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EXPERIENCE IN EBR-II

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HEDL MIXED OXIDE FUEL PIN BREACH EXPERIENCE IN EBR-II

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INTRODUCTION

Mixed plutonium-uranium oxide fuel pins clad with stainless steel are being irradiated to cladding breach in the EBR-II for the Hanford Engineering Development Laboratory's (HEDL) Run-to-Cladding-Breach (RTCB) program.¹ The objectives of these tests are to: 1) establish the burnup capability of mixed oxide fuel pins, 2) identify and characterize cladding breach mechanisms, 3) determine the effects of cladding breach on neighboring fuel pins and 4) provide data for developing fuel pin failure criteria.

Breaches have occurred in 16 HEDL fuel pins in the RTCB program. Examination and characterization has been completed on 13, with the remaining three in some phase of detailed examination. Characterization, includes identification and classification of the probable cause of each breach. The breaches evaluated have been classified as being due to: local cladding overtemperature, subassembly coolant overtemperature, and cladding and wire wrap wear. The cause for one breach has not been conclusively established. This paper will review characteristics, and discuss the probable mechanisms and causes.

TEST DESCRIPTION

The breached fuel pins, containing 343 mm long columns of 25 w/o PuO_2 -75 w/o UO_2 sintered pellets, were clad with solution annealed type 304 or cold worked type 316 stainless steel wrapped with a spacer wire at a 12 inch pitch, Table I. Twelve pins were from subassemblies fabricated to the Fast Flux Test Facility (FFTF) reference fuel and cladding specifications current at the time except for P-12AA-63K. The cladding on this pin was 30% cold worked and the plenum to fuel volume ratio was 0.62:1. The remaining four were from three subassemblies which contained pins with advanced oxide fuel design parameters.² The plenum to fuel volume ratios in P-40-D91, P-41-A3R, P-41-B10 and P-42-B71 were 2.24:1, 0.2:1, 0.51:1 and 0.51:1 respectively. Also the P-40, P-41 and P-42 fuel pins had thinner cladding than the reference fuel pin design. Fuel smeared density of pins ranged from 85% Theoretical Density (TD) in the reference fuel tests

*Operated by the Westinghouse Hanford Company for the United States Department of Energy.

TABLE I

Cladding Variables and Irradiation Conditions of HEDL Breached Fuel Pins.

PIN	CLADDING		FUEL BURNUP	CLADDING FLUENCE	INTERNAL PRESSURE	CALCULATED CLADDING MIDWALL TEMPERATURE AT TOP OF FUEL	PEAK PIN POWER
	TYPE/CONDITION	O.D./WALL THICKNESS (mm)/(mm)	EOL ⁽¹⁾ a/o	EOL $n/cm^2 \times 10^{23}$ (E > 0.1 MeV)	EOL MPa	BOL ⁽²⁾ /EOL °C	BOL/EOL KW/m
PNL 5A-1	304/SA	6.35/0.41	13.1	0.85	8.27	475/457	45.8/32.8
PNL 5B-17	304/SA	6.35/0.41	13.7	0.96	9.82	480/471	44.3/33.8
PNL 10-14	316/20%	5.84/0.38	6.6	0.50	3.93	564/540	30.3/26.9
PNL 11-39	316/20%	5.84/0.38	10.7	1.03	7.34	536/534	41.3/31.5
N/E-N122	316/20%	5.84/0.38	4.4	0.31	2.62	598/585	41.0/37.1
E/F-N054	316/20% CW	5.84/0.38	3.9	0.29	2.48	602/578	41.2/36.1
P-12AA-63K	316/30% CW	5.84/0.38	3.4	0.23	2.96	590/642	38.3/33.1
P-12AB-11B	316/20% CW	5.84/0.38	7.1	0.52	6.00	622/628	40.8/31.8
P-23B-1A	316/20% CW	5.84/0.38	5.3	0.5	2.96	627/612	39.1/35.8
P-23A-37	316/20% CW	5.84/0.38	10.5	1.0	7.01	595/575	40.0/28.2
P-23B-73E	316/20% CW	5.84/0.38	9.6	0.95	5.87	706/660	37.5/29.2
P-14-29	*316/20% CW	5.84/0.38	10.4	0.78	7.20	602/578	43.7/34.1
P-41-A3R	*316/20% CW	6.86/0.28	1.7	0.19	2.77	568/565	39.5/39.3
P-41-B10	*316/20% CW	6.86/0.28	1.7	0.19	1.02	568/564	39.3/39.2
P-42-B71	*316/20% CW	6.86/0.28	1.5	0.12	0.88	541/525	44.7/41.8
P-40-D91	*316/20% CW	5.84/0.25	12.1	0.9	4.01	614/581	50.8/40.0

(1) EOL - END OF LIFE

(2) BOL - BEGINNING OF LIFE

*PINS ARE CLAD WITH FFTF CORE 1-2 CLADDING

HEDL 7811-084.1

to 91.9% in some of the advanced oxide fuel pins. The pins were irradiated as 61, 37 or 19 pin hexagonal subassemblies to peak burnups as high as 13.7 a/o at maximum linear powers from 26.0 to 50.8 kW/m and calculated maximum cladding temperatures of 475°C to 706°C. Cladding variables and dimensions and calculated irradiation conditions at the beginning and end of life are given in Table I. Five of the breached pins have cladding from the FFTF first core cladding lots. The claddings on the remaining breached pins are from earlier developmental heats of stainless steel.

BREACH DESCRIPTIONS

The breached pin identity was determined by measuring ^{133}Xe gamma activity in the plenum, pin weight changes, activity in the water after washing selected pins, visual examination, or a combination of these. Non-destructive examination including visual and photographic coverage, profilometry, gamma scanning and eddy current scanning preceded the deliberate puncturing of the plenum. Breach locations were confirmed by internally pressurizing the pins with helium until a bubble stream showed in a bath of alcohol or a mass spectrometer pin-pointed the leaking helium. The breach was then examined metallographically. Where possible each breach was characterized as to its axial and circumferential location, size, cladding total diametral strain, total inelastic strain, and cladding thickness

reduction, Figure 1. The gamma scan measurements were analyzed for abnormally low or high gamma activity at or near the breach site as possible indicators of fission product associations with breach causes. The metallographic examination revealed failure modes (intergranular or transgranular), associated cladding and fuel microstructure, and character and extent of any chemical attack on the cladding inner and outer surfaces.

In an ideal subassembly of wire wrapped fuel pins, the nominal design coolant flow produces a distribution of temperatures decreasing radially from the central pin to the outer rows of pins. Temperature increases axially along the fuel columns because the EBR-II coolant flows upward. Circumferentially, the temperatures of the cladding are a maximum on the side toward the center of the subassembly and decrease to the minimum on the opposite side. Local hot spots on the cladding are not expected to occur unless serious perturbations occur in the coolant flow. Results from the examination several breached pins, however, showed evidence that cladding temperature distributions around these pins were not as the nominal analysis predicted. Evidence will be shown of breaches resulting from 1) a local cladding overtemperature or a local cladding overtemperature superimposed on a general overtemperature of several pins in a group, and 2) a global overtemperature of the subassembly coolant.

Local Cladding Overtemperature

The breach in the 37 pin subassembly P-23A occurred in a corner position pin, P-23A-37, at a peak burnup of 10.5 a/o. The breach, though not detectable during visual examination was located in a direction facing the adjacent duct wall and at an axial location, 250 mm above the bottom of the fuel (ABOF), where the wire wrap would not have supported the pin away from the duct wall. Diameter measurements at the breach location showed essentially the same total cladding strain, 1.2%, as in the cladding adjacent to the breach. Gamma activity measurements indicated no associated concentration of fission products. Metallographic examination showed the breach to be an intergranular failure with some short branched and parallel cracks, Figure 2. The deformation of the cladding, shown on the outer surface in Figure 2, and found at the same angular orientation 38 mm lower on the pin must have occurred by mechanical interaction between the cladding and duct, possibly during the assembly reconstitution. In a narrow angular sector on both sides of the breach, the carbides were enlarged and fewer in number than elsewhere on the cladding. Second phase particles, believed to be sigma and chi, were found at numerous grain boundary intersections. A 10 μm thick layer of ferrite was present on the cladding outer surface near the breach and not elsewhere. There was no evidence of creep voids or grain boundary precipitates associated with the breach. The ferrite layer implies a temperature at the breached cladding outer surface of at least 600°C. The evidence of sigma and chi phase indicates temperatures between 675°C and 700°C. These are 135 to 165°C higher than the calculated

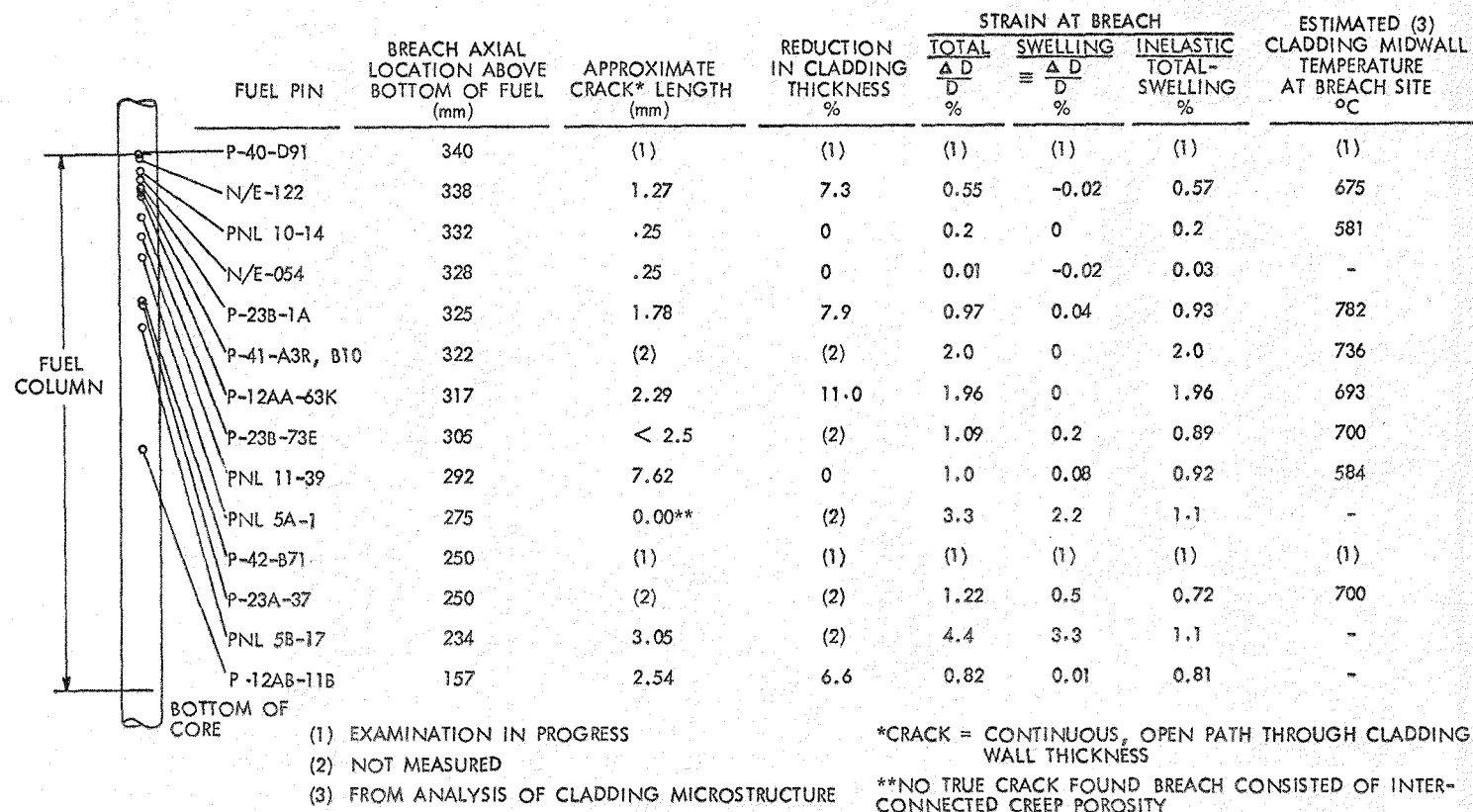


Figure 1. Breach Location, Cladding Strain Measurements and Estimated Cladding Temperature at the Breach.

temperatures for the cladding facing the duct wall. Overheating is postulated to have occurred in a localized volume of cladding by contact or near contact between the cladding and the duct wall. The duct for this particular test assembly was thermally insulated, such that heat dissipation from a contacting pin would be inhibited. A crack, initiated either at the cladding inner surface or on the outer surface in the deformed region, propagated along grain boundaries weakened by the coalesced carbides and second phase precipitates until it penetrated the cladding, releasing fission gases.

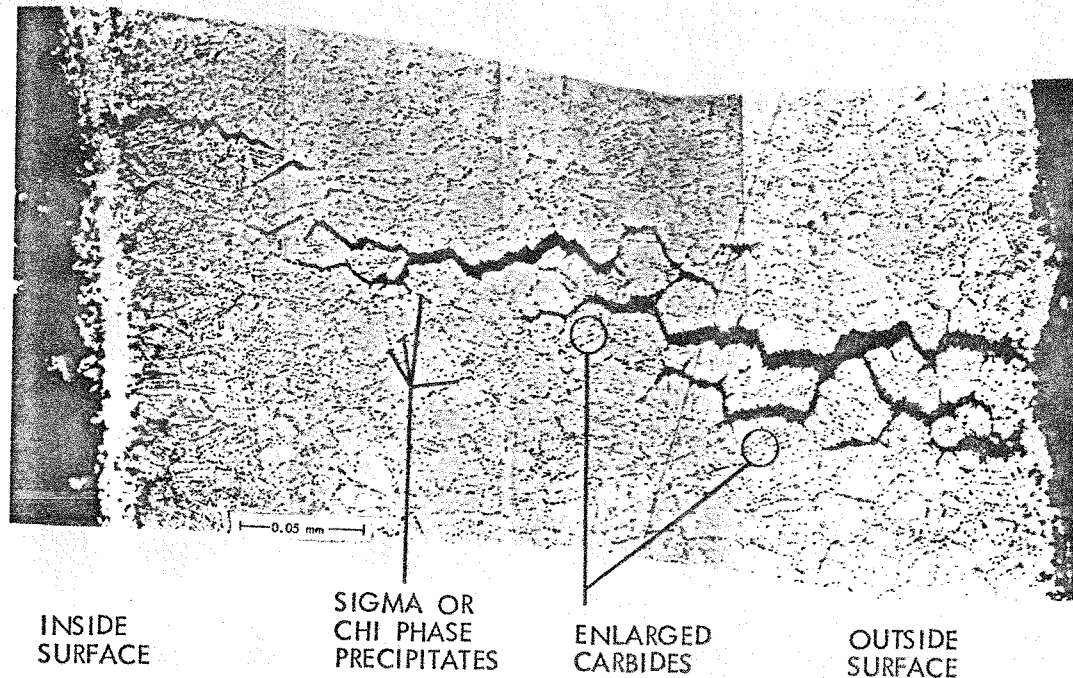


Figure 2. Breach and Cladding Microstructure on Fuel Pin P-23A-37.

The breach in corner pin P-23B-1A at a peak burnup of 5.3 a/o is postulated to have also resulted from contact or near contact with an insulated duct.³

A second breach in the P-23B subassembly occurred at a higher peak burnup, 9.6 a/o, in an interior pin P-23B-73E. In this case the breach was located in the center of a circular white spot on the cladding outer surface, Figure 3, at an angular position facing an adjacent interior pin. The adjacent pin showed a similar white spot facing P-23B-73E. Metallographic examination revealed a breach and cladding microstructure very similar to that of P-23A-37. The breach in this pin is believed to have occurred from local overheating of the cladding caused by pin-to-pin contact.

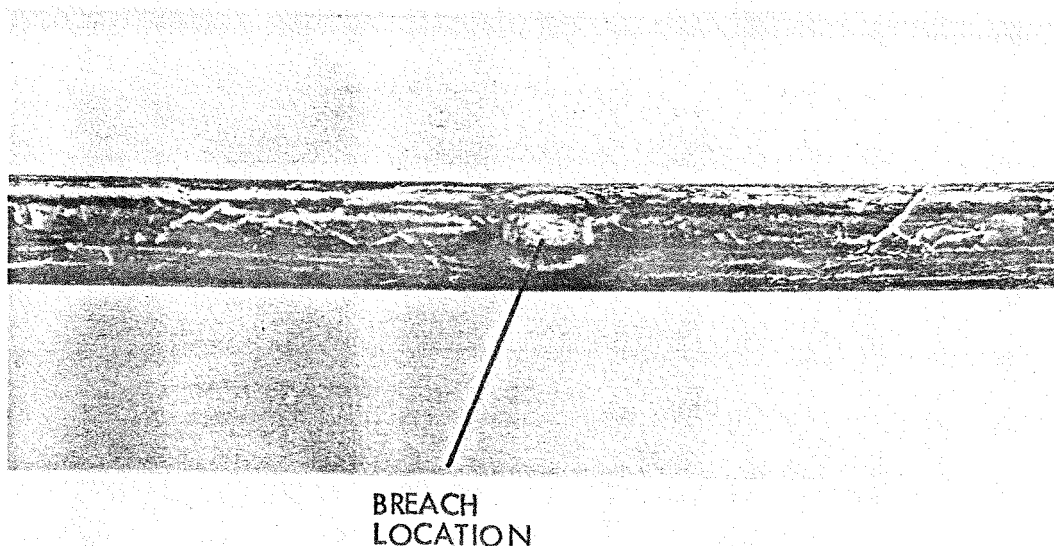


Figure 3. "White Spot" at Breach on Fuel Pin P-23B-73E.

The accumulated evidence from the breach and sibling pin examinations showed that similar pin-to-pin or near pin-to-pin contact occurred for P-12AB-11B, PNL-5A-1 and PNL-5B-17 breaches.^{4,5}

The pin N/E-122, which breached at a burnup of 4.4 a/o, was the central pin in a 37 pin subassembly. Diameter measurements of the cladding showed an abrupt increase of 0.55% in the total strain at the breach, 338 mm ABOF. Metallographic examination revealed the cladding breach as a connection of large creep voids, Figure 4. The grain boundaries were denuded of the normal carbides over a 50° arc centered on the breach and instead contained large precipitates. The creep voids were frequently formed at the interfaces between the precipitates and the denuded zone leading to a failure similar to that described in Reference 3. Energy dispersive X-ray analysis with a scanning electron microscope identified the large grain boundary precipitates as sigma and chi phases. [Similarities in the morphology and etching behavior led to the conclusion that the larger precipitates observed in the claddings of other breached and sibling pins were also sigma and chi phase.] Measurements of cladding thickness on N/E-122 indicated a localized reduction of approximately 7.3%. There was a 6 μm thick continuous layer of ferrite around the outer surface. A zone, 0.05 μm thick, on the cladding inner surface was void of intergranular and grain boundary carbides. Examination of a neighboring pin revealed a ferrite layer adjacent to N/E-122 that was significantly thicker. Eddy current scans of the central breached pin and the surrounding six neighbors showed similarities in their signals that indicated all the central grouping of seven pins had an increase in the thickness of ferrite. These results indicate the cladding temperatures in the breached N/E-122 pin and its neighbors

were higher than designed. The reduction in cladding thickness, the abrupt diameter increase, and the concentration of creep voids plus higher temperature phases as grain boundary precipitates at the breach site of N/E-122 indicate a localized temperature increase was superimposed on a general overtemperature of the pins in the central grouping. We postulate that pin-to-pin or near pin-to-pin contact occurred between N/E-122 and one of its neighbors to cause the local overtemperature.

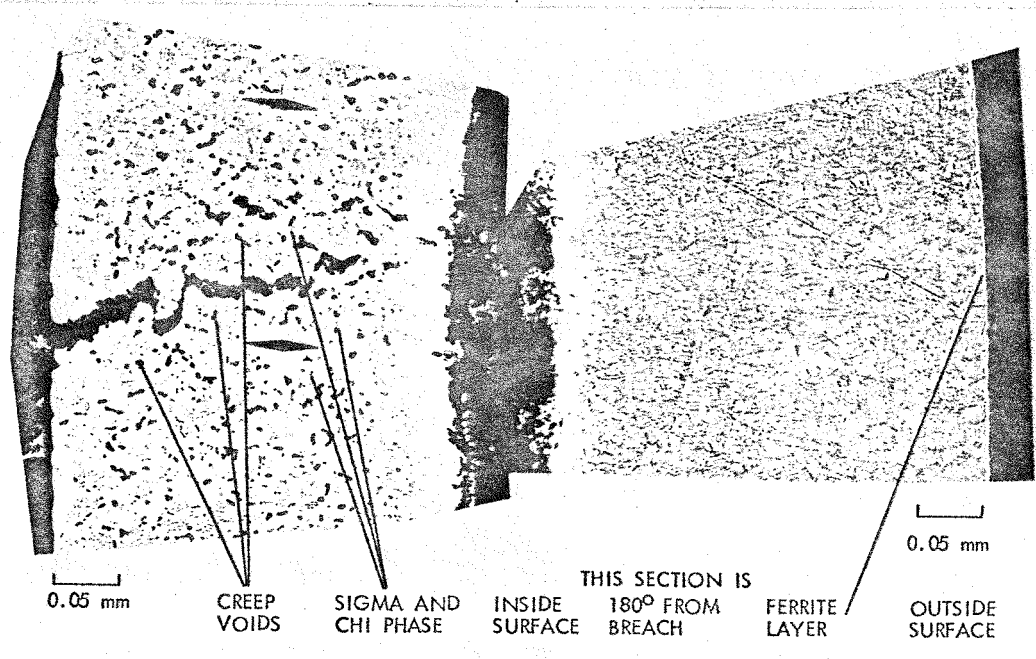


Figure 4. Breach and Cladding Microstructure on Fuel Pin N/E-122.

The breached pin P-12AA-63K and its siblings in the P-12AA subassembly displayed a similar behavior indicating a localized cladding overtemperature on top of a general overtemperature of the central group of pins.

Subassembly Coolant Overtemperature

The 19 pin P-41 and P-42 subassemblies sustained breaches very early in life, at 1.5 a/o burnup.² During the examination of the P-41 subassembly, breaches were visible on two pins, P-41-A3R and P-41-B10, although it is believed only one breach occurred in reactor. The breaches were small open holes located where the wire wraps from adjacent pins had contacted the cladding. Destructive examinations of P-41-A3R showed that the cladding had been pulled away from the fuel and that if the edges of the breach were pieced together a wedge of cladding material was missing. This wedge of material had adhered to the wire wrap of a neighboring pin where it contacted the breach area. The breaches are believed to have been pulled

open as a result of forces, applied during dismantling of the subassembly, acting on solid state welds formed between the cladding and the wire of neighboring pins during the irradiation. Metallographic examination confirmed that wires had welded to the cladding in some location and showed that large second phase precipitates of sigma and chi were present in the wire and cladding. Metallography of the wire wraps from several unbreached pins showed that formation of the higher temperature phases extended throughout much of the P-41 subassembly. A comparison with microstructure from isothermally annealed cladding indicated that sodium temperatures in P-41 were in excess of 760°C at the top of the fuel column in the subassembly interior. The cladding total diametral strain increased rapidly from zero at 228 mm above the bottom of the fuel to over 2% at the breach at 322 mm. We postulate that the original in-reactor breach occurred in P-41-A3R as a creep type failure due to unexpectedly high sodium temperatures. Subsequently, during dismantling, the forces on wire-to-cladding welds caused the open breaches on both pins.

The single breached pin found in the P-42 subassembly, P-42-B71, was located in a corner position in the subassembly. The breach, not detectable during visual examinations, was located at 250 mm ABOF orientated toward the subassembly duct wall. There were no cladding-to-wire wrap welds found in this subassembly. Metallographic examination of wire wrap from all P-42 pins showed large amounts of second phase material in wires from the interior of the subassembly. These results indicate that the P-42 subassembly had also operated with unusually high sodium temperatures. Examination of the breach pin is underway.

Cladding Wire Wrap Wear

Flow induced vibrations caused moderate to severe wear and thinning of the cladding by the wire wrap of adjacent pins in the 61 pin subassemblies PNL-10, PNL-11 and N/E.¹ Each subassembly was subsequently reconstituted as a 37 pin subassembly and irradiation continued until a fuel pin breached.

PNL 10-14 breached in a cladding wear mark, 332 mm ABOF, at a peak fuel burnup of 6.6 a/o. Total inelastic strain at the failure, calculated from diameter and cladding swelling measurements, was approximately 0.2%. Metallographic examination revealed a very short axial breach in the wear mark at a minimum cladding thickness of 0.15 mm. Severe cold working on the surface of the cladding in the wear mark by the wire of an adjacent pin, and intergranular attack on the cladding inner surface further reduced the unaffected cladding to 0.13 mm, 1/3 the original 0.381 mm thickness.

The breach in PNL 11-39, which was visually located during postirradiation examination, also occurred in a wear mark at 292 mm ABOF. Total inelastic strain at the failure was approximately 1%. Metallographic examination showed that the breach, approximately 7.6 mm long, was a mixed intergranular and transgranular failure with numerous transgranular microcracks. The minimum cladding thickness was 0.178 mm (Figure 5).

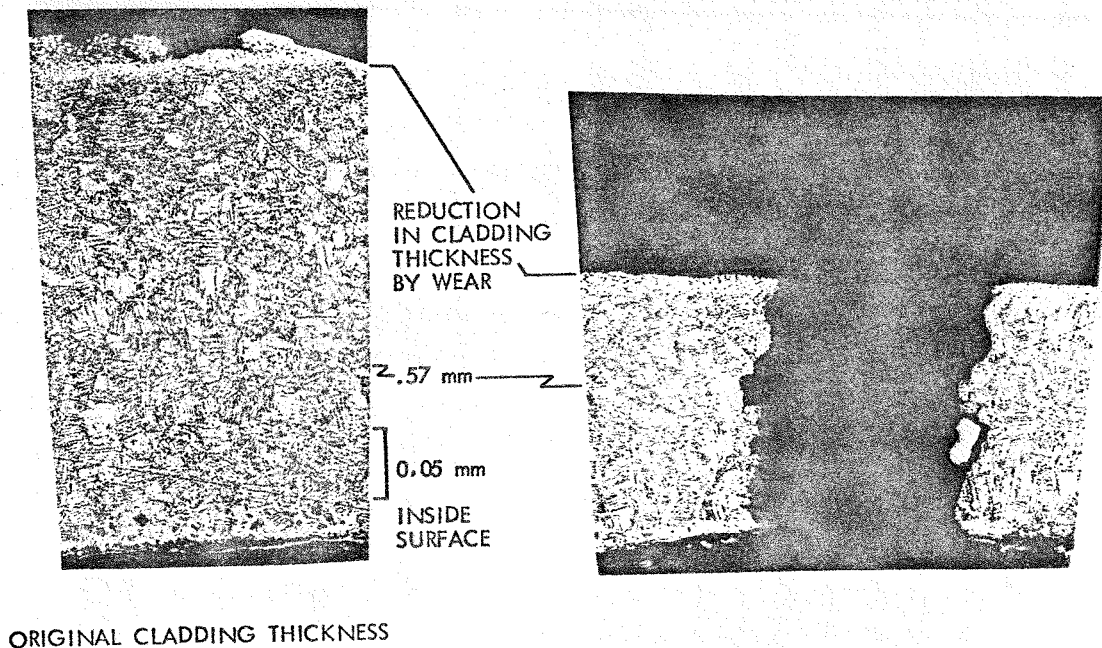


Figure 5. Breach in Cladding on PNL 11-39 in Wear Mark.

Unknown Cause

Pin N/E-054 breached at 328 mm ABOF in a shallow wear mark after irradiation to a burnup of 3.9 a/o. The diameter increase was essentially zero at the breach. Metallographic examination showed only one indication of the cladding breach and that, only on a roughly polished surface. The crack, which did not completely penetrate the cladding, intersected the outer surface of the cladding near the edge of the wear mark. After final polishing to remove less than 0.03 mm material, the crack was no longer visible. Chemical etching revealed a series of small intergranular cracks near the cladding inner surface that could have been the extension of the crack seen in the rough polished condition. Unusual etching effects on the cladding inner surface suggest abnormal phase or compositional variations. Analysis for nitrogen in the irradiated cladding of these N-series pins showed nitrogen higher by a factor of 3 in the region of the fuel column than in the unirradiated cladding. The source of the nitrogen was believed to be the mixed oxide fuel in these pins which contained an average of 170 ± 95 ppm N_2 versus less than 25 ppm N_2 in the fuel of the other reference pins.

The mechanism of cladding breach is unknown for this pin because of the very limited opportunity to examine the character of the crack. The unusual etching behavior and the excessive increase in nitrogen content suggests that the microstructure and chemistry in the cladding was different than normally observed. This may have adversely influenced strength and ductility.

Examinations in Progress

Examinations of three breached pins are still in progress. The P-14-29 pin breached while being irradiated in an interior position in a 37 pin subassembly at a peak burnup of 10.4 a/o. The breach was not visible, and diameter measurements showed no localized cladding strain. Examination to locate and characterize the breach is continuing.

Characterization of the P-42-B71 breach is continuing also.

A breach occurred at the top of the fuel column in pin P-40-D91, in a 61 pin subassembly, after irradiation to a peak burnup of 12.1 a/o. Although the breach on P-40-D91 is visible as a large open crack, it is believed that the original breach was a small hairline crack or pinhole. We postulate that water entered the pin during subassembly cleaning and reacted with fission products to form a larger volume reaction product which extended the original crack. Characterization is continuing.

DISCUSSION

After the accumulated evidence of local and general overtemperatures in the cladding was reviewed, a classification of probable causes of the breaches into three or more groups became evident, Figure 6. The similarity of breaches in the category of local cladding overtemperature is that they occurred where the wire wrap did not prevent pin-to-pin or near pin-to-pin contact. However, as this condition exists at many points along the fuel pin, a driving force must have been present to cause the contact. Radial temperature gradients, as much as 1000°C on corner pins, result in post-irradiation pin bowing of several centimeters in exterior and to a lesser extent in interior pins when the restraint of the duct is removed. During reconstitution and the subsequent irradiation, pin-to-pin forces resulting from the bow can cause pins to be displaced from their normal positions while still retaining the basic hexagonal arrangement. It is postulated that the pins which breached, by local cladding overtemperature were forced into contact or near contact with the insulated duct (P-23A-37 and P-23B-1A) or their neighbors in such a manner that coolant flow was severely restricted locally both circumferentially and axially. In the cases of P-12AA-63K and N/E-122, restrictions of flow in the interior of the subassembly are postulated to account for the general overtemperature as shown by high temperature microstructure around the breached pins and their neighbors. The condition of local cladding overtemperature is not, however, prototypic of that found in most of the other pins examined in these subassemblies. The severe pin displacements resulting in pin-to-pin and pin-to-duct contact appear to be related to the reconstitution of these subassemblies.

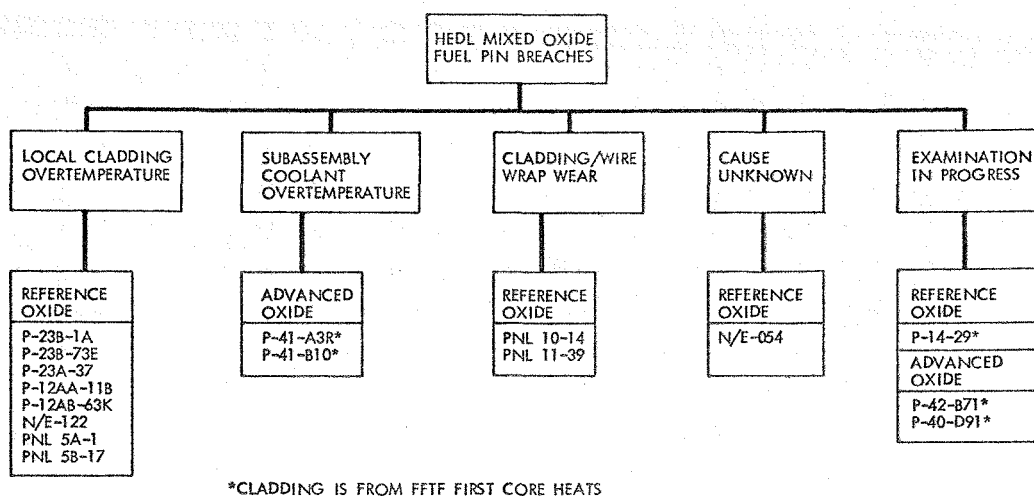


Figure 6. Classification of HEDL Mixed Oxide Fuel Pin Breaches by Probable Cause.

Estimates of the cladding midwall temperatures at the breach site have been made for several of the pins, Figure 1, by comparing the cladding microstructure with the isothermal annealing microstructure of unirradiated cladding. The estimate for the time at temperature assumed the coolant perturbations started as a result of the most recent subassembly reconstitution prior to breach. The temperature estimates for PNL 10-14 and 11-39 were based on observations of ferrite on the cladding outside surface. It appears from examinations of pins in the PNL-10 and PNL-11 subassemblies that, in general, they operated at higher than design temperatures. However, no evidence of localized cladding overtemperature was found.

The condition of overtemperature in the cladding is more widespread and more severe in the P-41 and P-42 subassemblies than in any others. The conditions in the subassembly that led to this general coolant overtemperature are not yet clear, but it is suggested that they are related to the subassembly hardware design. It is possible that the overtemperature of these two subassemblies and the P-12AA, N/E, PNL-10 and PNL-11 subassemblies are related in that all were irradiated in similar EBR-II hardware with segmented liners.

Although nine of the sixteen pins breached at a burnup less than the 8.5 a/o peak goal for FFTF, only one, N/E-054, breached for causes that could not definitely be characterized as prototypic. The burnup capability of the reference FFTF fuel pins has been demonstrated in the achievement of almost or greater than 10 a/o burnup by PNL-11, P-23A and P-14. Considering the loss of more than 50% of the cladding thickness at the wear mark before the breach occurred, PNL 11-39 demonstrated the potential for much

higher burnup capability. Irradiation of another HEDL subassembly of reference fuel pins, P-14A, was terminated after it reached 8.7 a/o burnup without breaching. The HEDL P-14 and P-14A EBR-II tests with tighter bundles did not show a vibration and wear problem.⁶

The inelastic diametral strain at the breaches was greater than 0.5%, Figure 1, except for PNL 10-14 and N/E-054. The low strain to failure on PNL 10-14 is believed related to the presence of the cold work layer and perhaps the intergranular attack at the deep wear mark. The reason for the low strain at failure in N/E-054 is not obvious from the examination. However, the excessively high bulk nitrogen in the cladding and the unusual microstructure on the cladding inner surface suggest that the cladding was not prototypic under these conditions and that its mechanical behavior should not be expected to be prototypic.

All of the breaches have demonstrated a benign nature, even those which occurred under relatively severe irradiation conditions of high temperatures, such as the P-41-A3R pin, and high internal pressure, as in the PNL-11, P-14 P-23A and PNL-5 pins. The large volumes of fission gas released from the high burnup pins, over 200 cm³ @ STP in PNL-11 and PNL-5 pins, had no apparent effect on the breached or neighboring pins nor on the subassembly in general. Although the evidence strongly points to pin-to-pin contact in several of the breaches there was no failure propagation to other pins.

Breaches in four pins were detected during the visual examination phase, P-41 (2 pins), P-40, and PNL-11. Breaches in the remaining pins were microscopic and in most cases invisible even after they were located by the internal pressure and bubble test. Of the four visible breaches only that in PNL 11-39 appeared to be in approximately the same condition as it was in-reactor. The others had been altered from their original in-reactor state by the conditions imposed during washing and disassembly. However, there was some sodium ingress in the PNL 11-39 pin that could have expanded the breach open enough to make it more visible. For this pin, sodium was present along the entire length of the fuel column, in the central void and in a rim of fuel around the circumference. There was no indication of fuel loss although there was a delayed neutron signal from the reactor coolant following the detection of fission gas activity in the reactor cover gas. Sodium was also detected in the fuel of P-12AB-11B.

One objective of the metallographic examinations of the breaches was to find, if possible, the site of crack initiation and to compare the condition of the cladding at the breach with that elsewhere on the fuel pin. The results indicated that breaches did not necessarily initiate at the point on the fuel pin cladding where grain boundary attack or weakening of the cladding from FCCI or sodium cladding chemical attack (SCCI) was most severe. Another important observation made on each breached fuel pin relating to fission products was that no anomalous concentrations of gamma active fission products were detected at the breach sites.

CONCLUSIONS

From the results of the irradiation and examinations of the 16 breached pins and their siblings the following conclusions can be made:

- 1) All of the examinations suggest that the fuel pin breaches occurred as the result of nonprototypic conditions of irradiation.
- 2) All breaches were benign, nonpropagating, and, except for PNL 11-39, microscopic.
- 3) High burnups, >10 a/o, and high fluences, up to 0.8×10^{23} n/cm² ($E > 0.1$ MeV), are achievable in FFTF fuel pins clad with the reference 20% cold worked type 316 stainless steel from both the earlier developmental steel heats and the FFTF first core steel heats.
- 4) The fuel pin cladding exhibited good ductility in that the inelastic diametral strain at the breach was generally greater than 0.5%.

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