the measured gas release; the gas trapped in the grain-edge tunnels would be released during grain-boundary separation. However, for the DEH tests having high fractional gas release, the amount of gas predicted to be trapped in the grain-edge tunnels at the end of the test is much too low to account for the difference between calculated and observed release values.

The above analysis demonstrates that GRASS is not releasing enough gas from within the grains either during the steady-state calculation, or during the DEH transient, or both. The following describes analyses that were performed to study the effect of changing uncertain materials properties used in GRASS on the calculation of transient fission-gas release.

a. Effect of Intragranular Bubble Diffusivities and Re-resolution Rates on Transient Gas Release

Intragranular gas-bubble diffusivities and gas-atom re-resolution rates in UO_2 are materials properties that can strongly affect intragranular gas release during steady-state conditions. Estimates of the diffusivity of atomic xenon at 1500°C , based on the results of various experiments,¹ differ by as much as three orders of magnitude. Gas-atom re-resolution rates are extremely difficult to obtain experimentally, and theoretical estimates are very model-dependent.

Tables II.1 and II.2 show the calculated end-of-life values for the fraction of retained gas trapped along grain edges for the H. B. Robinson fuel for various values of the intragranular gas-bubble diffusivities, D , and gas-atom re-resolution rates, b , respectively. These results were generated assuming that the fraction, F_{BL} , of the gas atoms knocked out of grain-boundary bubbles due to re-resolution that reentered the lattice equals 1.0.

TABLE II.1. Calculated Fraction of Retained Gas Trapped along Grain Edges as a Function of Intragranular Fission-gas-bubble Diffusivities, D , with $F_{BL} = 1.0$

Intragranular Gas-bubble Diffusivities, cm^2/s	Fraction of Retained Gas Trapped along Grain Edges, %
D_G^a	1.0
$10D_G$	2.0
$100D_G$	7.0
$1000D_G$	11.0

^a D_G is the diffusivity that has been used in GRASS.³

TABLE II.2. Calculated Fraction of Retained Gas Trapped along Grain Edges as a Function of Gas-atom Re-resolution Rate, b , with $F_{BL} = 1.0$

Gas-atom Re-resolution Rate, s^{-1}	Fraction of Retained Gas Trapped along Grain Edges, %
$0.01b_G^a$	17.0
$0.1b_G$	8.0
b_G	1.0
$10b_G$	0.1

^a b_G is the re-resolution rate that has been used in GRASS.³

Tables II.1 and II.2 show that intragranular gas release increases at the diffusivities increase and the re-resolution rate decreases. The intragranular gas release increases as the re-resolution rate decreases due to a corresponding decrease in the flux of "re-resolution" atoms from the boundaries back to the lattice. However, the assumption that all gas atoms leaving grain-boundary bubbles due to re-resolution reenter the lattice may not be realistic. A certain fraction of the gas atoms knocked out of grain-boundary bubbles may remain on the boundaries, or stop close enough to the boundaries to be quickly recaptured.

Tables II.3 and II.4 show the calculated end-of-life values for the fraction of retained gas trapped along grain edges for the H. B. Robinson fuel for various values of the intragranular gas-bubble diffusivities and the gas-atom re-solution rates, respectively, assuming that only a fraction, r_d/a , of the gas atoms leaving the boundaries due to re-solution reenter the lattice; r_d is the re-solution distance [$50 \text{ \AA}$ ($5 \times 10^{-9} \text{ m}$), in this example], and a is the UO_2 grain radius.

TABLE II.3. Calculated Fraction of Retained Gas Trapped along Grain Edges as a Function of Intragranular Fission-gas-bubble Diffusivities, D , with $F_{BL} = r_d/a$

Intragranular Gas-bubble Diffusivities, cm^2/s	Fraction of Retained Gas Trapped along Grain Edges, %
D_G	9.8
$10D_G$	10.5
$100D_G$	10.6
$1000D_G$	21.5

TABLE II.4. Calculated Fraction of Retained Gas Trapped along Grain Edges as a Function of Gas-atom Re-solution Rate, b , with $F_{BL} = r_d/a$

Gas-atom Re-solution Rate	Fraction of Retained Gas Trapped along Grain Edges, %
$0.01b_G$	5.6
$0.1b_G$	4.9
b_G	9.8
$10b_G$	12.0

Table II.4 shows that intragranular gas release increases as the re-solution rate increases. Table II.3 shows that intragranular gas release is relatively insensitive to changes in the intragranular fission-gas bubble diffusivities. These results suggest that, for $F_{BL} = r_d/a$, intragranular gas release is re-solution-controlled; i.e., the gas released from the grains consists mainly of single gas atoms. Fission-gas bubbles behave primarily as temporary gas-atom trapping sites.

Twenty-five percent of the retained gas (slightly less than the sum of the values from Table II.1 and II.2) is calculated to be trapped along

the grain edges when $D = 1000D_G$, $b = 0.01b_G$, and $F_{BL} = 1.0$. For this case (i.e., maximum steady-state gas release to the grain edges), Table II.5 lists the measured gas release, the predicted values of the transient gas release, and the fraction of gas trapped along the grain edges for DEH transient heating tests with available measurements of the pore-solid surface area. Parameters in the GRASS calculation describing the degree of interlinked porosity (see ANL-75-72, p. 30) were adjusted to maintain the predicted steady-state gas release at about the measured value.

TABLE II.5. GRASS Results for Gas Release during PCM-type DEH Transients with $D = 1000D_G$, $b = 0.01b_G$, and $F_{BL} = 1.0$

Test No.	Measured Gas Release, %	Calculated Gas Release, %	Calculated Fraction of Retained Gas Trapped along Grain Edges, %
22	13.3	2.6	35
24	12.0	5.7	57
29	2.0	0.81	25
32	16.1	5.4	63
26 ^a	62.3	7.1	80

^aMeasured surface temperatures were not available; estimated surface temperatures were used to obtain calculated results.

From Table II.5, the observed releases are within the capability of code prediction for these values of the intragranular bubble diffusivities and gas-atom re-resolution rates, assuming that the differences between the observed and calculated releases can be explained by the differing degrees of grain-boundary separation for the tests (i.e., increase in pore-solid surface area). The measured values of the pore-solid surface area for the tests in Table II.5 are listed in Table II.6, along with the total calculated releases obtained by using the measurements of pore-solid surface area to calculate the amount of gas released during each test due to grain boundary separation.

TABLE II.6. GRASS Results for Gas Release during PCM-type Transients Obtained by Normalizing the Amount of Grain-edge Gas Released per Unit Increase in Pore-solid Surface Area to Results of Test 29

Test No.	Measured Gas Release, %	Pore-Solid Surface Area per Unit Volume, 10^2 mm^{-1}	Calculated Total Gas Release, %
22	13.1	3.32	10.4
24	12.0	2.6	15.3
29	2.0	0.69	2.0
32	16.1	3.0	17.7
26 ^a	63.1	4.31	40.0

^aMeasured surface temperatures were not available. Estimated surface temperatures were used to obtain calculated results; 100% release from molten fuel was assumed.

The amount of gas released during each test was obtained by normalizing the amount of grain-edge gas released per unit increase in the pore-solid surface area to the results of Test 29. The total transient gas release, TGR, is then given by

$$\text{TGR} = A_1 + (1 - A_1/100)A_2\alpha S_V^{\alpha P}, \quad (1)$$

where A_1 is the calculated release listed in Table II.5, A_2 is the fraction of retained gas trapped along the grain edges in Table II.5, $S_V^{\alpha P}$ is the measured pore-solid surface area, and α is a coefficient relating $S_V^{\alpha P}$ to the release of gas from the grain edges due to grain-boundary separation. By normalizing Eq. 1 to the observed release from Test 29, we determined that α was 0.69×10^{-3} . The gas-release results of Table II.6 are also shown in Fig. II.1.

Fig. II.1

Predicted vs Measured Gas Release Using $D = 1000D_G$, $b = 0.01b_G$, and $F_{BL} = 1.0$.
Neg. No. MSD-64417.

The calculated results shown in Table II.6 and Fig. II.1 are in reasonable agreement with the experimental results. The linear relationship between the fractional gas release and $S_V^{\alpha P}$, Eq. 1, was assumed valid for all the tests shown in Table II.6 and Fig. II.1. However, the experimental results (see Sec. B.3.c) show that the relationship between gas release and $S_V^{\alpha P}$ is linear only up to a point; then there is a break in the curve as the slope increases. Test 26 lies on the curve above the break. Thus, using Eq. 1, which actually represents the curve below the break, for this test is not realistic. This fact partially accounts for the gas release from test 26 being underpredicted. However, the relatively good agreement between the measured and predicted gas release shown in Table II.6 and Fig. II.1 does not necessarily mean that $D = 1000D_G$, $b = 0.01b_G$, and $F_{BL} = 1.0$ are the correct values for these material properties.

There is no experimental evidence to indicate that 25% of the retained gas is trapped along the grain edges, as this case predicts. In addition, the choice of $D = 1000D_G$ is somewhat unreasonable; the GRASS diffusivities at low temperatures may be off by as much as an order of magnitude, as the results of the more definitive experiments vary by this amount. However, a further increase in the diffusivities is not warranted in light of the reasonable agreement between code predictions and experiment for steady-state gas release. Finally, the reasonable agreement between the calculated and measured transient-gas-release values shown in Fig. II.1 may simply be a reflection of the strong correlation between gas release and increase in pore-solid surface area (e.g., see Figs. II.19 and II.21 in Sec. B).

b. Effect of Nonequilibrium Phenomena and High-temperature Diffusivities on Transient Fission-gas Release

On the other hand, if the GRASS steady-state analysis is assumed to be correct (in what follows, $F_{BL} = r_d/a$), the fact that GRASS underpredicts DEH transient-gas release must be due to a low rate of release of gas from the grains during transient conditions. Two factors that can affect the release of gas from within the grains during transients are the bubble-size distribution (i.e., the larger the bubble, the smaller its mobility), and gas-bubble diffusivities at elevated temperatures.

(1) Effect of Bubble Size on Transient Gas Release. During transient conditions, coalescing bubbles have a limited rate of growth due to nonequilibrium effects. In GRASS, this limited rate of growth is ascribed to a shortage of available vacancies in a lattice that is trying to maintain thermodynamic equilibrium (see ANL-76-121, p. 62). However, various material properties associated with the availability of vacancies in the lattice are uncertain (e.g., vacancy diffusivities, and activation and formation energies). If GRASS is overpredicting bubble growth during the transient, then the rate of release of gas from the lattice to the grain surfaces will be reduced due to the reduced mobility of the fission-gas bubbles.

Sensitivity studies made with the GRASS code for cases of increasing lattice-bubble relaxation times indicate that the increase in gas release from the grains due to a distribution of bubbles having much smaller sizes is not enough to account for the high yields of released fission gas observed from some of the DEH-tested pellets. For example, Table II.7 lists

TABLE II.7. Maximum Potential Gas Release for Several DEH Tests for Two Values of Vacancy Diffusivity

Test No.	Measured Release, %	Maximum Calculated Release Using D_v , cm^2/s	Maximum Calculated Release Using $10^{-3}D_v$, cm^2/s
24	12.0	8.5	15
27	33.3	10	15

the maximum potential release predicted for two DEH tests for different values of the vacancy diffusivity, D_v : the lower the vacancy diffusivity, the longer the bubble relaxation time and the smaller the final bubble size. Thus, although this mechanism can account for the moderate gas release observed in Test 24, the higher release test, Test 27, is still underpredicted.

(2) Effect of High-temperature Diffusivities during Nonequilibrium Conditions on Transient Gas Release. Another material property that

can have a strong effect on gas release during a transient is the gas-bubble diffusivities during transient conditions at high temperatures. Actually, very little is known about the bubble diffusivities in UO_2 at high temperatures ($>1500^\circ\text{C}$). For example, FRAS code calculations are made using diffusivities based on the theory of surface diffusion.⁴ Between 1800 and 2800°C (UO_2 melts at about 2850°C), there are six to seven orders of magnitude separating bubble diffusivities based on an extrapolation (see ANL-75-58, p. 35) of Gulden's measurements at 1500°C and those obtained from the theory of surface diffusion (e.g., see Fig. II.2).

Fig. II.2. Intragranular Diffusivity of a 100-atom Fission-gas Bubble.
Neg. No. MSD-64419.

T_c , and connects the steady-state diffusivities with those given by the theory of surface diffusion. In general, the transition curve could also depend on the

In general, intragranular bubble transport by surface diffusion under typical steady-state conditions is thought unlikely. Bubble movement by surface diffusion requires that the matrix atoms move out of the UO_2 crystal and onto the bubble surface easily, and vice versa. However, during equilibrium conditions, the bubbles may be faceted. The rate of motion of a faceted bubble is determined by the frequency of nucleation of steps, instead of the time required for atoms to move from a step on one side of the bubble to a step on the other side.⁵ During transient conditions, step nucleation could be rapidly enhanced by high fuel temperatures⁶ and by nonequilibrium effects. This mechanism could lead to a rapid increase in the fission-gas-bubble mobilities.

For example, two possible transition curves are shown in Fig. II.2; the transition curve starts at some critical temperature,

heating rate (i.e., the degree of nonequilibrium in the lattice). For the purposes of the present discussion, the transition curves in Fig. II.2 will be assumed valid for a typical PCM-type DEH heating rate of 25°C/s. The critical temperature, T_c , represents the temperature at which the step nucleation becomes rapid enough so that this mechanism no longer totally dominates the bubble mobility.

Figures II.3 and II.4 show the results of GRASS calculations of transient-gas release plotted against the measured results for available PCM-type DEH tests. The calculated results in Figs. II.3 and II.4 were

Fig. II.3

Predicted vs Measured Gas Release Using Transition Curve A. Neg. No. MSD-64416.

Fig. II.4

Predicted vs Measured Gas Release Using Transition Curve B. Neg. No. MSD-64415.

reached using the transition curves A and B, respectively, shown in Fig. II.2. Except for Tests 24 and 32, the results for transition curve A are surprisingly good. The surface-temperature measurements for Test 26 were not available, and estimated surface temperatures were used to calculate the radial temperature profiles; the spread in the calculated results for Test 26 reflects a 200°C difference in the maximum surface temperature. The effect of using transition curve B, which starts at a higher critical temperature, is to decrease the predicted gas release. The gas-release predictions shown in Fig. II.3 and II.4 were made assuming that all gas trapped along the grain edges escaped during the transient due to grain-boundary separation. For example, the predictions shown in Fig. II.3 were made by adding to the GRASS calculated release (i.e., no release via grain-boundary separation) the quantity of gas calculated to be trapped along the grain edges, as shown in Table II.8.

TABLE II.8. GRASS Results for Gas Release during PCM-type DEH Transients Using Transition Curve A

Test No.	Calculated Release (No release via grain-boundary separation), %	Fraction of Retained Gas Trapped along Grain Edges, %	Total Release Assuming Gas Trapped along Grain Edges Is Released, %
22	9.5	4.0	13.1
23	4.0	2.0	5.92
24	22.7	13.3	33.0
26	34.9	33.5	56.7
27	20.1	12.1	29.8
29	0.03	1.01	1.07
30	0.04	1.06	1.1
32	24.0	15.0	35.4
33	25.3	23.7	43.0

At present, GRASS does not include a model for the formation of cracklike intergranular separations. A comparison of the GRASS calculations with the posttest microstructures indicates that the radial zones in which large amounts of gas were calculated to be transported to the grain boundaries coincide with the regions in which measured swelling exceeded the assumed value for long-range porosity interconnection (19%). The assumption that all the grain-edge gas escapes leads, in most cases, to a slight overprediction of the measured gas release.

The gas release from Tests 24 and 32 is overpredicted by about a factor of two (e.g., see Figs. II.3 and II.4). The reason for this is not completely understood at this time. However, the GRASS gas-release predictions for these tests could be high because of DEHTTD overpredictions of fuel temperatures (e.g., see Sec. B.5). In this context, note that Test 24 is about a factor of two below the trend line on the experimental plot of fractional gas release versus maximum centerline temperature (see Fig. II.12 later).

Test 32 may also be below this trend line by about a factor of 2, but this is difficult to quantify due to the very steep slope of the data. Clearly, additional experimental information is required.

c. Conclusions

In summary, the effect of possible variations of the values of certain materials properties used in the GRASS code on the predicted gas release has been studied. There are arguments for and against all possibilities. Based on the available evidence, we lean toward using the higher diffusivities at high temperatures during transient conditions because:

1. The hypothesis that step nucleation is rate-controlling during steady-state conditions and that the step density increases rapidly during transient heating conditions and leads to higher gas-bubble mobilities is based on experimental evidence⁵ as well as theoretical arguments.^{5,6}
2. The GRASS code predictions are completely independent of the results of the experimental study.
3. The code predictions using these values of the gas-bubble diffusivities are in fairly good agreement with the observed releases.

These analyses are continuing. Work is in progress to develop an analytical formulation for use in place of the transition curves shown in Fig. II.2.

B. Experimental Program (S. M. Gehl and L. R. Kelman, MSD)

The initial series of direct-electrical-heating (DEH) tests to simulate hypothetical power-cooling-mismatch (PCM) accidents was continued this quarter, and our ability to calculate radial temperature profiles was improved. All PCM-type tests to date were evaluated, empirical correlations were developed between released fission gas and transient temperature history and between released fission gas and microstructural change, and some preliminary conclusions about the mechanisms of gas release during thermal transients were drawn. Progress was also made in the microstructural characterization of PBF-tested Saxton Load Follower fuel. A topical report on the initial series of PCM-type tests was written.

1. Transient Heating Tests (S. M. Gehl, D. R. Pepalis, and R. B. Holdsworth, MSD)

Table II.9 lists experimental conditions for three DEH transient heating tests (Tests 31-33) conducted in the last quarter, along with conditions for the other tests in the series of PCM simulations. In Test 31, the fuel was subjected to a steady-state power input of 500 W for ~15 min before transient heating was started. Surface temperatures reached 1270°C during the steady-state heating. In all other tests, the pretransient power was held to 300 W or less. Calculations of the pretransient and transient temperature histories of Test 31 have not been completed.

TABLE II.9. Experimental Conditions and Gas Release for PCM-type Tests

Test No.	Spec. No. 155-	Duration of Test, s	Power Ramp, W/s		Integrated Power, 10 ⁴ W-s	Average CL Heat Rate, °C/s	Xenon Release		Krypton Release		Estimated Release from Unmelted Fuel, ^a %	
			Programmed	Actual			cm ³ /g	% ^b	cm ³ /g	% ^b	Xe	Kr
9	AC18	15	500	500	1.40	430	0.08	12				
16	AC25	9	60	67	0.64	200	0.01	2	0.003	6		
18	AC26	31	7.5	21	1.88	80	0.05	8	0.008	16		
21	AC27	68	4.0	10	3.16	25	0.320 ± 0.007	50.0 ± 1.0	0.030	60	44 ± 1	55
22	AD6	36	4.0	7.3	1.64	36	0.085 ± 0.003	13.1 ± 0.5	0.012	25		
23	AD18	28	4.0	7.3	1.17	41	0.006-0.030	1-5	0.001-0.005	2-10		
24	AD29	50	4.0	8.4	2.54	28	0.082 ± 0.003	12.0 ± 0.5	0.016	30		
25	AD40	58	4.0	10	3.38	25	0.162 ± 0.015	43.0 ± 3.9	0.021	72	36 ± 4	68
26	CC4	54	4.0	2.1	4.32	15	0.427 ± 0.011	63.1 ± 1.6	0.032	61	59 ± 2	57
27	CC3	54	4.0	7.4	3.24	22	0.225 ± 0.004	33.3 ± 0.7	0.033	63		
28	CC5	50	4.0	6.8	2.55							
29	CC11	59	4.0	4.5	2.50	12	0.014 ± 0.004	2.0 ± 0.6	0.002	4		
30	CC6	70	4.0	0.7	1.46	10	0.003 ± 0.001	0.4 ± 0.1	4 × 10 ⁻⁴	0.8		
31	CC13	71	4.0	12	6.17		0.141 ± 0.011	20.1 ± 1.5	0.010	19		
32	CC14	60	4.0	5.5	2.59	22	0.110 ± 0.016	16.1 ± 2.4	0.008	15		
33	CC15	100	4.0	5.0	4.14	12	0.292 ± 0.015	42.6 ± 2.2	0.026	49		

^aAssuming that the liquid fuel released its entire gas burden.

^bCorrected for burnup and normalized to pretest xenon and krypton contents of 0.685 and 0.053 cm³/g, respectively.

A few notes are necessary to explain some of the entries in Table II.9. Because of problems with the power-control circuitry, the power ramp experienced by the test specimen usually differed somewhat from the programmed ramp. The columns headed "Power Ramp Programmed" and "Power Ramp Actual" permit an evaluation of these differences.

The confidence intervals indicated for the xenon release values were obtained by assuming a possible error of plus or minus one in the least significant digit of the gas-sample chemical analysis. Other sources of uncertainty, due to possible errors in calibrating the volume of the gas-measuring and -sampling system and in reading the pressure of the gas recovered from the charcoal, were insignificant compared to the assumed errors in the chemical analyses.

Gas release was also calculated as a percentage of the amount present in the fuel at the end of irradiation. Retained gas concentrations of 0.685 and 0.053 cm³/g for xenon and krypton, respectively, as determined by whole-pellet dissolution, were corrected for axial burnup variations and used for calculating the percentage released.

Figure II.5, a plot of xenon release as a function of total energy for all PCM-type DEH tests, shows that there is a general trend of higher gas-release fractions at higher total power depositions. Test 31 shows the only large deviation from the trend indicated by the parallel dashed lines in Fig. II.5. This deviation may be due to the long, high-power pretransient heating in Test 31. The details of the thermal history must be examined to explain the apparent scatter in the data for the other tests.

Fig. II.5

Xenon Release as a Function of Total Energy for PCM-type DEH Tests. Neg. No. MSD-64418.

2. Relationships between Gas Release and Transient Temperature History (S. M. Gehl, MSD)

a. Parameters for Describing Temperature History

Radial temperature profiles are calculated as a function of time during DEH tests with the DEHTTD code. The radial temperature profiles

of Test 24, plotted in Fig. II.6 for both the heating and cooling portions of a transient, show the general trends observed for all tests. Both centerline and surface temperature increase monotonically during the power-on portion of the transient. Since the centerline temperature increases more rapidly

than the surface temperature, the volume-averaged temperature gradient increased during this portion of the transient. After the power was turned off, the centerline temperature decreased more rapidly than the surface temperature, and the temperature gradient also decreased.

Fig. II.6. Time Sequence of Radial Temperature Profiles for Test 24. Neg. No. MSD-64420.

To develop relationships between gas release and thermal conditions, we had to describe the complex temperature behavior shown in Fig. II.6 with a set of simplified parameters. The quantities used for this purpose included time integrals of temperature and temperature gradient, referred to as integral parameters, and terms that express the extremes in temperature and temperature gradient, called maximum parameters. A partial list of the parameters considered and the notation used for this representation are given in Table II.10.

Because of the similarity in test conditions for the present series of DEH experiments, relationships were discovered between parameters that are, in principle, mutually independent. These relationships greatly reduced the number of independent parameters needed to describe the temperature histories. As an example, the relationship between two of the integral parameters is presented in Fig. II.7, in which $\int (\bar{dT}/\bar{dR}) dt$ is plotted against $\int \bar{T} dt$ for nine DEH tests. The integrals were evaluated separately for the heating and cooling portions of the transients. Both sets of data can be described by a single straight line passing through the origin. Relationships of this type were found among all the integral parameters.

Similarly, as shown in Fig. II.8, there is a relationship between the maximum parameters. However, a plot of $(dT/dR)_{\max}$ versus $\int \bar{T} dt$ (see Fig. II.9) shows that the maximum parameters are nearly

TABLE II.10. Parameters Used to Describe Transient Thermal History

Integral parameters	{	1a. $\int_P \bar{T} dt$	Time integral of volume-averaged temperature, powered portion of transient only
		1b. $\int_t \bar{T} dt$	Time integral of volume-averaged temperature, total transient
		2. $\int T_{CL} dt^a$	Time integral of centerline temperature
		3. $\int (\overline{dT/dR}) dt^a$	Time integral of volume-averaged radial temperature gradient
Maximum parameters	{	4. $T_{CL_{max}}$	Maximum centerline temperature
		5. $(\overline{dT/dR})_{max}$	Maximum volume-averaged temperature gradient

^aThese integrals may be evaluated separately for the powered portion and the totality of the transient as shown for items 1a and 1b.

Fig. II.7. Relationship between Time Integrals of Volume-averaged Temperature and Temperature Gradient for PCM-type Tests. ANL Neg. No. 306-77-314.

Fig. II.8. Relationship between Maximum Centerline Temperature and Maximum Volume-averaged Temperature Gradient for PCM-type Tests. ANL Neg. No. 306-77-316.

Fig. II.9. Maximum Volume-averaged Temperature Gradient as a Function of Time Integral of Volume-averaged Temperature. ANL Neg. No. 306-77-315.

independent of the integral parameters. Therefore, the transient temperature history can be fully described by two nearly independent parameters, one selected from the group of integral parameters and the other from the maximum parameters.

The small number of independent thermal-history parameters is due to the similarity in test conditions. For tests that cover a wider range of initial conditions and heating rates, a larger number of parameters would be independent. As the range of test conditions is expanded, it may be determined that other parameters are more useful for correlating with gas release. However, for the limited range of tests conducted, the parameters selected are adequate for describing the observed gas-release fractions.

b. Relationships between Gas Release and Temperature-history Parameters

Calculated temperature-history parameters for nine DEH experiments are tabulated in Table II.11. The transient temperature histories are still being evaluated for the remaining tests. The confidence intervals indicated were determined by assuming that the calculated values of T and TCL were in error by $\pm 200^\circ\text{C}$. The possible sources of error in the temperature calculations are discussed in Sec. B.5. Percentage xenon release is plotted against $\int_p \bar{T} dt$ in Fig. II.10 and against $(dT/dR)_{\max}$ in Fig. II.11. A general trend of higher gas-release fractions at higher values of $\int_p \bar{T} dt$ is observed (see Fig. II.10), although a large amount of scatter is evident. Gas release correlates more strongly with $(dT/dR)_{\max}$, as shown in Fig. II.10.

The data suggest that there is a threshold value of temperature gradient, equal to $\sim 3 \times 10^2 \text{ } ^\circ\text{C}/\text{mm}$, above which large amounts of gas release are observed. Because there is an empirical correlation between $TCL_{\max}$ and $(dT/dR)_{\max}$, a relationship also exists between xenon release and $TCL_{\max}$. This relationship, plotted in Fig. II.12, shows a rapid increase in gas release for centerline temperatures above $\sim 2500^\circ\text{C}$.

3. Relationships between Gas Release and Microstructural Change (S. M. Gehl, MSD)

During DEH testing of the Robinson fuel, a complex sequence or sequences of microstructural changes occurred that resulted in the formation

TABLE II.11. Temperature-history Parameters and Quantitative Stereology
Results for PCM-type Tests

Test No.	Xenon Release %	$\int \bar{T} dt,$ $10^5 \text{ } ^\circ\text{C} \cdot \text{s}$		$\int_p T_{CL} dt,$ $10^5 \text{ } ^\circ\text{C} \cdot \text{s}$	$\int (\bar{dT}/\bar{dR}) dt,$ $10^4 \text{ } ^\circ\text{C} \cdot \text{s/mm}$		$(\frac{\bar{dT}}{\bar{dR}})_{\max},$ $10^2 \text{ } ^\circ\text{C/mm}$	$T_{CL\max},$ $^\circ\text{C}$	V_V^P	% Swelling	$S_V^{\alpha P},$ $\frac{\text{mm}^2}{10^2 \text{ mm}^3}$	$(S_g^{\alpha P})^{xs}$ $10^4 \text{ mm}^2/\text{g}$
		Power On	Total		Power On	Total						
9	12.6											
16	2.3											
18	8.2											
21	44.1								0.224	25.0	4.06	4.12
22	13.1 ± 0.5	0.482	0.697	0.75	0.684	1.012	3.01	2510	0.155	14.8	3.32	2.93
23	1-5	0.354	0.524	0.47	0.464	0.717	2.60	2320				
24	12.0 ± 0.5	0.780	1.121	1.12	1.203	1.787	3.85	2830	0.137	12.4	2.60	2.10
25	42.9 ± 3.9											
26 ^a	59 ± 2	1.2	1.4	1.55	1.7	2.2	4.8	3100	0.264	31.8	4.31	4.69
27	33.3 ± 0.7	0.897	1.166	1.07	1.421	1.854	3.85	2740				
28									0.132	11.8	2.36	1.83
29	2.0 ± 0.6	0.613	0.738	0.86	0.869	1.024	1.65	1700	0.030	0	0.69	0
30	0.4 ± 0.1	0.735	0.909	0.92	0.890	1.093	1.69	1660				
31	20.1 ± 1.5											
32	16.1 ± 2.4	0.934	1.150	1.32	1.151	1.538	3.40	2700	0.179	18.1	3.00	2.68
33	42.6 ± 2.2	1.697	2.046	2.09	2.326	2.818	3.22	2660				

^aCalculated temperature parameters for Test 26 are based on estimated surface-temperature histories.

Fig. II.10

Percent Xenon Release as a Function of the Time Integral of Volume-averaged Temperature. ANL Neg. No. 306-77-318.

Fig. II.11

Percent Xenon Release as a Function of Maximum Volume-averaged Temperature Gradient. ANL Neg. No. 306-77-311.

Fig. II.12

Percent Xenon Release as a Function of Maximum Centerline Temperature. ANL Neg. No. 306-77-322.

of extensive networks of interconnected, intergranular porosity. The network provided escape paths for much of the fission gas that was able to diffuse to the grain boundaries. Quantitative relationships have been developed that link fission-gas release with the transient-induced microstructural changes. Before these relationships are presented, it is useful to describe the as-irradiated microstructure and the observed microstructural changes both qualitatively and quantitatively.

a. The As-irradiated Microstructure

During irradiation, the density of the Robinson fuel increased from the as-fabricated value of 92% to a final density of 94% of theoretical. Approximately half the porosity in the irradiated fuel is in the form of large, roughly spherical pores with diameters in the range 0.3-0.25 mm. These pores, formed during fabrication, were apparently not altered during irradiation. Examples of this kind of porosity are visible in the polished plane section through an as-irradiated fuel pellet shown in Fig. II.13. Most of the remaining porosity consists of isolated intergranular pores that are too small to be visible at the magnification of Fig. II.13. These pores, shown in Fig. II.14, range in size from 0.5 to 2.0 μm and have a specific surface area of 69 mm^2/mm^3 . The fine-scale pores, located predominantly at the grain boundaries, are remnants of the as-fabricated porosity after in-pile densification.

Fig. II.13

Partial Transverse Section through an As-irradiated Robinson Pellet. Neg. No. MSD-187872.

Fig. II.14

Grain Structure and Fine-scale Porosity in As-irradiated Robinson Fuel. Neg. No. MSD-190434.

Intra- and intergranular fission-gas bubbles with diameters between 10 and 50 mm were also present in the as-irradiated structure. Rough estimates of the density of fission-gas bubbles from replica fractographs of the type shown in Figs. II.15 and II.16 gave values of 10^{10} - 10^{11} bubbles/mm³.

Fig. II.15. Intragranular Fission-gas Bubbles in As-received Robinson Fuel. Neg. No. MSD-185902.

Fig. II.16. Intergranular Fission-gas Bubbles in As-received Robinson Fuel. Neg. No. MSD-185901.

b. Microstructural Changes during DEH Transients

The most striking feature of the post-DEH microstructure is the pattern of intergranular separations that occur in the hotter portions of the fuel. Intergranular separations are present in the unmelted regions of specimens in which central melting occurred and in some of the specimens in which there was no melting. Figure II.17 shows examples of the various

morphologies identified as directional and nondirectional intergranular separations. The initial stages in the formation of directional and nondirectional patterns of intergranular separations are shown in Figs. II.17A and II.17B, respectively. These separations are cracklike in appearance. The patterns of intergranular separation are nearly fully developed in Figs. II.17C and II.17D.

Fig. II.17. Optical Micrographs of Intergranular Separations in DEH-tested Fuel. Neg. No. MSD-190928.

Note that, for both the directional and nondirectional patterns almost every grain is adjacent to a separation, and the separations are interlinked over long distances through the structure. Examples of varying degrees of development of both classes of intergranular separation are present in a single test specimen. In some cases, as shown in Fig. II.17E, wide variations in the amount of directionality occur over distances of $\sim 100 \mu\text{m}$. For individual test specimens, an appreciable degree of directionality was present in $\sim 40\%$ of the area that showed intergranular separations. Directional separations tended to be oriented perpendicular to the pellet radius. However, the local orientation direction was influenced by the presence of stress concentrators, such as large pores and cracks, and near contact points between adjacent pellet fragments. The density of intergranular separations also increased in the vicinity of sources of stress concentration.

A final example of a form of intergranular separation, shown in Fig. II.17F, was present in fuel that was heated to temperature close to the melting point. These separations occupy grain boundaries of all orientations with equal frequency. The rounded outline of these features contrasts with the sharper edges and cracklike appearance of the separations shown in Figs. II.17A-E. The structure shown in Fig. II.17F is characteristic of high-temperature grain-boundary swelling.

Fig. II.18

Radial Profiles of Swelling and Pore-Solid Surface Area Produced by DEH.
ANL Neg. No. 306-77-317.

The structure shown in Fig. II.17F is characteristic of high-temperature grain-boundary swelling.

The extent of intergranular separation can be quantitatively characterized by measuring the values of pore volume fraction, V_V^P , and pore solid surface area, $S_V^{\alpha P}$, as a function of radial position. If pretest values for these parameters are also known, the volume increase (i.e., swelling) and the new pore-solid surface area produced during the transient can be calculated. Radial profiles of swelling and new surface area per gram of fuel, $(S_g^{\alpha P})^{xs}$, are plotted in Fig. II.18. These graphs show that the radial gradients are quite large and that, for some tests, the maximum surface-area increase may occur at an intermediate radial position. A plot of $(S_g^{\alpha P})^{xs}$ against % swelling (Fig. II.19) shows that a single relationship describes the data for several radial positions in each of seven DEH tests.

Volume averaging of the radial profiles of swelling and $(S_g^{\alpha P})^{xs}$ was used to calculate the quantitative stereology parameters listed in Table II.9 for seven DEH tests. The volume averaged parameters were also used for the quantitative relationships between gas release and microstructural change developed in Sec. B.3.c. Because of the extensive interlinkage

of the intergranular separations, escape paths exist for much of the fission gas that diffuses to the grain boundaries.

Fig. II.19

New Pore-Solid Surface Area vs Percent Swelling
for All Radial Positions in Seven DEH-tested
Pellets. ANL Neg. No. 306-77-319.

Evidence that fission-gas effects were in part responsible for the formation of the separations was obtained by examining the posttest microstructure in the SEM. Specimens were prepared by fracturing DEH-tested specimens at room temperature. Figure II.20 shows a series of micrographs along a radial traverse of the specimen from Test 24. Radial temperature profiles at various times during this test are shown in Fig. II.6. At the outermost radial positions (see Figs. II.20A and II.20B), intergranular fracture predominated. The grain surfaces are covered with fission-gas bubbles ranging in size from the limit of resolution (~ 30 nm) to ~ 50 nm. Where transgranular fracture occurred, no bubbles are visible.

A marked increase in bubble size and density (values of $\sim 10^{13}$ bubbles/mm³ were determined for the area in Fig. II.20A) has occurred over the as-irradiated microstructures shown in Figs. II.15 and II.16. Average bubble size increases toward the center of the pellet, i.e., where transient temperatures were higher, as shown in Figs. II.20C and II.20D. When the bubbles reach ~ 0.2 μ m in diameter, continued bubble coalescence results in the formation of sinuous grain-boundary channels.

Grain-boundary-channel formation and bubble growth appear to proceed at different rates on individual grain faces, even for faces on the same grain, as seen in Figs. II.20C and II.20E. The arrow in Fig. II.20F indicates a grain face on which the channels have coalesced to form a nearly complete separation between two adjacent grains. More extensive grain-boundary separation has occurred at the location shown in Fig. II.20G. The predominant fracture mode has changed to transgranular at this position. Note that, in addition to the intergranular separations, intragranular bubbles with diameters of 0.1 to 0.3 μ m are present. This sequence of micrographs shows how intergranular separations may be formed by a mechanism involving growth and coalescence of fission-gas bubbles.

A

 $R/R_0 = 0.92$

B

C

 $R/R_0 = 0.83$

D

E

 $R/R_0 = 0.75$

F

 $2 \mu\text{m}$ $R/R_0 = 0.53$

G

 $20 \mu\text{m}$

Fig. II.20

Scanning-electron Fractographs
at Four Radial Positions in a
Robinson Fuel Pellet after
DEH Test 24. (A-D) Neg.
No. MSD-190926; (E-G) Neg.
No. MSD-190925.

c. Relationships between Gas Release and Microstructural Change

Since there is a direct and nearly linear relationship between $(S_g^{\alpha P})_{xs}$ and the amount of swelling, as shown in Fig. II.19, a relationship between gas release and swelling will have nearly the same form as a relationship between gas release and surface area. Therefore, to avoid repetition, only the relationship between gas release and swelling will be discussed.

Plots of xenon release against both V_V^P and swelling are presented in Figs. II.21 and II.22. The significance of the similarity of the two plots arises from the fact that the initial and final microstructural conditions must be known to calculate swelling, while V_V^P is a measure of the final microstructural state only. Comparison of the two graphs in Figs. II.21 and II.22 indicates that no information is lost by plotting xenon release against a parameter that measures only the final microstructural state. Figure II.22 shows that ~2% gas release was obtained without any microstructural change. For tests in which swelling was less than 17-19%, an approximately linear increase in gas release with swelling was observed. For swelling in excess of 18%, the percentage gas release increased more rapidly with increasing swelling.

The change in slope in the curve of percent release versus percent swelling is believed to be the result of the rapid interlinkage of the

Fig. II.21. Percent Xenon Release as a Function of Post-DEH Volume-averaged Pore Volume Fraction. ANL Neg. No. 306-77-313.

Fig. II.22. Percent Xenon Release as a Function of Volume-averaged Swelling. ANL Neg. No. 306-77-310.

intergranular separations when the extent of separation reaches a critical value. The interlinked porosity allows fission gas on grain boundaries and edges to escape. For swelling below the critical value, gas release occurs mainly by diffusional mechanisms. This explanation is consistent with studies of percolation probabilities⁷⁻⁹ in which extensive long-range interconnection of a second phase in a matrix is predicted if the volume fraction of the second phase exceeds a critical value.⁷⁻⁹ Interconnection of the second-phase particles is negligible for volume fractions less than the critical value. Critical volume fractions of 0.17,⁷ 0.20,⁸ and 0.36,⁹ reported in the literature, depend on the size distribution and shape of the second-phase particles. Figure II.21 shows that the critical volume fractions for the interconnection of intergranular separations is 0.17-0.19, which is within the range of reported values.

4. Gas-release Mechanisms (S. M. Gehl and J. Rest, MSD)

Empirical correlations were developed between gas release and transient temperature history and between gas release and microstructural change for the initial series of PCM-type tests that suggest some preliminary conclusions about the mechanisms of gas release.

The relationships between gas release and maximum temperature parameters contrast with the weaker relationships between gas release and integral parameters (see Fig. II.10). Appreciable scatter was observed for the PCM-type tests, for which the test conditions were similar. For tests covering a wider range of initial conditions and heating rates, an even larger amount of scatter would be expected. Since the integral or time-at-temperature parameters determine the extent to which diffusional processes occur, the weak relationships between gas release and the integral parameters suggest that diffusional processes are not the only mechanisms that affect gas release. This conclusion is supported by GRASS calculations, which underpredict the measured gas releases when only diffusional processes are considered.

GRASS is also being used to model the formation of the grain-surface channels, shown in Fig. II.20, and the venting of these channels to grain-edge tunnels. GRASS calculations using these models indicate that large quantities of gas can be transported to the grain edges if the high-temperature bubble mobilities are allowed to approach the limiting case of surface diffusion control (see Sec. A.1). Figure II.3 shows that, if the gas in the grain-edge tunnels is assumed to be released by the interlinkage of the intergranular separations, the total gas-release values calculated by GRASS are in good agreement with the experimentally measured release fractions.

At present, GRASS does not include a model for the formation of the cracklike intergranular separations shown in Fig. II.17. A comparison of the GRASS calculations with the posttest microstructures indicates that the radial zones in which large amounts of gas were calculated to be transported to the grain boundaries coincide with the regions in which measured swelling exceeded the assumed value for long-range porosity interconnection (19%). The assumption that all the grain-edge gas escapes leads, in most cases, to a slight overprediction of the measured gas release. An additional source for the apparent discrepancies between the calculated and experimental gas releases in Fig. II.3 is the uncertainty in the DEHTTD-calculated temperature profiles used in GRASS.

The series of micrographs in Fig. II.20 indicates that intergranular separations can form by the diffusion-controlled processes of growth and coalescence of fission-gas bubbles. However, optical micrographs of areas in the same specimen (Fig. II.23) show that extensive cracklike intergranular separations are present at radial positions where the predominant features on the intergranular fracture surfaces are isolated fission-gas bubbles. That

A. $R/R_0 = 0.93$

B. $R/R_0 = 0.70$

0.20 mm

Fig. II.23. Optical Micrographs of a Polished Section at Selected Radial Positions in a Robinson Fuel Pellet after DEH Test 24. Compare with the scanning-electron fractographs in Fig. II.20. Neg. No. MSD-190927.

is, the gradual processes of bubble growth and coalescence to form channels, and channel coalescence to form separations, can be interrupted by the more rapid process of crack propagation. The stresses responsible for cracking are the result of the applied axial load, differential thermal expansion, and the pressurization of intergranular fission-gas bubbles.

The intergranular propagation of the cracks is, in part, due to the reduction in grain-grain contact area by the intergranular fission-gas bubbles. Crack propagation is accompanied by varying amounts of plastic flow in the adjacent grains, depending on the local temperature. For temperatures close to the melting point, stresses are relieved by plastic flow without cracking, and the bubble and channel coalescence processes are wholly responsible for intergranular separation. Therefore, the formation of the network of intergranular separations responsible for fission-gas release is a result of both diffusion-controlled (bubble growth and coalescence) and mechanical (cracking) mechanisms.

5. Calculation of Transient Radial Temperature Profiles (S. M. Gehl and J. Rest, MSD)

The DEHTTD computer code can be used to calculate the transient temperature histories for DEH tests by solving the time-dependent heat-transfer equation. The code contains expressions taken from the literature for the temperature dependence of thermal and electrical conductivity of UO_2 , and requires as input the time histories of pellet voltage, current, power, and surface temperature. The radial temperature profile is initialized by solving the steady-state heat-transfer equation for the input values at time = 0. Note that, because voltage, current, and power are input to the code, and electrical conductivity is known, the problem is overdetermined. The extra information is used in the code to adjust the electrical-conductivity expression until a convergence criterion is met in a solution of the steady-state problem.

The electrical conductivity is altered by incrementing a variable called STOICH, which affects the electrical conductivity in the same way as fuel stoichiometry in the conductivity expressions of Aronson et al.¹¹ The variable STOICH is not the same as stoichiometry. STOICH accounts for effects such as fuel chemistry and microstructure in addition to stoichiometry. The value of STOICH obtained for the steady-state solution is carried over for use in the transient solutions of the heat-transfer equation.

The available literature expressions for thermal and electrical conductivity, i.e., the expressions used in the code, are appropriate for high-density, high-purity UO_2 . There are no comparable data for high-burnup fuel. In addition, the microstructure is continually changing during the DEH test as intergranular separations are formed. As a result, attempts at calculating temperature profiles of Robinson-fuel specimens using the literature data gave centerline temperatures that were in error by as much as 600°C. A

comprehensive solution to this problem, e.g., by detailed physical-property measurements on high-burnup fuel, was beyond the scope of the program. At the same time, some corrective measures were necessary. The problem was addressed by identifying two factors considered the most likely to cause the observed errors.

The first factor was the discrepancy between the literature values of electrical conductivity and the measured values for the Robinson fuel. Because the resistance of pellet stacks is routinely monitored during the initial preheat period prior to DEH testing, we can easily determine the effective electrical conductivity of the stacks, which consist of a high-burnup fuel pellet sandwiched between two depleted UO_2 pellets. As described in ANL-77-34 (p. 64), the measured conductivity values were fitted to an equation of the form

$$\sigma = \frac{A}{T} \exp\left(-\frac{B}{T}\right),$$

where σ is electrical conductivity, T is absolute temperature, and A and B are constants. At 300°C , the measured conductivities were approximately two orders of magnitude greater than the literature value used in the code. At 1000°C , the difference was approximately a factor of 3. The empirically determined electrical conductivities were incorporated into the DEHTTD code by adding a term to the existing expression. Because the empirical term was significant only at low temperatures and because the STOICH term already in the code gave a partial correction for the difference in electrical conductivity, the effect on calculations of radial temperature profiles was negligible.

The second factor thought likely to affect the accuracy of the heat-transfer calculation was the change in thermal conductivity during the transient caused by the formation of intergranular separations. Preliminary investigations showed that the calculated radial temperature profiles were sensitive to small changes in thermal conductivity. To model the formation of intergranular separations, we introduced a cracking factor into the thermal-conductivity equation to give an expression of the form

$$T'_k = (1 - F)T_k,$$

where F is the cracking factor, T_k is the original thermal conductivity calculated by the code, and T'_k is the modified value used for temperature-profile calculation.

The cracking factor is time-dependent, having the value zero at the start of transient heating, and increasing linearly with time to a maximum value at the time of maximum power input. The maximum value of the cracking factor is an adjustable parameter. For a physically realistic treatment, the maximum cracking factor would be a function of the extent of intergranular separation that occurs during each transient.

For the calculations presented in this report, the maximum cracking factor was assumed to be 0.5 for all tests. This value was determined by a trial-and-error matching of the calculated centerline temperature for Test 24 to the melting temperature of UO_2 (2850°C). This value was chosen because the posttest examination of the microstructure of Test 24 revealed that the very center of the fuel pellet had been heated to incipient melting. With a cracking factor of 0.5, the maximum centerline temperature calculated for Test 24 was 2824°C , which closely matched the microstructural result. In addition, with a maximum cracking factor of 0.5, the code accurately predicted the melt radius obtained in Test 26.

These correlations indicate that the use of the cracking factor allows accurate calculations of the transient radial temperature profiles. However, because the maximum cracking factor was held constant for all tests, the calculated temperatures may be in error for the tests in which small amounts of intergranular separation occurred. To account for possible errors arising from this source, the temperature-history parameters in Table II.11 have confidence intervals that assume potential errors of as much as 200°C in T_{CL} and $\bar{T}$ calculations. The resulting uncertainty in $(dT/dR)_{\text{max}}$ values range from 9 to 25%. Sensitivity studies indicate that the probable errors are in the range 50 - 150°C . The calculated temperatures can be continually refined by developing a relationship between the maximum cracking factor and a measure of intergranular separation, such as swelling or surface area.

Finally, the uncertainty in the temperature profiles calculated by the DEHTTD code compares favorably with the uncertainties in standard temperature calculations performed for in-reactor fuel rods.

6. ANL-EG&G Crosscheck Program (S. M. Gehl and L. R. Kelman, MSD)

The intergranular separations in the fuel from PBF Test IE-1 were found to be similar to the separations in DEH-tested Robinson fuel. Preparations are continuing for DEH testing and total fission-gas determination in the Saxton fuel.

Intergranular separations similar to those described for the DEH-tested Robinson fuel are present in the irradiated PWR fuel tested in the film-boiling region of rod 007 from Test PBF IE-1. Figure II.24 shows the appearance of the separations on polished plane sections through DEH- and PBF-tested fuel. The extent of directional-separation formation is less in the PBF-tested fuel, but the separations were otherwise similar. Scanning-electron fractographs of Saxton fuel from the film-boiling region of rod 007 of Test IE-1 (see Figs. II.25A-II.25C) show features nearly identical to those observed for DEH tests of Robinson fuel and presented in Fig. II.20. The intergranular fractures in Fig. II.25D exposed grains that have almost completely separated from their neighbors. An isolated contact area is visible only in

| 0.2 mm |

a. PBF-tested

b. DEH-tested

Fig. II.24. Intergranular Separations in PBF- and DEH-tested Fuel. ANL Neg. No. 306-77-162.

A

B

C

D

20 μm

Fig. II.25. Scanning-electron Fractographs of Fuel from Film-boiling Region of Rod 007 from Test PBF-IE-1. Compare with Fig. II.20. Neg. No. MSD-190924.

the center of this region. This nearly complete separation explains a grain-pullout problem experienced in the metallographic preparation of specimens from rod 007 (see ANL-77-34, pp. 67-68).

The series of photos in Fig. II.25 shows that the intergranular separations in Test PBF-IE-1 were formed by the growth and coalescence of fission-gas bubbles to form grain-boundary channels and the coalescence of channels to completely separate the faces of adjacent grains, the same sequence of events observed for the DEH-tested Robinson fuel. Rapid, discontinuous formation of intergranular separations by crack propagation also occurred during the PBF test, as evidenced by the cracklike directional separations in the PBF-tested fuel. However, since the density of directional separations was smaller for the PBF-tested fuel, intergranular separation by crack propagation was probably not as prevalent as in the DEH-tested material.

The similarity of the intergranular separations obtained under electrical and nuclear heating conditions demonstrates that the DEH technique can accurately simulate many of the effects of in-reactor transient heating. Since gas release during DEH transients is related to the formation of intergranular separations, a similar correlation probably exists for nuclear heating.

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III. MECHANICAL PROPERTIES OF ZIRCALOY CONTAINING OXYGEN

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A. Summary

Uniaxial and biaxial mechanical-property data on Zircaloy-4 are being obtained over a wide range of temperatures and strain rates 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.

Several tensile tests were performed on reactor-grade zirconium for comparison with the tensile properties of Zircaloy-2 and -4. The results indicate that the yield stress and ultimate tensile strength of zirconium are lower than those of Zircaloy-4 at temperatures between 300 and 1300 K. In general, the uniform and total strains for zirconium are higher than for Zircaloy. Total strains of ~1.75 in zirconium compared with ~1.3 in Zircaloy-4 were obtained at 1123 K, i.e., the temperature for maximum superplastic elongations in uniaxial tensile tests on Zircaloy.

Additional information was obtained on the tensile properties of composite Zircaloy-4/oxide specimens with total oxygen contents between 2.3 and 6.5 wt %. As the oxygen concentration in the material increases, the strength and ductility decrease for test temperatures between 300 and 1300 K due to cracking of the oxide layer and the smaller section thickness, which decreases the load-bearing capability of the α -phase material.

Additional information has been obtained on the diametral expansion and rupture characteristics of axially constrained Zircaloy-4 cladding in a steam environment. To determine whether the pellet-cladding diametral-gap distance had any effect on the deformation characteristics, burst experiments were performed on 300-mm-long tubes with gap distances of 0.2 and 0.5 mm at heating rates of 5, 45, and 115°C/s in steam. The effect of the pellet-cladding gap distance on the circumferential temperature variations in the cladding was also determined at the three heating rates. The radial-strain-localization parameter W was evaluated from measurements of the cladding thickness in the region of maximum circumferential strain, and the parameter was used to quantitatively correlate the circumferential strain at failure with the maximum circumferential temperature variation around the tube for the 0.5-mm gap. The maximum circumferential strain at burst temperatures in the vicinity of the 800°C strain peak was relatively insensitive to the gap distance in the range of 0.07-0.5 mm for heating rates of 5-10°C/s; however, for heating rates $\geq 45^\circ\text{C}$, the maximum circumferential strain increased by a factor of tv.

for the larger gap distance. The present results will provide the basis for interpretation of the deformation behavior of Zircaloy fuel cladding in nuclear-heated fuel rods.

A model has been developed for the deformation and fracture behavior of Zircaloy fuel cladding during reflood of the reactor core that follows the blowdown stage of a LOCA. The finite-element model, which incorporates the thermal and elastic properties of Zircaloy and the ZrO_2 surface layer, was used to perform the heat-transfer and stress analyses for the cladding during the rapid thermal quench from the Leidenfrost temperature to $\sim 100^\circ\text{C}$. A fracture analysis was made for Zircaloy cladding in which the oxide and α layers comprise 10, 20, and 40% of the wall thickness for initial crack distributions of 10 axial cracks uniformly distributed around the circumference of the tube and one circumferential crack every 1 and 10 cm along the axial length.

The initial results of the analysis indicate that the number and spacing of the cracks are important parameters in the crack-propagation behavior of the cladding under thermal-shock conditions. Based upon the fracture properties of oxidized Zircaloy and the assumptions used in the analysis, circumferential cracks are more likely to propagate through the wall thickness of heavily oxidized cladding, whereas axial cracks only rupture the oxide layer. Thermal-shock tests on oxidized Zircaloy cladding will be performed to correlate the initial crack distribution and the degree of oxidation with predictions of the crack-propagation behavior based on the analysis.

B. Mechanical Properties of Zirconium and Zircaloy-Oxygen Alloys

1. Comparison of the Tensile Properties of Zircaloy-4 and Reactor-grade Zirconium

To obtain a better understanding of the influence of the various alloying elements in Zircaloy on the high-temperature mechanical properties, a series of tensile tests was performed on reactor-grade zirconium for comparison with information developed on Zircaloy-2 and -4.¹ Cold-rolled and vacuum-annealed zirconium in the form of 0.625-mm-thick sheet with a recrystallized grain size of $\sim 5\ \mu\text{m}$ was purchased from Teledyne Wah Chang, Albany, OR. The composition of the material, which meets ASTM Specification B352-67, is given in Table III.1.

Tensile specimens with a 12.6-mm gauge length were fabricated in accordance with ASTM Specification E8-69. The specimens were reinforced in the grip region by spot-welding coupons of zirconium to the shoulder. Tensile tests were conducted in vacuo by the methods described in Ref. 2.

Figure III.1 shows the temperature dependence of the yield stress Zircaloy-4 and reactor-grade zirconium. The yield stress of both materials decreases as the temperature increases because of the dynamic recovery that

occurs at the higher temperatures. Because of the presence of alloying elements, Zircaloy-4 is stronger than zirconium at all temperatures. Figure III.1 shows the temperature dependence for the uniform strain of zirconium and Zircaloy-4. Below 1300 K, the uniform strain of Zircaloy-4 is lower than that of zirconium, except near 473 K, where dynamic strain-aging phenomena are significant.

TABLE III.1. Composition of
Reactor-grade Zirconium

Element	ppm	Element	ppm
Al	<35	O	940
B	0.2	Mn	<25
Cd	<0.2	Ni	<35
C	65	N	28
Cr	50	Si	<40
Co	<10	Ti	<50
Cu	10	W	<25
Hf	61	U	1.5
Fe	362	Zr	Balance
H	12

Fig. III.1. Effect of Temperature on the Yield Stress of Reactor-grade Zirconium and Zircaloy-4. ANL Neg. No. 306-77-349.

Fig. III.2. Uniform Strain as a Function of Temperature for Reactor-grade Zirconium and Zircaloy-4. ANL Neg. No. 306-77-335.

Alloying elements influence the uniform strain as a result of their effect on the yield stress (flow stress) and the work-hardening rate. The uniform strain in a tensile test is commonly defined by the Considère criterion $d\sigma/d\epsilon = \sigma$, i.e., equating the work-hardening rate and the flow stress. If alloying elements increase the work-hardening rate of the material more than the flow stress, the uniform strain will increase; the converse situation will produce a decrease in the uniform strain. The results in Figs. III.1 and III.2 suggest that the alloyed zirconium, i.e., Zircaloy-4, falls into the latter category, except at ~ 473 K, where the uniform strain is the same for both materials (Fig. III.2). Above 1300 K, grain growth, in addition to alloy composition, complicates the uniform strain behavior.

Figure III.3 shows the influence of temperature on the total strain of zirconium and Zircaloy-4. Both materials exhibit superplastic strain maxima at ~ 1123 and 1300 K. The larger ductility peak near 1123 K for pure zirconium can be attributed to a greater extent of dynamic recovery and a higher strain-rate sensitivity in the unalloyed material.

Fig. III.3

Temperature Dependence of the Total Strain of Reactor-grade Zirconium and Zircaloy-4. ANL Neg. No. 306-77-332.

Figure III.4 shows the strain-rate dependence of the uniform strain of zirconium and Zircaloy-4 at 1123 K. The uniform strain of zirconium is always greater than that of Zircaloy-4. Figure III.5 shows the effect of strain rate on the total strain for the two materials. The total strain of zirconium is relatively independent of strain rate, whereas the ductility of Zircaloy-4 decreases as the strain rate increases. This observation can be explained in terms of the mode of accommodation of grain-boundary sliding in the two

materials at 1123 K. The accommodation of grain-boundary sliding in Zircaloy-4 at high strain rates ($\geq 5 \times 10^{-4} \text{ s}^{-1}$) occurs by dislocation slip, whereas at lower strain rates, diffusional processes become important.³ The results in Fig. III.5 imply that, at strain rates $\geq 6 \times 10^{-4} \text{ s}^{-1}$, accommodation of grain-boundary sliding by dislocation slip is more efficient, and hence, the ductility of pure zirconium is higher than that of Zircaloy-4. Apparently, accommodation by diffusional flow is more effective in the "impure" material at lower strain rates and results in a higher ductility for Zircaloy-4.

Fig. III.4. Effect of Strain Rate on the Uniform Strain of Reactor-grade Zirconium and Zircaloy-4 at 1123 K. ANL Neg. No. 306-77-350.

Fig. III.5. Total Strain as a Function of Strain Rate for Reactor-grade Zirconium and Zircaloy-4 at 1123 K. ANL Neg. No. 306-77-351.

Figure III.6 shows the effect of specimen geometry (a factor-of-two change in specimen width) on the temperature dependence of the total strain of Zircaloy-4. The ductility peak near 1123 K is higher for the specimen with a larger width-to-thickness (w/t) ratio; however, the ductility peak at $\sim 1300 \text{ K}$ is independent of specimen geometry. This observation can be attributed to the anisotropy of deformation of the α -phase material at the lower temperature. The close-packed-hexagonal structure of α -phase Zircaloy has a limited number of slip systems, and most of the slip occurs in the a -axis rather than the c -axis direction, which results in significant anisotropy. The high-temperature β phase has a body-centered-cubic (bcc) structure, which has multiple slip systems and therefore is less anisotropic. Since the initial texture of the Zircaloy sheet consists of grains with most of the basal planes parallel to the plane of sheet, longitudinal elongation of the specimen gauge length is accompanied by a substantial decrease in the specimen width at temperatures in the α -phase region. The change in the thickness of the specimen is small because the c -axis direction is parallel to the thickness direction. In β -phase Zircaloy above 1300 K, elongation of the gauge length occurs with a significant decrease in specimen thickness and a minimal change in width.

Because of the "geometrical constraint" associated with a low width-to-thickness ratio for α -phase material, smaller total elongations and elongated grains are observed for specimens with $w/t = 5$ compared with larger elongations and an equiaxed structure for $w/t = 10$ at 1123 K. The

Fig. III.6. Temperature Dependence of the Total Strain of Zircaloy-4 Specimens with Two Width-to-Thickness Ratios. ANL Neg. No. 306-77-343.

decreases the total and uniform strains as a result of the brittle oxide layer and oxygen diffusion in the α -phase material. Figure III.9 shows the effect of temperature on the total and uniform strains for composite specimens with 2.3 wt % oxygen. The uniform strain exhibits a maximum at 673 K, and the total strain exhibits maxima at ~673 and 973 K. The minimum in the total strain at ~823 K

elongation peak at ~1300 K is not dependent on specimen geometry because the bcc structure is less anisotropic. The above reasoning may explain the difference in the relative magnitude of the α -phase superplastic strain maxima in uniaxial³ and biaxial⁴ deformations. Since the width direction in a tensile specimen is analogous to the tube axis in a biaxial tube-burst specimen, in which tensile stresses are imposed in the axial and hoop directions, a reduction in the width of a Zircaloy tensile specimen is easier. Consequently, the α -phase strain maximum is larger under uniaxial-loading conditions. Differences in the grain size and texture of the tube and sheet materials and strain rate are other factors that can influence the uniaxial and biaxial strain maxima.

2. Mechanical Properties of Zircaloy-4/Oxide Composite Specimens

The effect of oxygen on the uniform and total strains of Zircaloy-4/oxide composite specimens is shown in Figs. III.7 and III.8, respectively, for several temperatures between 300 and 1073 K. Oxygen de-

Fig. III.7

Effect of Oxygen Concentration on the Uniform Strain of Zircaloy-4/Oxide Composites at Several Temperatures. ANL Neg. No. 306-77-337.

Fig. III.8. Oxygen-concentration Dependence of the Total Strain of Zircaloy-4/Oxide Composites at Several Temperatures. ANL Neg. No. 306-77-336.

Fig. III.9. Effect of Temperature on the Uniform and Total Strain of Zircaloy-4/Oxide Composite Specimens with 2.3 wt % Oxygen. ANL Neg. No. 306-77-333.

may be due to dynamic strain aging. The elongation peak at ~973 K, with essentially zero uniform strain, may be indicative of superplastic behavior of α -phase Zircaloy with an oxygen-concentration gradient in the material, since the surface-oxide layer cracks at very small strains during deformation at temperatures ≤ 1300 K.⁴

Figure III.10 shows the effect of temperature on the yield stress and ultimate tensile strength of Zircaloy-4/oxide specimens with 2.3 wt % oxygen, both of which decrease as the temperature increases. The uniform and total strains and the ultimate tensile strength are compared in Figs. III.11-III.13,

Fig. III.10. Ultimate Tensile Strength and Yield Stress as a Function of Temperature for Zircaloy-4/Oxide Composites with 2.3 wt % Oxygen. ANL Neg. No. 306-77-344.

Fig. III.11. Temperature Dependence of the Uniform Strain of Zircaloy-4/Oxide Composites with 2.3, 5.2, and 6.5 wt % Oxygen. ANL Neg. No. 306-77-348.

Fig. III.12. Total Strain as a Function of Temperature for Zircaloy-4/Oxide Composites with 2.3, 5.2, and 6.5 wt % Oxygen. ANL Neg. No. 306-77-334.

Fig. III.13. Temperature Dependence of the Ultimate Tensile Strength of Zircaloy-4/Oxide Composites with 2.3, 5.2, and 6.5 wt % Oxygen. ANL Neg. No. 306-77-345.

respectively, for Zircaloy-4/oxide composites with 2.3, 5.2, and 6.5 wt % oxygen. The decrease in the strength of the composites with an increase in oxygen concentration is associated with cracking of the oxide layer and the smaller cross section of the load-bearing α -phase material.

C. Biaxial Deformation and Rupture Characteristics of Zircaloy-4 Cladding in Steam

1. Effect of Pellet-Cladding Diametral-gap Distance on the Rupture Characteristics at Temperatures in the α -phase Region

Results reported previously⁵ indicated that temperature nonuniformities in the cladding during transient-heating experiments decreased the circumferential strain at failure for initial internal pressures ≥ 7 MPa, which resulted in bursts in the α -phase region. At lower internal pressures, the cladding typically ruptures at temperatures in the two-phase ($\alpha + \beta$) or β -phase regions, and oxidation of the Zircaloy rather than the temperature nonuniformity limits the strain at failure.

During the present reporting period, the effect of diametral-gap distance between the alumina pellets and the cladding on the degree of temperature nonuniformity and the maximum circumferential strain at failure was investigated at heating rates of 5, 45, and 115°C/s. Diametral-gap distances of 0.2 and 0.5 mm were used in the present experiments for comparison with previous results⁵ obtained with a smaller diametral gap of 0.07 mm. Figure III.14 shows a qualitative comparison of the temperature distributions, as evidenced by the "hot" and "cold" spots, produced in two tubes with diametral-gap distances of 0.07 and 0.5 mm for a heating rate of 45°C/s and an internal

Fig. III.14. Comparison of the Temperature Distributions in Zircaloy, as Evidenced by the "Hot" and "Cold" Regions, at a Heating Rate of 45°C/s and an Internal Pressure of 9.7 MPa for Pellet-Cladding Diametral-gap Distances of (A) 0.07 and (B) 0.5 mm. ANL Neg. No. 306-77-330.

pressure of 9.65 MPa. The tube with the smaller diametral-gap distance (Fig. III.14A) exhibits a higher density of "cold" spots that are smaller in size than those obtained with a 0.5-mm gap (Fig. III.14B). The probability of developing a relatively large uniform-temperature zone, as shown by the arrow in Fig. III.14B, is greater for a larger diametral gap. The temperature nonuniformities in the cladding with 0.2- and 0.5-mm diametral gaps were similar.

Figure III.15 shows the maximum recorded local temperature difference for cladding temperatures between ~ 750 and 800°C as a function of the heating rate for diametral-gap distances of 0.07, 0.2, and 0.5 mm. At low heating rates ($\leq 20^{\circ}\text{C/s}$), the temperature variations in the cladding are small (~ 40 - 60°C) and relatively insensitive to the diametral-gap distance, whereas at higher heating rates, the temperature nonuniformity increases as the diametral-gap distance decreases. The large temperature differences associated with the higher heating rates result from conduction

and radiation heat transfer between the cladding and pellets; heat flow in the radial and axial directions by convection in the gas annulus and conduction

Fig. III.15

Relationship between the Maximum Local Temperature Difference at Temperatures near 800°C and the Heating Rate for Pellet-Cladding Diametral-gap Distances of 0.07, 0.2, and 0.5 mm. ANL Neg. No. 306-77-331 Rev. 1.

through the tube cross section, respectively, is comparatively small during the short-time periods at the higher heating rates.

The relationships between the maximum circumferential strain and the burst temperature for the different diametral-gap distances are shown in Figs. III.16-III.18 for heating rates of 5, 45, and 115°C/s, respectively. At the lowest heating rate, the diametral-gap distance does not have a strong effect

Fig. III.16. Maximum Circumferential Strain vs Burst Temperature for Axially Constrained Zircaloy-4 Cladding with Pellet-Cladding Diametral-gap Distances of 0.07, 0.2, and 0.5 mm at a Heating Rate of 5-10°C/s in Steam. ANL Neg. No. 306-77-340.

Fig. III.17. Maximum Circumferential Strain vs Burst Temperature for Axially Constrained Zircaloy-4 Cladding with Pellet-Cladding Diametral-gap Distances of 0.07, 0.2, and 0.5 mm at a Heating Rate of 45°C/s in Steam. ANL Neg. No. 306-77-352.

Fig. III.18

Maximum Circumferential Strain vs Burst Temperature for Axially Constrained Zircaloy-4 Cladding with Pellet-Cladding Diametral-gap Distances of 0.07, 0.2, and 0.5 mm at a Heating Rate of 115°C/s in Steam. ANL Neg. No. 306-77-342.

on the circumferential strain; however, as the heating rate increases ($\geq 45^\circ\text{C/s}$) the circumferential strain for burst temperatures near 800°C is approximately a factor of two higher for the larger gap distances. No significant difference was observed in the circumferential strains at failure for cladding with 0.2- and 0.5-mm diametral-gap distances. This is consistent with the results in Fig. III.15 for the variation of the maximum temperature difference with heating rate for the two gap sizes.

The increase in the circumferential strain from ~ 30 to 60% for gap distances of 0.07 to 0.2-0.5 mm, respectively, at heating rates of 45 and 115°C/s is due to the larger regions of more uniform temperature in the case of the larger diametral-gap distances (e.g., Fig. III.14), although the maximum local temperature variations, as evidenced by the bars in Figs. III.16-III.18, are not strongly dependent on the gap size for burst temperatures near 800°C . Based upon the relatively small differences between the circumferential strain behavior of pellet- and mandrel-constrained cladding (0.07-mm diametral gap) for burst temperatures in the $(\alpha + \beta)$ or β -phase regions,⁵ the effect of diametral-gap distance on the maximum circumferential strain will be quite small for burst temperatures $\geq 850^\circ\text{C}$.

Figures III.19-III.21 show the effect of diametral-gap distance on the relationship between the maximum circumferential strain and the initial pressure for heating rates of 5, 45, and 115°C/s , respectively. At heating rates $\geq 45^\circ\text{C/s}$, the diametral-gap distance has a significant effect on the circumferential strain for internal pressures between 8 and 13 MPa, whereas no

Fig. III.19. Maximum Rupture Circumferential Strain as a Function of Initial Internal Pressure for Axially Constrained Zircaloy-4 Cladding with Pellet-Cladding Diametral-gap Distances of 0.07, 0.2, and 0.5 mm at a Heating Rate of $5\text{--}10^\circ\text{C/s}$ in Steam. ANL Neg. No. 306-77-341.

Fig. III.20. Maximum Rupture Circumferential Strain as a Function of Initial Internal Pressure for Axially Constrained Zircaloy-4 Cladding with Pellet-Cladding Diametral-gap Distances of 0.07, 0.2, and 0.5 mm at a Heating Rate of 45°C/s in Steam. ANL Neg. No. 306-77-346.

Fig. III.21. Maximum Rupture Circumferential Strain as a Function of Initial Internal Pressure for Axially Constrained Zircaloy-4 Cladding with Pellet-Cladding Diametral-gap Distances of 0.07, 0.2, and 0.5 mm at a Heating Rate of 115°C/s in Steam. ANL Neg. No. 306-77-347.

effect of gap distance was observed at the lowest heating rate. At low heating rates, i.e., $\leq 15^\circ\text{C/s}$, and internal pressures ≥ 5 MPa, circumferential strains between ~ 0.6 and 0.8 are likely, and, for a limited internal pressure range of 6-8 MPa, strains as high as 1.0 are possible in a steam environment. With regard to the ballooning behavior of Zircaloy cladding under nuclear-heating conditions, one data point from the in-reactor test program at the Karlsruhe Research Center, Federal Republic of Germany,⁶ is shown in Fig. III.19. Although this test is not sufficient for a comparison between in-reactor and laboratory measurements, the in-reactor result, obtained at a heating rate of $\sim 10^\circ\text{C/s}$, an internal pressure of 4.9 MPa, and a diametral gap of 0.19 mm, is consistent with the results in Fig. III.19.

In general, the burst opening in the cladding is in the axial direction, since the tangential stress is higher than the axial stress during the final stages of ballooning. However, for pellet-constrained cladding that ruptures at temperatures in the α - or predominantly α -phase region, tangential rupture orientations, as shown in Fig. III.22, are occasionally observed. The internal pressures and heating rates for the specimens shown in Fig. III.22 were 11.2 and 9.7 MPa and 51 and 11°C/s, respectively. The tangential failure orientation results from a large circumferential temperature gradient in the ballooning region in which the higher temperature was opposite the burst opening.

The ballooning sequence that leads to this type of failure is as follows. Initially, as the hotter side of the tube begins to balloon, the material undergoes tangential expansion as well as axial contraction because wall thinning is not favorable during the deformation of α -phase cladding. At this stage, axial contraction of the cooler side of the tube is negligible and the tube bends in the direction of the ballooning region (Fig. III.22B). As a result of the bending moment on the tube, the cooler side also deforms and the axial stress in this region can exceed the tangential stress; this results in the type of failure shown in Fig. III.22.

2. Evaluation of the Strain-localization Parameter for α -phase Cladding Burst under Different Test Conditions

In Ref. 5, a method was developed to correlate the maximum circumferential strain with a quantitative measure of the extent of radial strain

localization in the tube wall for burst temperatures $\leq 830^{\circ}\text{C}$, such that the circumferential strain decreases as the radial strain becomes more localized. A radial-strain-localization parameter W was defined by

$$W = 1 - \frac{1}{2\pi} \int_0^{2\pi} \frac{e_r(\theta)}{e_r^0} d\theta, \quad (1)$$

where

θ = tangential angle from the fracture tip,

$e_r(\theta) = \ln(h/h_0)$, i.e., the true local fracture strain at different θ positions around the circumference of the cladding,

and

h_0, h = the initial and local thicknesses, respectively, of the cladding at the region of maximum circumferential strain.

Fig. III.22. Pellet-constrained Zircaloy-4 Cladding with a 0.5-mm Diametral Gap after Rupture in Steam That Shows Tangential Rather Than the Typical Axial Burst Openings. The initial pressures and heating rates are (A) 11.2 MPa and 51°C/s , and (B) 9.7 MPa and 11°C/s . ANL Neg. No. 306-77-329.

The W parameter has been evaluated for 24 tests in which the diametral-gap distance was 0.5 mm. These data have been included in the plot shown in Fig. III.23 and are in good agreement with the previous results, which encompass a wide range of experimental conditions. Figure III.24 shows a

comparison of the W parameter as a function of the maximum recorded circumferential temperature difference for cladding with 0.5- and 0.07-mm diametral-gap distances. The burst temperatures for these tests at heating rates between 5 and 130°C/s lie within ±25°C of the ~800°C strain peak (see Figs. III.16-III.18).

Fig. III.23

Maximum Circumferential Strain vs Radial-strain-localization Parameter for α -phase Cladding Burst under Different Test Conditions at Temperatures between 700 and 850°C. Test Conditions can be identified from the following code abbreviations: V = vacuum environment, S = steam environment, X = preoxidized cladding in steam at a transient-heating rate of 115°C/s for 7 s and burst in vacuo, U = unconstrained cladding, M = mandrel-constrained cladding with a 2.5-mm axial gap, P = pellet-constrained cladding with a 2.5-mm axial gap, F = specimen length of 153 mm, L = specimen length of 300 mm, D = pellet-cladding diametral gap of 0.07 mm, Z = pellet-cladding diametral gap of 0.5 mm, numerals denote the heating rate in °C/s, and 0 represents isothermal stress-rupture tests. ANL Neg. No. 306-77-143 Rev. 1.

Fig. III.24

Radial-strain-localization Parameter as a Function of Maximum Circumferential Temperature Difference in Zircaloy Tubes with Pellet-Cladding Diametral-gap Distances of 0.07 and 0.5 mm Ruptured in Steam at Temperatures Near 800°C. ANL Neg. No. 306-77-140 Rev. 1.

For heating rates of ~ 5 - 10°C/s , which are typical of the values during refill from the "evaluation-model" worst-case analysis of a double-ended cold-leg guillotine break in a postulated LOCA for a reference pressurized water reactor,⁷ W parameters between 0.5 and 0.55 would be expected from Fig. III.24. The corresponding values of the maximum circumferential strain from Fig. III.23 are ~ 0.75 - 0.9 for this range of heating rates and pellet-cladding diametral-gap distances between ~ 0.07 and 0.5 mm. Peak cladding temperatures obtained from a "best-estimate" analysis, which is based upon currently accepted "best-judgment" mathematical models for a LOCA in a pressurized water reactor at its most probable state, are below $\sim 650^\circ\text{C}$, and the maximum circumferential strain predicted for the cladding, if failure occurs, would be ≤ 0.4 .

Fig. III.25. Effect of Heating Rate on the Maximum Circumferential Strain Near 800°C for Unconstrained and Mandrel- and Pellet-constrained Zircaloy-4 Cladding in Steam. ANL Neg. No. 306-77-338.

3. Effect of Heating Rate and Type of Axial Constraint on Maximum Circumferential Strain at Temperatures in the α -phase Region

Figure III.25 shows the heating-rate dependence of the maximum circumferential strain at burst temperatures near 800°C for unconstrained and mandrel- and pellet-constrained cladding (2.5-mm axial gap) in steam. The influ-

ence of heating rate is more pronounced between ~ 5 and 50°C/s and is greatest for pellet-constrained cladding with a small diametral gap and least for unconstrained cladding. The range of circumferential strains between ~ 0.3 and 1.5 for burst temperatures near 800°C indicated in this figure are the result of a number of interdependent variables, e.g., the diametral-gap sizes and the axial and circumferential temperature variations in the cladding, the effects of which have been quantified in this investigation. These factors, rather than the heating mode per se, i.e., direct versus indirect (internal or external), determine the circumferential strain at failure.

D. Analysis of Crack Growth in Oxidized Zircaloy Cladding during Thermal-shock Conditions

The ballooning characteristics of Zircaloy cladding under postulated LOCA conditions in LWR's have been investigated in considerable detail. Far less attention has been given to the behavior of the cladding during reflood that follows the blowdown stage of a LOCA. In the latter stages of a LOCA transient, the cladding undergoes a quench as a result of the rapid increase in heat-transfer coefficient between the cladding and the steam at the so-called Leidenfrost temperature. The exact value of the Leidenfrost temperature for oxidized Zircaloy cladding at 0.1-MPa total pressure is not accurately known, but is expected to be $\sim 500^{\circ}\text{C}$. A rapid quench can introduce large stresses that may result in brittle fracture of the cladding, particularly when it is heavily oxidized.

Realistic modeling of the deformation and fracture behavior of Zircaloy cladding following a LOCA is quite complex. Not only are the loadings (both mechanical and thermal) not fully known, but the mechanical properties of the anisotropic Zircaloy-oxide composite material are dependent on the time-temperature transient experienced by the cladding during the blowdown phase. Any model of cladding deformation during the thermal quench will necessarily involve several simplifying assumptions. As a first assumption in this study, the cladding is oxidized to the extent that it is relatively brittle compared with as-received Zircaloy, and consequently, linear-elasticity theory can be used in the analysis. Second, only thermal stresses are considered, and the temperature distribution is assumed to be axisymmetric so that the stress distribution is also axisymmetric. Third, the cladding is assumed to be infinitely long so that a generalized plane-strain deformation mode is applicable. Fourth, all interactions between cracks are ignored in the fracture analysis.

The cladding has been modeled by a series of ring elements, each of which can have temperature-dependent anisotropic heat-conduction and elasticity properties. The same finite-element model is used to perform both the heat-transfer and stress analyses. As high strain-rate data become available, the material properties, as a function of oxygen content, can be incorporated into the model. A general flow chart for the code is shown in Fig. III.26. The initial temperature distribution in the cladding is assumed to be known. At every time increment, a heat-transfer analysis is performed with either the boundary-temperature history or a specified heat-transfer coefficient and environment-temperature history.

Initially the cladding is assumed to be crack-free. A specified number of equally spaced axial cracks, to be input by the user, is initiated as soon as the hoop stress in the outermost element exceeds the ultimate hoop strength. Similarly, a predetermined number of equidistant circumferential cracks per unit axial length is initiated at the outer boundary as soon as the axial stress in the outermost element exceeds the axial ultimate strength. Any interactions

between cracks are ignored in the analysis. The size of the outermost finite element determines the size of the initial crack. It is further assumed that the hoop stresses are zero in all elements that extend from the outermost element to the element containing the current axial-crack tip. This assumption is reasonable if the number of axial cracks in the cladding is large so that the hoop continuity from the outer surface to the crack tip is essentially destroyed. A similar assumption is made for the axial stresses and circumferential cracks.

Fig. III.26. Flow Diagram for Computation of Temperature Distribution and Fracture of Zircaloy Cladding under Thermal-shock Conditions. ANL Neg. No. 306-77-328.

At every time increment, the initial total potential energy of the system is determined by

$$PE = U + GE + SE,$$

(2)

where U is the elastic strain energy in the cladding (the stress concentration due to the cracks is ignored), GE is the additional strain energy due to the presence of the cracks, and SE is the surface energy. The quantity GE is calculated from the crack-extension force for a finite-width anisotropic strip with a single edge crack, which is subjected to a bending moment. The crack is allowed to extend to the next element if it results in a decrease of the total potential energy.

In case the potential energy is lowered, the crack is extended to succeeding elements until the total potential energy equals the initial total potential energy. All rate effects on crack propagation are ignored, and the extension of the cracks from element to element at any step is assumed to occur instantaneously. This assumption is not too severe, provided the thermal shock diffuses through the material at a much lower rate than the propagation of elastic waves through the material.

The thermal and elastic properties of Zircaloy and ZrO_2 were obtained from the MATPRO handbook.⁸ The anisotropic properties of α -Zircaloy were obtained from Ref. 9. Because of insufficient property data, the oxide layer was assumed to be isotropic. The analysis was carried out for tubes in which the thickness of the ZrO_2 and oxygen-embrittled α layers at the outer surface comprised 0, 10, 20, and 40% of the tube wall. The outer diameter (OD) and inner diameter (ID) of the cladding were 10.90 and 9.63 mm, respectively.

Figures III.27-III.30 show the temperature distribution across the cladding wall for various oxide and α -layer thicknesses at several times during the transient for a thermal shock in which the OD surface temperature decreased from 500 to 100°C and the ID surface temperature was maintained at 500°C. The maximum effective rate at which the temperature profile (thermal shock) moves through the oxide or metal layer is ~ 5 m/s, which is much less

Fig. III.27. Temperature Distribution across the Wall of an Unoxidized Zircaloy Tube after a Thermal Shock from 500 to 100°C; Heat-transfer Coefficient = ∞ . ANL Neg. No. 306-77-361.

Fig. III.28. Temperature Distribution across the Wall of an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 10% of the Wall Thickness after a Thermal Shock from 500 to 100°C; Heat-transfer Coefficient = ∞ . ANL Neg. No. 306-77-358.

Fig. III.29

Temperature Distribution across the Wall of an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 20% of the Wall Thickness after a Thermal Shock from 500 to 100°C; Heat-transfer Coefficient = ∞ . ANL Neg. No. 306-77-357.

Fig. III.30

Temperature Distribution across the Wall of an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 40% of the Wall Thickness after a Thermal Shock from 500 to 100°C; Heat-transfer Coefficient = ∞ . ANL Neg. No. 306-77-359.

than the velocity of elastic waves through the material. Also note that the steady-state temperature distribution is reached in ~ 0.04 s. Because of the poor thermal conductivity of the oxide layer, most of the temperature gradient through the wall occurs in the oxide, which is subjected to a high tensile stress. Figures III.27-III.30 were calculated on the basis that the temperature at the outside surface was reduced to 100°C at time $(t) = 0$.

A second series of transient analyses was performed for a uniform initial cladding temperature of 500°C. The temperature of the steam at the outer surface was set at 100°C at $t = 0$, and the heat-transfer coefficient between the cladding and steam was assigned a value of 180 kW/m²·K. The resultant time-dependent temperature distributions are shown in Figs. III.31-III.33. Note that the heat-transfer coefficient is sufficiently large so that the

Fig. III.31

Temperature Distribution across the Wall of an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 10% of the Wall Thickness after a Thermal Shock from 500 to 100°C; Heat-transfer Coefficient = 180 kW/m²·K. ANL Neg. No. 306-77-360.

Fig. III.32

Temperature Distribution across the Wall of an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 20% of the Wall Thickness after a Thermal Shock from 500 to 100°C; Heat-transfer Coefficient = 180 kW/m²·K. ANL Neg. No. 306-77-366.

Fig. III.33

Temperature Distribution across the Wall of an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 40% of the Wall Thickness after a Thermal Shock from 500 to 100°C; Heat-transfer Coefficient = 180 kW/m²·K. ANL Neg. No. 306-77-367.

Figures III.34-III.36 show the growth of an axial and circumferential crack in Zircaloy cladding in which the oxide and α layers comprise 10, 20, and 40% of the tube wall, respectively. The cladding contained 10 axial cracks distributed uniformly around the circumference of the tube and one circumferential crack per centimeter along the axial length. Note that the axial cracks never penetrate below the oxide-layer thickness. The circumferential cracks, on the other hand, grow to ~65, 55, and 40% of the wall thickness for total oxide and α -layer thicknesses of 10, 20, and 40% of the tube wall, respectively.

Fig. III.34

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 10% of the Tube Wall; Initial Crack Depth and Distribution: 0.05 mm and 10 Axial Cracks and One Circumferential Crack per Centimeter of Tube Length. ANL Neg. No. 306-77-362.

Fig. III.35

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 20% of the Tube Wall; Initial Crack Depth and Distribution: 0.05 mm and 10 Axial Cracks and One Circumferential Crack per Centimeter of Tube Length. ANL Neg. No. 306-77-363.

temperature distribution quickly approaches that of the previous case. The true heat-transfer coefficient between Zircaloy and steam at the Leidenfrost temperature is not known; however, a value in the range of 60-300 kW/m²·K is a reasonable estimate. The effect of varying the heat-transfer coefficient on the thermal-shock behavior of oxidized Zircaloy cladding will be investigated in the future.

An initial fracture analysis was made with the assumptions that the cladding is initially stress-free and the surface energies for the oxide and underlying metal are 45 and 100 J/m², respectively. A series of drop-weight impact tests will be conducted to obtain a more quantitative estimate for the surface-energy values.

Fig. III.36

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 40% of the Tube Wall; Initial Crack Depth and Distribution: 0.05 mm and 10 Axial Cracks and One Circumferential Crack per Centimeter of Tube Length. ANL Neg. No. 306-77-356.

Figures III.37-III.39 show the growth of an axial and circumferential crack for an initial distribution of 10 axial cracks and one circumferential crack every 10 cm along the axial length. Although the axial cracks do not propagate, as in the previous case, the circumferential cracks almost penetrate the tube for oxide and α -layer thicknesses of 20 and 40% of the wall. The number and spacing of the cracks, produced during oxidation and deformation under transient-heating conditions, will be an important factor in crack propagation due to thermal shock in the reflood stage of a LOCA. Thermal-shock tests on oxidized Zircaloy cladding will be performed to correlate the initial crack distribution with crack-propagation behavior for comparison with these predictions.

Figure III.40 shows the effect of a decrease in the thermal-shock temperature from 500 to 400°C on axial and circumferential crack growth for cladding in which the oxide and α layers comprise 20% of the wall thickness for a tube with an initial distribution of 10 axial cracks and one circumferential crack every 10 cm along the length. The circumferential cracks traverse 75% of the wall, compared with 95% in the case of a thermal shock upon cooling from 500°C.

Fig. III.37

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 10% of the Tube Wall; Initial Crack Depth and Distribution: 0.05 mm and 10 Axial Cracks and One Circumferential Crack per 10 cm of Tube Length. ANL Neg. No. 306-77-364.

Fig. III.38

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 20% of the Tube Wall; Initial Crack Depth and Distribution: 0.05 mm and 10 Axial Cracks and One Circumferential Crack per 10 cm of Tube Length. ANL Neg. No. 306-77-368.

Fig. III.39

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 40% of the Tube Wall; Initial Crack Depth and Distribution: 0.05 mm and 10 Axial Cracks and One Circumferential Crack per 10 cm of Tube Length. ANL Neg. No. 306-77-365.

Fig. III.40

Crack Growth as a Function of Time after a Thermal Shock from 400 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 20% of the Tube Wall; Initial Crack Depth and Distribution: 0.05 mm and 10 Axial Cracks and One Circumferential Crack per 10 cm of Tube Length. ANL Neg. No. 306-77-370.

The results thus far were based on a model with 35 finite elements and a 0.05-mm-wide outermost element (initial crack size). The number of finite elements was increased to 69 so that the outermost element (initial crack width) decreased to 0.025 mm, and the crack growth during a thermal shock upon cooling from 500°C was determined for cladding in which the oxide and α layers comprised 20% of the wall thickness. The result is shown in Fig. III.41. Figure III.42 shows a similar analysis for a larger circumferential crack spacing, i.e., one crack per 10-cm length. A comparison of Fig. III.35 with Fig. III.41 and Fig. III.38 with Fig. III.42 shows that the initial crack size does

Fig. III.41

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 20% of the Tube Wall; Initial Crack Depth and Distribution: 0.025 mm and 10 Axial Cracks and One Circumferential Crack per Centimeter of Tube Length. ANL Neg. No. 306-77-369.

Fig. III.42

Crack Growth as a Function of Time after a Thermal Shock from 500 to 100°C for an Oxidized Zircaloy Tube in Which the ZrO_2 and α Layers Comprise 20% of the Tube Wall; Initial Crack Depth and Distribution: 0.025 mm and 10 Axial Cracks and One Circumferential Crack per 10 cm of Tube Length. ANL Neg. No. 306-77-355.

not greatly influence subsequent crack growth for these crack distributions because the crack-extension forces are quite small for cracks of this size and the forces result in a negligible contribution to the total potential energy. The situation would be different if the initial crack size was larger.

The initial results of this analysis indicate that the number and spacing of the cracks are important parameters in the crack-propagation behavior of the cladding under thermal-shock conditions. Based upon the fracture properties of oxidized Zircaloy and the assumptions used in the analysis, circumferential cracks are more likely to propagate through the wall thickness. This could result in multiple fractures of the cladding in heavily oxidized regions, whereas axial cracks only rupture the oxide layer. The fracture behavior of oxidized Zircaloy fuel cladding is dependent on a number of parameters such as the ultimate strength, surface energy, oxide-layer thickness, degree of thermal shock, and number and spacing of the cracks. For example, a high ultimate tensile strength of the oxide layer will delay the initiation of cracks; however, a higher stored elastic-strain energy at fracture will result, which, together with a low surface energy, might drive the cracks, once initiated, rapidly through the wall. The effect of these parameters will be studied in detail in the future.

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IV. STEAM-EXPLOSION STUDIES

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A. Combined Vapor Explosions (R. E. Henry, L. M. McUmber, and E. L. Martinec, Jr., RAS)

Previous experiments with FREON-22 and mineral oil, as reported in Ref. 1, demonstrated that a system pressure of 0.1 MPa resulted in vigorous physical explosions with maximum peak pressures of about 2.2 MPa. When the ambient pressure was increased to 0.22 MPa, no explosive behavior was observed. The mechanistic reasons for this abrupt termination of the explosive behavior are given in Ref. 1, and these basically relate to the propagation phase of the explosion and, in particular, the ability of the system to continually fragment itself as the explosive event progresses.

To provide an additional test sequence with a different exploding fluid, experiments were conducted with FREON-11 and mineral oil. Freon-11 has a higher boiling point (20°C) than Freon-22 (-41°C) and a higher critical temperature as well (198° versus 96°C). The higher critical temperature means that Freon-11 will have a significantly higher homogeneous nucleation temperature range as shown in Table IV.1 for a system pressure of 1 atm (0.1 MPa). As discussed in Ref. 2, a nucleation rate required for large-scale

TABLE IV.1. Nucleation Rates for Freon-11^a

Interface Temperature, °C	P_v , bars	σ , dynes/cm	r , Å	J , cm ⁻³ .s ⁻¹	Waiting Time for 1 cm ³
140	17.1	4.75	56	1.7×10^{-14}	1.9×10^6 yr
142	17.6	4.58	52	4.4×10^{-7}	26 days
144	18.3	4.36	48	7.3×10^1	1.4×10^{-2} s
146	19.1	4.16	44	1.1×10^8	8.9×10^{-9} s
148	19.9	3.93	40	6.6×10^{13}	1.5×10^{-14} s
150	20.5	3.77	37	1.3×10^{17}	7.8×10^{-18} s
155	22.3	3.29	30	1.5×10^{24}	6.6×10^{-25} s
160	24.3	2.83	23	2.1×10^{28}	4.7×10^{-29} s
165	26.4	2.38	18	4.6×10^{30}	2.2×10^{-31} s

^aConversion factors: 1 bar = 0.1 MPa; 1 dyne = 10^{-5} N;
1 Å = 10^{-10} m.

apor explosions would correspond to an interface temperature of 155-160°C for Freon-11. This higher temperature range for homogeneous nucleation results in a much higher oil temperature for the onset of explosive events. Consequently, this provides an additional test of the viability of the interface temperature model.

These experiments were conducted in the experimental apparatus shown in Fig. IV.1 in which the Freon is dropped vertically into the interaction vessel, which contains about 900 mL of mineral oil at the prespecified temperature. Since these experiments involve very high oil temperatures, the system is kept in an inert argon environment and the Freon is added immediately before the test is to be executed. The instrumentation in this system includes piezoelectric pressure and force transducers to monitor the explosive event and initial temperatures of both the Freon and mineral oil.

Fig. IV.1. Experimental Apparatus for Large-scale Vapor Explosions in a Contained Volume. ANL Neg. No. 900-77-69.

The experimental data are tabulated in Table IV.2 for initial temperatures, amounts of interactants, amount of oil lost from the vessel, maximum interaction pressure, and system impulse derived from the pressure and force measurements. These results are graphically illustrated in Figs. IV.2

and IV.3 for maximum system pressures and system impulse, respectively. The onset of explosions occurs for an interface temperature between 160 and 170°C, which is in good agreement with the analytical prediction, and at very high interface temperatures there is a cessation of explosive events. This is in agreement with the high-temperature results reported in Ref. 1, where it

TABLE IV.2. Table of Runs

Date	Ambient Pressure, MPa	Temperature, °C			Initial Amount, mL		Amount of Oil Released, mL	Maximum Interaction Pressure, MPa	Impulse, N·s		
		Oil	Freon-11	Interface	Oil	Freon-11			Force	P _{bottom}	P _{middle}
7/14	0.25	326	49	220	900	100	-	-		No event	
7/18	0.25	289	34	191	900	100	-	-		No event	
7/19	0.25	270	36	180	900	100	-	-		No event	
7/19	0.25	260	44	177	900	100	-	-		No event	
7/21	0.1	260	26	170	900	200	350	0.34	1.2	-	0.9
									1.1	-	0.2
7/22	0.1	282	23	183	900	200	750	0.91	16.8	6.0	8.9
7/22	0.1	300	23	194	900	200	460	0.38	6.4	-	3.6
7/25	0.1	310	23	200	900	200	750	0.67	20.7	-	17.5
7/27	0.1	317	24	205	900	200	450	0.53	3.8	2.5	2.3
7/27	0.1	341	24	220	900	200	900	0.63	6.7	7.6	6.0
7/28	0.1	360	24	231	900	200	900	0.91	12.9	16.8	14.3
7/28	0.1	360	24	231	900	50	-	-		No event	
7/29	0.1	340	24	219	900	100	?	3.22	30.6	20.2	18.9
7/29	0.1	339	25	219	900	50	-	-		No event	
8/2	0.1	331	23	213	900	50	-	-		No event	
8/2	0.1	330	23	212	900	50	900	0.91	24.3	20.3	18.0
8/5	0.1	310	23	200	900	50	-	Explosion: not recorded			
8/5	0.1	310	23	200	900	50	650	0.71	18.0	11.6	12.5
8/8	0.1	290	23	138	900	50	550	0.29	6.7	-	3.7
8/12	0.1	220	24	145	900	50	-	-			
8/12	0.1	240	23	157	900	50	170	-			
8/15	0.1	260	23	169	900	50	270	0.21	1.2	-	0.2
8/15	0.1	277	23	180	900	50	?	0.20	1.9	-	1.2
8/16	0.1	266	23	173	900	50	340	0.17	0.6	-	0.7

Fig. IV.2. Maximum Interaction Pressures for Freon-11 and Mineral Oil. ANL Neg. No. 900-77-1284.

Fig. IV.3. System Impulses for Freon-11 and Mineral Oil. ANL Neg. No. 900-77-1283.

was concluded that, for an interface temperature in the near vicinity of the thermodynamic critical point for the exploding liquid, the nucleation rate should become so overwhelming that explosive interactions will be terminated. Although this temperature is somewhat higher than the thermodynamic critical point, it is certainly in close agreement with the value of 198°C for Freon-11. As illustrated in Table IV.2, systems with ambient pressures of 0.25 MPa did not observe any explosive interactions. This is also in agreement with the behavior reported in Ref. 1 for Freon-22/mineral-oil systems as well as Freon-22/water systems.

In summary, experiments have been conducted with a different Freon, one having a much higher normal boiling point than the thermodynamic critical point than those previously used. The interface temperature-spontaneous nucleation model provides an excellent representation of the temperature range at which explosive interactions will be observed. The previously observed behavior for high-pressure and high-interface-temperature terminations was again observed in this system.

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