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HEAT TRANSFER COEFFICIENTS BETWEEN FUEL AND CLADDING IN OXIDE FUEL RODS

BY
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BREEDER REACTOR DEVELOPMENT OPERATION

GENERAL ELECTRIC

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ABSTRACT

Coefficients of heat transfer between fuel and cladding were determined from in-pile irradiations. Values of 1150, 750, and 520 Btu/h-ft²-°F were obtained for initial nominal fuel-cladding gaps of 0.002, 0.004, and 0.010 inch in the power range from 17.5 to 20.0 kW/ft.

Heat transfer coefficients obtained for mixed oxide fuel rods irradiated in a fast flux for burnups ranging from 1500 to 50,000 MWd/Te are also presented.

SECTION I

INTRODUCTION

Knowledge of fuel behavior under in-pile conditions is essential for any reactor design. This knowledge is particularly important at power levels resulting in fuel center temperatures near incipient melting.

The effective size and thermal resistivity of the diametral gap between fuel and cladding is an important design variable since it is related to the initial cold gap size, cladding expansion characteristics, fuel density and expansion characteristics, and the fuel operating temperature. Other variables also affect the thermal resistivity of the fuel-cladding gap: the composition of the gas bond; surface roughness; hardness and strength of the cladding and fuel; and reactions between

fission products, cladding, and fuel at the interface. With these variables in mind, an in-pile experiment was conceived and carried out to determine the gap conductance as a function of initial cold gap size. In addition, information on the effect of initial cold gap size on fuel-cladding interaction, fuel restructuring, and operating temperature was obtained.

The experiment was conducted with close control of fuel temperature and power through the use of thermocouples both in the fuel and exterior to the cladding. Irradiation of all fuel pins was carried out simultaneously to eliminate possible discrepancies in irradiation exposure and operating conditions.

SECTION II

SUMMARY

Heat transfer coefficients between fuel and cladding were determined as a function of fuel-cladding gap size from in-pile irradiation experiments. Fuel pins with nominally 0.002, 0.004, and 0.010 inch diametral gaps between fuel and cladding were irradiated simultane-

ously in the GETR. The heat transfer coefficients between fuel and cladding were determined from post-irradiation metallographic measurements of the extent of equiaxed and columnar grain growth and of the extent of fuel melting. Heat transfer coefficients of

1140, 750, and 520 Btu/h-ft²-°F were calculated for diametral gaps of 0.0022, 0.0042, and 0.0101 inches, respectively.

Metallographic examination of the fuel-cladding gap showed that the 0.0042 and 0.0101 inch diametral gaps had decreased approximately 0.002 and 0.005 inches, respectively. The 0.0022-inch gap decreased approximately 0.001 inch.

Heat transfer coefficients were also determined for mixed oxide fuel rods irradiated in a fast flux to burnups ranging from 1800 to 50,000 MWd/Te. The heat transfer coefficients ranged from approximately 2000 Btu/h-ft²-°F to 1000 Btu/h-ft²-°F for diametral gap sizes ranging from 0.001 to 0.005 inch. In the high burnup fuel pins no significant change was observed in the heat-transfer coefficient even though the gap between fuel and cladding was filled with fuel or fission products, or both.

SECTION III

DESCRIPTION OF EXPERIMENT

3.1 PHYSICAL DESCRIPTION

Four argon-bonded fuel pins containing (Pu_{0.2}U_{0.8})O_{1.98} and having as-fabricated diametral fuel-cladding gaps of 0.0022, 0.0042, 0.0101 and 0.0040 inch were irradiated simultaneously in the trail cable facility of the General Electric Test Reactor (GETR). The fuel pins were filled with argon gas to simulate the conductivity of the gas bond in an operating fuel pin after considerable burnup. One pin (0.0040-inch gap) was instrumented with a tungsten-rhenium thermocouple to measure fuel center temperature and to establish a correlation with the irradiated fuel microstructure.

Each fuel pin was designed to operate with a maximum cladding temperature of 1300°F and to a peak power of 23 kW/ft. Cladding temperatures were controlled by varying the thickness of the capsule thermal dams. Capsule power was monitored by both thermal dam Δt and capsule cooling water Δt measurements.

Fuel pin and capsule drawings are shown in Figures 3-1 and 3-2.

Inlet and outlet cooling water temperatures were recorded by thermocouples. Thermocouples were also placed in the annuli between the thermal dams around fuel pins No. 2 and No. 3. These thermocouples were used to monitor the capsule performance and calculate the power generated by the fuel pins. NaK was used to provide a thermal bond between the concentric thermal dams and containment cans.

A unique feature of this capsule was the double containment of the fuel pins. This design made it possible to detect any bubbles in the NaK bond since the assembly was easily x-rayed prior to assembly into the capsule.

3.2 FUEL FABRICATION

The powder for the fuel pellets was made by the standard co-precipitation process for mixed oxide powder.⁽¹⁾ All pellets were made from a single powder batch.

Pellets were made by pressing to a green density of 46% to 56% of theoretical density

FIGURE 3-1. CAPSULE DRAWING

and sintered for 3 hours in a dry 94% helium—6% hydrogen atmosphere at 1600°C. The pellets produced had densities between 95% and 97% of theoretical density. The oxygen-to-metal (O/M) ratio of the pellets was in the range of 1.976 to 1.986. Oxygen-to-metal ratio was determined by the technique

described by W. L. Lyon.⁽²⁾ Density values were determined by pellet volume and weight measurements.

Fuel fabrication parameters for each fuel pin are listed in Table 3-1.

TABLE 3-1

FUEL FABRICATION PARAMETERS FOR CAPSULE E1B

PARAMETER	FUEL PIN			
	E1B-1	E1B-2	E1B-3	E1B-4
%PuO ₂	20.96	20.96	20.96	20.96
%U-235	0.22	0.22	0.22	0.22
Inside Diameter of Clad (in.)				
Minimum	0.2218	0.2200	0.2278	0.2219
Maximum	0.2221	0.2206	0.2280	0.2221
Diametral Gap (mils)				
Range	4-4.5	1.8-2.5	9.7-10.8	3.6-4.9
Average	4.25	2.2	10.1	4.2
Fuel Weight (gr)	7.087	7.836	7.915	7.996
Fuel Density (% TD)				
Range	95.6-96.1	95.8-97.3	95.7-96.7	96.1-96.4
Average	95.9	96.4	96.1	96.2
Smeared Density (% TD)	92.1	94.3	91.5	92.4
Oxygen-to-Metal Ratio				
Range	1.984-1.986	1.987-1.984	1.976-1.984	1.976-1.984
Average	1.985	1.980	1.980	1.980
Cladding Material	347 SS*	347 SS*	347 SS*	347 SS*

*Stainless Steel

SECTION IV

IRRADIATION OF EXPERIMENT

The initial 40-hour irradiation was carried out at an average linear power of approximately 18 kW/ft based on cooling water Δt measurements. The operating power of each fuel pin determined from the thermal dam thermocouple Δt 's are listed in Table 4-1 for the initial 40-hour irradiation periods.* As can be seen from Figure 4-1, which shows the

operating power history of fuel pin E1B-3, the power was maintained quite constant during the 40-hour irradiation.

*The final irradiation period was shortened from 5 hours to 1-1/2 hours because of a reactor scram.

TABLE 4-1

FUEL PIN POWER CALCULATED FROM CAPSULE CALORIMETRY

<u>Fuel Pin</u>	<u>Linear Power (kW/ft)</u>			
	<u>Thermal Dam Thermocouples</u>		<u>Cooling Water Δt</u>	<u>Burnup Analysis</u>
	<u>40 hour Irradiation</u>	<u>5 hour* Irradiation</u>		
E1B-1	17.7	19.55	17.7**	NA***
E1B-2	18.0	20.05		20.7
E1B-3	17.8	19.55		NA
E1B-4	17.1	18.75	17.7**	18.7

*The 5-hour irradiation was shortened to 1.5 hours because of a reactor scram.

**Determined during 40-hour irradiation only.

***NA not analyzed.

FIGURE 4-1. POWER VERSUS TIME IN REACTOR FOR FUEL PIN EIB-3

SECTION V

DESTRUCTIVE EXAMINATION

The capsule was removed from the reactor, gamma scanned and disassembled. After removal from the capsule, each fuel pin was physically examined and accurately measured to determine any changes in length and diameter. The pre- and post-irradiation measurements are listed in Table A-1, and diametral changes observed are discussed in Appendix A.

Subsequent to the physical examination, the fuel pins were punctured to determine percent gas release and composition of the fission gas. Each fuel pin was then sectioned and the fuel and cladding examined metallographically. The procedures and observations made during each part of the examination are given in Appendix A.

SECTION VI

DISCUSSION OF RESULTS

6.1 CALCULATION OF GAP CONDUCTANCE

Conductance values were calculated using the data of Baily, et al.,⁽³⁾ for the integrated conductivity of mixed oxides of plutonium and uranium.* The diameters for equiaxed and columnar grain growth were used as temperature reference points across the diameter of each fuel pin. The molten boundary was also used as a temperature reference point when melting occurred. Temperatures used for the formation of equiaxed and columnar grains were 1650°C and 2150°C, respectively. These temperatures are in close agreement with temperatures determined experimentally both in and out-of-pile by Hausner and Nelson⁽⁴⁾ and by Potter and Elyard⁽⁵⁾ for UO_2 . Values reported for the melting temperature of unirradiated $(\text{Pu}, \text{U})\text{O}_{2+x}$ by Lyon and Baily⁽⁶⁾ were used to calculate integral conductivities to melting.

Although a fixed temperature was assumed in the calculations for the temperatures at which equiaxed and columnar grain growth occurs, it is important to realize that the rate of growth and final grain size observed may be dependent on the following:

- Initial grain size of the material.
- The amount and distribution of impurities and porosity.
- Oxygen-to-metal ratio.
- Stress gradients.
- Temperature and time at temperature.
- Irradiation exposure.

The effects of many of these variables on the formation of equiaxed and columnar grains have not been studied, especially at high temperature and in a temperature gradient. MacEwan and Hayashi⁽⁷⁾ have recently summarized the work which has been done to determine the effects of time and temperature, porosity, impurities, stoichiometry, compressive stress, and irradiation exposure on equiaxed grain growth in UO_2 . A brief summary of their conclusions is given below:

*A sample calculation, illustrating the methods used, is given in Appendix B.

- Effects of pores and inclusions:

Porosity has been found to inhibit movement of grain boundaries. Estimates of the reduction in the rate of grain growth showed that it could not have been reduced more than 30%.

- Effects of stoichiometry:

Increases in the O/U ratio from 2.0 to 2.23 have been found to increase the growth constant, k , for grain growth four orders of magnitude at 1325°C. In addition the activation energy decreased by 40% at a composition of $\text{UO}_{2.02}$.

- Effect of impurities:

Impurities initially present in UO_2 have been found to increase the activation energy for grain growth.

- Effect of compressive stress:

Stress or strain gradients have been found to accelerate grain growth in regions having the highest stress. However, uniformly applied compressive stresses did not affect the rate of grain growth, even if sufficient to produce plastic flow.

- Effect of irradiation exposure:

In-pile and post-irradiation experiments have shown that irradiation exposure inhibited grain growth under near isothermal conditions. Indirect evidence from in-pile fuel irradiations indicates that in a steep temperature gradient the effect of the thermal gradient decreases the inhibition of grain growth due to irradiation. Golyariov and Pravdyuk⁽⁸⁾ have reported that the temperature necessary to produce grain growth in UO_2 increases 200°C in the range of 10^{16} to 10^{19} fissions/cm³.

From the above it must be concluded that use of equiaxed grain growth as an absolute temperature indicator is probably not accurate to more than $\pm 200^\circ\text{C}$.

Little has been done to investigate the effects of impurities, irradiation, porosity, etc., on the growth of columnar grains. Potter and Elyard⁽⁵⁾ studied the effect of porosity and additions of metallic Ir and Y_2O_3 on the formation of columnar grains in UO_2 . The impurity additions were found to inhibit the development of columnar grains. Tests on high density samples showed that columnar grain growth is a function of both the porosity present and a diffusion mechanism. Temperatures determined for equiaxed and columnar grain growth were 1600°C and 2200°C.

6.2 GAP CONDUCTANCE - CAPSULE E1B

The effect of the cold gap sizes tested on fuel operating temperature is shown graphically in Figure 6-1. Fuel in pin E1B-3, which had an initial cold fuel cladding gap of 0.0101 inch, showed definite evidence of melting. The large central void in this pin is a result of solidification and shrinkage of molten fuel. Pin E1B-4, which had an 0.0042-inch cold gap showed no evidence of having melted, but a small central void (~ 0.010 inch in diameter) was formed indicating it operated with a higher central temperature than E1B-2 (0.0022-inch gap) in which no central void was observed to have formed even though it operated at a higher power.

Since the fuel pins in capsule E1B operated at two different power levels, conductance values calculated could reflect either the start-of-life, low power condition (~ 18 kW/ft), or the high power conditions (~ 20 kW/ft) experienced at the end-of-life. To determine which power conditions fixed the diameters of equiaxed and columnar grain growth, gap conductances were calculated for each case.

FIGURE 6-1. STRUCTURAL CHANGE AS A FUNCTION OF GAP SIZE

The gap size, operating power, fuel surface temperature, integral conductivity to the fuel center temperature, and the gap conductance determined for the start-of-life and end-of-life operating conditions for each fuel pin are listed in Table 6-1. The gap conductances are plotted against the cold "as fabricated" diametral gap sizes in Figure 6-2. In both cases (start and end-of-life) the conductances obtained for columnar grain growth for the same gap size are higher than those obtained for equiaxed grain growth.

Re-plotting the conductances obtained for the end-of-life operating conditions against the average fuel-cladding gap at the end of the irradiation, leads to a logical explanation of the differences observed between the conductances determined from the equiaxed and columnar grain growth diameters. Figure 6-3 shows the end-of-life conductances based on equiaxed grain growth diameters and those determined for the end-of-life columnar grain growth diameters. Based on knowledge of the temperature profiles in the fuel and their effect on equiaxed and columnar grain growth, as well as the molten boundary, a logical explanation of the data plotted in Figure 6-3 is possible.

The data in Table 6-1 indicate that the end-of-life center fuel temperatures are higher than at start-of-life, while the fuel surface temperatures are lower at end-of-life than at the beginning of the irradiation. In order for these two conditions to be achieved, the temperature profiles across the radius of the fuel pins would be required to change as shown in Figure 6-4. Under the conditions illustrated, if the columnar grain growth radius is assumed to be determined by the operating conditions at end-of-life, then the conductance value calculated for the columnar radius based on the start-of-life conditions would be higher than the "true" value at the beginning of the irradiation since the apparent radius of columnar grain growth at the end of the irradiation would be smaller than it actually

should be. The data plotted in Figure 6-3 indicate that the conductances determined from columnar grain growth based on start-of-life conditions are indeed higher than those based on end-of-life conditions. The above discussion requires that the columnar grain growth diameter at end-of-life be smaller than at the start of the irradiation. This requires restructuring of the columnar grains from a larger to a smaller diameter. Hausner, et al.⁽⁹⁾ have shown that restructuring of the columnar grains can occur in UO_2 irradiated at a high power followed by a lower power irradiation. Both Hausner, et al., and Bain⁽¹⁰⁾ have observed that evidence of fuel melting at high operating power can be eradicated by operating at lower powers. The excellent agreement between the conductances calculated based on the diameter of the molten boundary and the columnar, grain-growth diameter based on the end-of-life operating conditions indicates that the columnar grains did restructure.

An analysis similar to that carried out for columnar grain growth can be applied to the conductances determined from equiaxed grain growth diameters. If it is assumed that the equiaxed grain growth diameter measured during the post-irradiation examination was fixed at the start of the irradiation it can be seen from Figure 6-3 that conductances based on the end-of-life operating conditions would result in lower conductance values than at start-of-life. The data in Figure 6-3 based on equiaxed grain growth at start-of-life support this conclusion.

From the above discussion, it was concluded that:

- The end-of-life operating conditions fixed the diameters of columnar grain growth in the E1B capsule fuel pins.
- The start-of-life operating conditions fixed the diameters of equiaxed grain growth in the E1B capsule fuel pins.

TABLE 6-1

SUMMARY OF CAPSULE E1B DATA

FUEL PIN	TIME	GAP (mils)	POWER (kW/ft)	FUEL SURFACE TEMPERATURE (°C)		$\int_{500}^{T_c} kd\theta$ W/cm	GAP CONDUCTANCE (Btu/h-ft ² -°F)	
				EGG	CGG		EGG	CGG
E1B-2	Start of Life	2.2	18.0	1104	976	55.2	1140	1520
	End of Life	1.2	20.1	993	866	57.7	1643	3000
E1B-3	Start of Life	10.1	17.8	1652*	1213	67.9	524*	611
	End of Life	4.5	19.6	1652*	1452	68.6	599*	741
E1B-4	Start of Life	4.2	17.1	1348	1130	55.5	750	959
	End of Life	1.8	18.8	1267	1066	57.1	875	1910

*Since equiaxed grain growth was observed to occur at the surface of the fuel in pin E1B-3, the fuel surface temperature was set at 1650°C (3004°F). Since the fuel surface temperature could have been greater than 1650°C, the gap conductances calculated for the E1B-3 pin should be taken as maximum values.

FIGURE 6-2. GAP CONDUCTANCE VERSUS COLD DIAMETRAL GAP

FIGURE 6-3. GAP CONDUCTANCE VERSUS DIAMETRAL GAP

FIGURE 6-4. TEMPERATURE PROFILES IN EIB CAPSULE FUEL PINS AT START-OF-LIFE AND END-OF-LIFE

- The gap conductances determined from the equiaxed grain growth diameters for pre-irradiation fuel-cladding gaps of 0.0022, 0.0042 and 0.0101 inch were 1140, 750, and 520 Btu/h-ft²-°F.
- The gap conductances determined from the columnar grain growth diameters for post-irradiation gaps of 0.0012, 0.0018, and 0.0045 inch were 3000, 1910, and 740 Btu/h-ft²-°F.

6.3 GAP CONDUCTANCE—FAST FLUX IRRADIATIONS

Gap conductances for two groups of fuel pins irradiated in EBR-II to burnups of approximately 1500, and 50,000 MWD/Te are presented and discussed in this section. Pre- and post-irradiation fuel pin and operating parameters for the two groups of fuel pins are listed in Table 6-2. Gap conductances were determined using the methods discussed in Section 6.1.

Values of gap conductance determined from the diameters of equiaxed grain growth for each group of fuel pins are plotted against the cold "as-fabricated" gap sizes in Figure 6-5. Since these pins were irradiated in EBR-II (constant flux), the maximum temperatures in the rods were reached at the beginning of their irradiation. Therefore, the diameter of equiaxed grain growth should be representative of the temperature conditions in the fuel at start-of-life. As can be seen in Figure 6-5, three of the Group F2 fuel pins were found to have much lower gap conductances than the remaining fuel pins. One of the pins (F2P) operated at a much lower power than the remainder of the pins (9.9 kW/ft versus 16-17 kW/ft). Since the gap coefficient is a function of operating power, this pin would be expected to have a lower conductance than the high powered fuel pins.* The other two fuel pins

contained vibratory compacted powder fuel and, therefore, would be expected to exhibit low gap conductivities because of the decreased fuel-cladding contact area.

As discussed in Section 6.1 and 6.2, the diameter of columnar grain growth can change during irradiation because of restructuring and unless affected by irradiation, impurities, etc., and should be representative of end-of-life temperature conditions in the fuel. Gap conductances calculated based on the columnar grain growth diameters and end-of-life conditions are plotted in Figure 6-6. The data plotted in Figure 6-6 show the fuel-cladding gap of the high burnup fuel pins decreased to zero and the gap conductances fall within a narrow range from approximately 1100 to 1600 Btu/h-ft²-°F. The gap conductances obtained from the lower burnup Group F1 fuel pins also fall in the 1100 to 1600 Btu/h-ft²-°F range, indicating that the gap conductance approaches a constant value rather than increasing with burnup. This is in conflict with present models for gap conductance which predict an increase in the gap conductance as the fuel-cladding gap becomes smaller. Most gap conductance models consider only the area of contact and the interfacial pressure between surfaces as important parameters.

It is hypothesized that the observed trend toward a constant gap conductance at higher burnups is caused by the filling of the fuel-cladding gap with fuel and fission products, and the formation of oxide layers on the cladding surface. Three stages in the closing of the fuel-cladding gap with increasing burnup are shown in Figures 6-7 through 6-9. The pictures were taken from representative sections of fuel pins E1B-3, F1A, and F2Q, which had burnups of approximately 400, 1600, and 58,000 MWD/Te. At low burnup (see Figure 6-7), a fuel-cladding gap still exists after irradiation, even though the gap decreased to about one-half of the pre-irradiation size. At low burnup it appears that the change in the gap size is caused by

*The gap conductance is approximately proportional to power.

TABLE 6-2
FAST FLUX IRRADIATIONS - FUEL PIN AND OPERATING PARAMETERS

	PIN	FUEL	O/M	POWER (kW/ft)	DIAMETRAL GAP		HEAT FLUX i.d. [Btu/h-ft ² F(×10 ⁻⁵)]	CLADDING i.d. TEMP (°F)	CLADDING i.d. TEMP (°F)		BURNUP (MWd/Te)	CLADDING			DIAMETER OF STRUCTURE CHANGES			GAP CONDUCTANCE (Btu/h-ft ² °F)	
		DENSITY			Pre-	Post-			EGG	CGG		Type	o.d.	i.d.	EGG	CGG	Void	EGG	CGG
		(%)																	
Group One	F1A	94.1	1.998	16.1	0.0041	0.0030	9.53	1092	1128	978	1,750	347	0.2488	0.2203	0.1840	0.1311	0.0169	982	1360
	F1B	95.6	2.000	16.4	0.0012	0.0028	9.70	1101	925	714	1,615	347	0.2500	0.2204	0.1705	0.0968	0.0032	1626	
	F1C	96.1	2.026	16.8	0.0040	0.0013	9.94	1089	1144	1003	1,519	I-800	0.2510	0.2203	0.1839	0.1345	0.0178	1016	1330
	F1D	96.7	1.978	16.2	0.0018	0.0008	9.59	1069	953	-	1,596	I-800	0.2500	0.2203	0.1656	-	-	1412	-
	F1E	90.1	1.988	14.8	0.0042	0.0030	8.76	1063	-	970	1,615	316	0.2492	0.2203	-	0.1271	0.0308	-	1224
	F1F	85.3	1.986	16.4	-	0.0010	9.71	1083	-	-	1,711	I-800	0.2524	0.2203	0.1769	0.1481	0.0438	2002	
Group Two	F2A	95.7	1.999	16.1	0.0022	0	9.52	1069	922	914	56,200	347	0.2510	0.2204	0.1646	0.1196	0.0110	1530	1566
	F2B	96.8	1.998	16.4	0.0022		9.71	1048	938	970	57,100	316	0.2496	0.2203	0.1642	0.1259	0.0055	1445	1376
	F2E	96.7	1.975	16.7	0.0028		9.87	1056	984	1003	58,100	316	0.2510	0.2205	0.1699	0.1332	0.0059	1320	1306
	F2N	97.3	2.000	16.6	0.0052		9.81	1061	967	1025	57,700	347	0.2410	0.2205	0.1773	0.1319	0.0306	1020	1203
	F2P	96.7	1.999	9.9	0.0032		5.86	936	-	-	34,600	347	0.2504	0.2202	0.1220	-	-	713	-
	F2Q	96.4	2.000	16.7	0.0012		9.89	1085	958	944	58,100	347	0.2524	0.2202	-	0.1261	0.0105	2072	1531
	F2S	98.2	2.004	17.5	0.0008		10.37	1152	945	1128	61,000	347	0.2808	0.2200	0.1677	0.1470	0.0108	1656	1139
	F2U	89.7	1.986	16.3	0.0038		9.65	1074	953	1016	56,700	347	0.2509	0.2202	0.1798	0.1433	0.0470	1273	1227
	F2Z	90.2	1.983	16.1	0.0041		9.53	1043	1044	1013	56,100	316	0.2501	0.2203	0.1817	0.1411	0.0459	1098	1211
	F2W	83.8	1.990	14.1	-		8.36	1027	-	1188	49,200	347	0.2505	0.2200	0.1859	0.1503	0.0471	911	731
	F2Y	83.9	1.993	13.8	-		8.18	998	-	1195	48,200	316	0.2407	0.2200	0.1828	0.1482	0.0521	918	702

FIGURE 6-5. FAST FLUX IRRADIATIONS: GAP CONDUCTANCE VERSUS DIAMETRAL GAP FOR EQUIAXED GRAIN GROWTH

FIGURE 6-6. FAST FLUX IRRADIATIONS: GAP CONDUCTANCE VERSUS DIAMETRAL GAP FOR COLUMNAR GRAIN GROWTH

(a) FUEL-CLADDING INTERFACE SHOWING ASPERITES FORMED ON FUEL SURFACE AFTER 40 HOURS IN-PILE—AS POLISHED (100X)

(b) FUEL-CLADDING INTERFACE SHOWN IN INSET ABOVE—AS POLISHED (500X)

(c) SAME AS (b) ABOVE—ETCHED.

FIGURE 6-7. FUEL-CLADDING GAP, LOW BURNUP IRRADIATION

(a) FUEL-CLADDING INTERFACE SHOWING FUEL AND FISSION PRODUCTS—AS POLISHED (100X)

(b) FUEL-CLADDING INTERFACE SHOWN IN INSET ABOVE—AS POLISHED (500X)

(c) SAME AS (b) ABOVE — ETCHED

FIGURE 6-8. FUEL-CLADDING GAP, INTERMEDIATE BURNUP FUEL

FIGURE 6-9. FUEL-CLADDING GAP, HIGH BURNUP FUEL

fuel movement, either by cracking or plastic flow or a combination of both processes.

Figure 6-8 shows a region of the fuel cladding interface observed during the post-irradiation examination of fuel pin F1A. A layer of material was found in the gap consisting of fuel and white metallic particles. This deposit appears to be separate from the original fuel pellet surface and is more porous than the main body of fuel. Thus, the fuel-cladding gap appears to be partially filled not only by cracking or plastic flow of the fuel, or both, but by deposition of fuel and impurities or fission products by vapor transport.

The fuel-cladding interface of pin F2Q after a burnup of 58,000 MWd/Te is shown in Figure 6-9. As can be seen in the as-polished picture the fuel-cladding gap has completely disappeared. Upon etching, material is removed from the interface, and the original fuel pellet surface can be distinguished. Since the material at the fuel-cladding interface is much more reactive with the etchant than the fuel, it must have a different composition or a much higher surface area than the original fuel. Recent analysis of cladding samples at this laboratory have shown that Cs-137, a fission product, was present in significant amounts in the cladding of fuel pin F2Z.⁽¹¹⁾ Work carried out by Stalica and Seils⁽¹²⁾ using a microprobe has shown that the fission products Cs and Ba are present at the fuel-cladding interface and that changes in the concentrations of Ni, Fe, and Cr occurred at the inside surface of cladding containing irradiated mixed oxide fuel. Microprobe work on unirradiated compatibility samples of hyperstoichiometric fuel heated for 1000 hours at 1400°F by Lauritzen and Cummings⁽¹³⁾ has shown that a reaction layer is formed on the cladding surface composed of Cr₂O₃ and possibly an oxide of Fe. Wheeler⁽¹⁴⁾ and Skipper and Wootton⁽¹⁵⁾ have reported a decrease in the gap conductance between surfaces by factors of 4 or 5, or more, if one of the contacting surfaces became oxidized.

Based on the above discussion, the following conclusions are drawn:

- The gap conductances obtained from equiaxed grain growth diameters reflect the gap conductance at the start of life and increase with decreasing gap size as shown in Figure 6-5.
- Gap conductances determined from columnar grain growth diameters reflect the end-of-life operating conditions and decrease slightly with burnup to a constant value in the range of 1000 to 1600 Btu/h-ft²-°F.
- The deposition of fission products and fuel at the fuel-cladding interface affects the heat transfer across the interface.

6.4 COMPARISON OF GAP CONDUCTANCE DATA

Several authors have conducted experiments to determine the gap conductance in operating UO₂ fuel rods. Some of the most noted work is that reported by Bain and Robertson,⁽¹⁶⁾ Cohen,⁽¹⁷⁾ and Belle.⁽¹⁸⁾ Results from these three investigators and others have been discussed and summarized in the UO₂ Handbook by Belle.⁽¹⁸⁾ Shown in Figures 6-10 and 6-11 are curves of Belle's and Cohen's data and Bain and Robertson's data along with curves determined from the experiments discussed in this report. In general, the data obtained for the gap conductance values for mixed oxide are higher than those obtained for UO₂. One fact of significance is that the gap conductance values obtained from pins containing argon in the gaps are lower than for helium bonded pins. Belle concluded that the conductance across the fuel-cladding interface appeared to be independent of the atmosphere within the fuel pins. It should be noted that for Belle's data, plotted in Figure 6-10, the fuel-cladding gap was closed because of the difference in thermal expansion of the cladding and fuel. The experimental data reported herein indicate that the gap coefficients for argon and helium

FIGURE 6-10. COMPARISON OF GAP CONDUCTANCE DATA OF COHEN AND BELLE (from Belle⁽¹⁸⁾)

FIGURE 6-11. COMPARISON OF GAP CONDUCTIVITY DATA OF BAIN AND ROBERTSON (from Belle⁽¹⁸⁾)

atmospheres become comparable when the diametral gap is approximately 0.002 inch ($\frac{\text{Assembled Diametral Gap}}{\text{Fuel Diameter}} = 0.01$) or less. At the power which the E1B capsule and the Group F1 and F2 fuel pins operated, the fuel and cladding gaps would be closed because of expansion differences between the fuel and cladding for diametral gaps ranging from approximately 0.0020 to 0.0025 inch. Therefore, it may be that Belle observed no effect due to the type of gas used for bonding because of the interference fit between fuel and cladding. As discussed in Section 6.3, as the gap closes, the gap conductance tends to approach a constant value. This would tend to suggest that closure of the gap would result in the gas within the pin having little effect on the conductance across the interface. Bain, et al. (19) and Duncan (20) have also observed that the conductivity of the gap gas does affect the heat transfer across the fuel-cladding interface.

Several investigators have tried to develop models for use in calculating heat transfer coefficients across an interface. Cetinkale and Fishenden (21) and Ross and Stoute (22) have both done considerable work in this area. Shown in Figure 6-12 is a comparison of the curves of gap conductance determined from the pins discussed in this report and comparable gap conductances calculated using Ross and Stoute's model for a fresh fuel pin, assuming:

- Sodium coolant at 950°F
- Constant film coefficient of 25,000 Btu/h-ft²
- Fuel pellet diameter of 0.220 inch
- Cladding thickness = 0.015 inch
- Cladding yield strength of 20,700 psi
- Fractional expansion of the fuel and cladding varying with temperature according to the expressions:

$$\left(\frac{\Delta L}{L}\right)_{\text{fuel}} = 3.16 \times 10^{-4} + 3.716 \times 10^{-6} T + 9.01 \times 10^{-10} T^2$$

$$\left(\frac{\Delta L}{L}\right)_{\text{cladding}} = -6.253 \times 10^{-6} + 7.69 \times 10^{-6} T + 1.747 \times 10^{-9} T^2$$

- Gamma-neutron heating in the cladding was 5% of the total Btu/ft³ - sec leaving the fuel pin
- Coefficients used in the gap model were:

$$\alpha_0 = 0.91 \text{ ft}^{1/2}$$

$$\beta_0 = 2.5$$

$$\alpha_e = 18 \times 10^{-6} \text{ inch (cladding)}$$

$$\alpha_e = 36 \times 10^{-6} \text{ inch (fuel)}$$

$$\psi = 60,000 \text{ psi}$$

$$E = 28 \times 10^6 \text{ psi}$$

$$g_f + g_e = 3.28 \times 10^{-5} \text{ (pure He)}$$

- Linear power = 18 kW/ft

Gap conductances calculated using the Ross and Stoute model fall above the curves determined from equiaxed grain growth diameters for both the helium bonded Group F1 and F2 as well as the argon bonded E1B capsule pins. Because of the variety of variables in the input to the model, it is difficult, if not impossible, to determine any reason for the discrepancy between the models and experimental values.

Summarizing, the experimental data reported indicate the following:

- Gap conductance is sensitive to the gas atmosphere inside a fuel pin for diametral gaps greater than 0.002 inch.
- In general, the conductance values obtained for mixed oxide fuel, clad in stainless steel are slightly higher than for UO₂ clad in Zircaloy-2.

FIGURE 6-12. COMPARISON OF GAP CONDUCTANCE WITH ROSS AND STOUTE MODEL

SECTION VII

SUMMARY OF CONCLUSIONS

An experimental determination of the effect of diametral gap size on the gap conductance of stainless-steel clad mixed oxide in the power range of 18 to 20 kW/ft at a burnup of approximately 500 MWd/Te, indicates the following:

- Based on the diameter of equiaxed grain growth, the gap conductance for as fabricated diametral fuel-cladding gaps of 0.002, 0.004, and 0.010 inch were 1140, 750, and 520 Btu/h-ft²-°F.
- Based on the columnar grain growth diameters and the cold post-irradiation diametral gap sizes of 0.0012, 0.0018, and 0.0045 inch, gap conductances of 3000, 1910, and 740 Btu/h-ft²-°F were determined.

Examination of fuel rods irradiated in a fast flux to burnups of 1600 and 50,000 MWd/Te showed the following:

- Deposition of fission products and fuel at the fuel-cladding interface and the forma-

tion of oxide layers on the cladding surface affects the heat transfer across the fuel cladding interface.

- After 50,000 MWd/Te burnup, the fuel-cladding gap is obliterated and gap conductance values determined from the diameter of columnar grain growth indicate that the fuel-cladding gap conductance approaches a constant value in the range of 1100 to 1600 Btu/h-ft²-°F.

Comparison of the gap conductance values determined and results obtained from UO₂ experiments indicates the following:

- In general gap conductance values determined for mixed oxide fuel clad in stainless steel are slightly higher than reported for UO₂ clad in Zircaloy-2.
- Gap conductances were higher in helium bonded fuel pins than in argon for initial fuel cladding gaps greater than 0.002 inch.

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APPENDIX A

DESTRUCTIVE EXAMINATION -
CAPSULE E1B FUEL PINS

The E1B capsule and fuel pins were destructively examined at the General Electric - Radioactive Materials Laboratory (RML) located at Vallecitos, California.

The post-irradiation examination consisted of the following:

- Gamma scan of capsule and fuel pins.
- Diametral and length measurements on the fuel pins.
- Fission gas analysis.
- Radiochemical burnup analysis.
- Destructive examination of the fuel pins.
 - Alpha and beta-gamma autoradiography
 - Metallographic examination

The procedures, observations and results of each part of the examination are presented briefly below:

A.1 GAMMA SCAN

Prior to gamma scanning, the outer tube was removed from the capsule assembly. The remaining assembly was then scanned using settings which would detect gamma activity above an energy of approximately 0.65 MeV. A 10-mil slit was used to resolve details such as cracking or redistribution of the fuel more clearly. Gamma activity was nearly uniform, except for flux peaking effects, in all four fuel pins. The level of activity in Pin E1B-4 appeared to be slightly less than in the other three fuel pins (approximately 1/20 lower).

The gamma scans did not indicate any gross cladding failure nor was any evidence of fuel redistribution observed.

A.2 CAPSULE DISASSEMBLY

Before the inner and outer NaK containers were disassembled, a sample of the NaK was removed from the inner container and analyzed for fission products and alpha activity. Only Zr-95 and Zn-65, which are activation products, not fission products, were detected. It was concluded that none of the fuel pins had ruptured, and the outer containment can was removed.

A.3 EXAMINATION OF THE FUEL PINS

The fuel pins were removed from the inner container and were observed to be in excellent condition by visual inspection. Except for a slight discoloration of the ends of each fuel pin, there was no change in the appearance of the fuel pins. Fuel pin E1B-4 appeared somewhat darker than the other three.

A.4 DIMENSIONAL MEASUREMENTS

Pre- and post-irradiation length and diameter measurements were performed using a micrometer and dial gauge. The measurements are tabulated in Table A-1. In addition, continuous diameter and profile scans were made at 0° and 90° orientations. The results of the profile measurements of each fuel pin indicated no significant mechanical interaction between fuel and cladding. The profilometer traces are shown superimposed above the pins in Figures A-1 through A-4. Randomly occurring ridges 0.1 to 0.3 of a mil high were observed at pellet interfaces. The height of

TABLE A-1

PRE- AND POST-IRRADIATION DIAMETRAL AND LENGTH
MEASUREMENTS - CAPSULE E1B FUEL PINS*

<u>PIN 1</u>						
<u>Position**</u>	<u>DIAMETER 0° (inches)</u>			<u>DIAMETER 90° (inches)</u>		
	<u>Pre-Irradiation</u>	<u>Post-Irradiation</u>	<u>ΔD</u>	<u>Pre-Irradiation</u>	<u>Post-Irradiation</u>	<u>ΔD</u>
1	0.2510	0.251	0	0.2510	0.251	0
2	0.2510	0.251	0	0.2505	0.251	+0.0005
3	0.2510	0.251	0	0.2505	0.251	+0.0005
4	0.2510	0.251	0	0.2505	0.251	+0.0005
5	0.2510	0.251	0	0.2505	0.252	+0.0015
Length	***					
<u>PIN 2</u>						
1	0.2495	0.250	+0.0005	0.2495	0.250	0
2	0.2495	0.250	+0.0005	0.2495	0.250	0
3	0.2495	0.250	+0.0005	0.2495	0.250	0
4	0.2495	0.250	+0.0005	0.2495	0.250	0
5	0.2495	0.250	+0.0005	0.2495	0.250	0
Length	2.146	2.146	0			
<u>PIN 3</u>						
1	0.2565	0.257	+0.0005	0.2560	0.256	0
2	0.2565	0.257	+0.0005	0.2560	0.256	0
3	0.2565	0.257	+0.0005	0.2555	0.256	+0.0005
4	0.2570	0.257	0	0.2540	0.254	0
5	0.2575	0.258	+0.0005	0.2535	0.254	+0.0005
Length	2.131	2.131	0			
<u>PIN 4</u>						
1	0.2515	0.252	+0.0005	0.2505	0.252	+0.0015
2	0.2505	0.252	+0.0015	0.2505	0.251	+0.0005
3	0.2505	0.251	+0.0005	0.2505	0.251	+0.0005
4	0.2505	0.250	-0.0005	0.2505	0.251	+0.0005
5	0.2510	0.251	0	0.2510	0.251	0
Length	2.187	2.186	-0.001			

*Measurements were made with a micrometer.

**Measurements were taken at 1-inch intervals from top of fuel pin.

***No length measurement was taken because of thermocouple extending from this pin.

FIGURE A-1. CLADDING PROFILE OF PIN E1B-1

FIGURE A-2. CLADDING PROFILE OF PIN E1B-2

FIGURE A-3. CLADDING PROFILE OF PIN E1B-4

FIGURE A-4. CLADDING PROFILE OF PIN E1B-3

the ridges was independent of gap size. Ridges of the order of 1 mil and larger have been observed on water-cooled uranium dioxide fueled reactor rods, and ridges up to 1 mil were observed on rods fueled with mixed oxides of plutonium and uranium. (A-1, A-2, A-3) In the cases noted, cladding temperatures were in the 500°F to 600°F temperature range, which resulted in maximum fuel surface temperatures of 1400°F (750°C). Under the operating conditions for the present experiment, minimum cladding and fuel temperatures were approximately 1000°F and 1700°F respectively. Therefore, it is hypothesized that the higher fuel and cladding temperatures result in less fuel-cladding interaction because of an increase in plasticity of mixed oxide fuel.

Measurements of the fuel-cladding gap during post-irradiation examination indicated that the fuel-cladding gap had decreased to approximately one-half the pre-irradiation size in each fuel pin. The post-irradiation gap in pins E1B-2, -3, and -4 were respectively 0.0012, 0.0045, 0.0017 inch.* A gradual decrease in the fuel center temperature of pin E1B-1 was recorded by the in-fuel W-Re thermocouple for the first seven hours of the irradiation.** This would be expected if the fuel-cladding gap was decreasing during the irradiation. Because of the ingress of the NaK bond into the fuel pin, it cannot be concluded

that the decrease in the fuel center temperature was entirely due to a decrease in the fuel-cladding gap. Movement of fuel into the fuel-cladding gap has recently been reported for mixed oxide fuel irradiated to low burnup in a fast flux where gap closures of up to 0.003 inch were observed. (A-3)

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*Average values determined from 100× photomicrographs taken at 0°, 90°, 180° and 270° orientations around each fuel section.

**The thermocouple failed after 7 hours of operation because of NaK leakage through the braze between the thermocouple and fuel-pin end plug.

APPENDIX B

CALCULATION OF GAP COEFFICIENT,
FUEL PIN F1A

PARAMETERS:

Cladding o. d. (in.)	0.2488
Cladding i. d. (in.)	0.2203
Fuel o. d. (in.)	0.2162
Fuel Central Void i. d. (in.)	0.01694
Equiaxed Grain Growth	
Diameter (in.)	0.1840
Theoretical Density (%)	94.1
Linear Power (kW/ft)	16.1

The temperature at the cladding i. d. (T_c) is calculated to be 1092°F.

The gap coefficient can be calculated from consideration of fuel microstructure. For instance, the extent to which equiaxed grains grow can be used as a temperature marker from which fuel surface and, hence, gap conductance can be determined. In this instance, equiaxed grain growth is assumed to occur at temperatures above 1650°C.

Following the treatment of Robertson*, the expression of the parameter $\int k dT$ for a hollow cylinder of outer diameter, a , inner diameter, b , and linear power, W , to any diameter, d , is:

$$\int_{T_a}^{T_d} k(T) dT = \frac{W}{4\pi} \frac{1}{a^2 - b^2} \left[(a^2 - d^2) - 2b^2 \ln \left(\frac{a}{b} \right) \right] \quad (1)$$

Substituting the parameters for fuel pin F1A into Equation (1),

*Robertson, J. A. L., " $\int k d\theta$ in Fuel Irradiations," April 1959, [AECL-807 (CRFD-835)]

$$\int_{T_a}^{T_{egg}} k(T) dT = 11.6 \text{ w/cm}$$

Now, Baily, et al.,* has reported a density-thermal conductivity relationship for stiochiometric mixed oxide fuel:

$$k(T, D) = 0.011 + \frac{1}{T(0.4848 - 0.4465D)} \quad (\text{watts/cm-}^\circ\text{C}) \quad (2)$$

where,

T = temperature ($^\circ\text{C}$) and

D = fractional theoretical pellet density.

Integrating Equation (2) between temperatures T_1 and T_2 ,

$$\int_{T_2}^{T_1} k(T, D) dT = 0.011 (T_2 - T_1) + \frac{\ln}{(0.4848 - 0.4465D)} \quad (\text{w/cm}) \quad (3)$$

Equation (3) can now be solved implicitly for surface temperature T_a , assuming $T_{egg} = 1650^\circ\text{C}$:

*Baily, W. E., et al., "Thermal Conductivity of Uranium-Plutonium Oxide Fuels," Nuclear Metallurgy, Volume 13, International Symposium on Plutonium Fuels Technology, Proceedings of the 1967 Nuclear Metallurgy Symposium held in Scottsdale, Arizona on October 4-6, 1967, American Institute of Mining, Metallurgical and Petroleum Engineers, Inc. 1968.

$\int_{T_a}^T \text{egg } k(T) dT$ was previously determined to

equal 11.6 w/cm,

$$11.6 = 0.011 (1650 - T_a)$$

$$+ \frac{\ln (1650/T_a)}{[0.4848 - 0.4465 (0.941)]}$$

Therefore,

$$T_a = 1128^\circ\text{C}$$

$$= 2063^\circ\text{F}$$

Now,

$$\Delta T_{\text{gap}} = T_c - T_a = 971^\circ\text{F}$$

The surface heat flux for pin F1A operating at 16.1 kW/ft,

$$\text{SHF} = \frac{(16.1 \text{ kW/ft}) (3413 \text{ Btu/h-kW}) (12 \text{ in./ft})}{\pi (0.2203 \text{ in.})}$$

$$= 952,748 \text{ Btu/h-ft}^2$$

The gap conductance for fuel pin F1A is $h_{\text{gap}} =$

$$\frac{\text{SHF}}{\Delta T_{\text{gap}}} = 982 \text{ Btu/h-ft}^2 - ^\circ\text{F}$$

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