POST-IRRADIATION EXAMINATION OF OVERHEATED FUEL BUNDLES

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ABSTRACT

Post-irradiation examinations (PIE) were conducted on prototype 43-element CANDU¹ fuel bundles that overheated during test irradiations in the NRU reactor. PIE revealed that the bundles remained physically intact, but on several elements the Zr-4 sheath collapsed into axial gaps between the pellet stack and end caps, between adjacent pellets within the stacks, and into missing pellet chips and cracks. Helium pressurization tests showed that none of the collapsed elements leaked. Hydride blisters were discovered on a few elements, but the source of the hydrogen was not linked to a breach of the cladding or end caps. These defects were attributed to primary hydriding. Microstructural changes in the fuel and cladding indicate that the cladding was briefly exposed to temperatures in the range 600-800°C and pressures above 11.2 MPa. The results show that Zr-4 cladding behaves in a highly ductile manner during such transient, high-temperature and high-pressure excursions.

INTRODUCTION

Five prototype 43-element CANDU fuel bundles (1,2) were irradiated in the U2 loop of the NRU reactor from 1990 November 5 to 19, at linear power ratings up to 65 kW/m (equivalent to 1250 kW bundle power). The bundles overheated following a reactor trip due to the loss of class IV electrical power to the site. The main pumps of the irradiation test loop were not equipped with flywheels or backup power, and they stopped almost immediately after the power outage. When the pumps stopped, the coolant flow stagnated briefly in the down-flow leg of the U2 loop, and then reversed direction as convective cooling was established. The bundles overheated during this brief stagnation period. On the subsequent startup of NRU, the loop-coolant gamma-activity monitors indicated that there was a defect in the string, and the fuel assembly was removed from the reactor. Post-irradiation examinations in the NRU bays and the hot cells revealed that the bundles were physically intact, but the Zr-4 sheath had collapsed into axial gaps in several elements. Hydride blisters with fission-product deposits were also observed.

This paper describes the prototype bundle fabrication and the irradiation conditions, and presents the main results from the post-irradiation examinations.

FUEL MATERIALS

The prototype 43-element bundle contains two element sizes, 11.5 mm diameter elements with 0.33 mm thick (nominal) Zr-4 cladding in the outer and intermediate rings, and 13.5 mm diameter elements with 0.35 mm thick (nominal) cladding in the inner ring and centre. The Zr-4 tubing was supplied in the annealed condition, according to the ASTM standard, B-353-83. The yield strength was 362-400 MPa, UTS 538-578 MPa, elongation 28%, and grain size 9 μ m. Bearing pads and spacer pads were brazed onto the sheaths by Zircatec Precision Industries (ZPI). All sheaths were CANLUB coated.

¹ CANDU: CANada Deuterium Uranium; registered trademark.

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The fuel pellets were also made by ZPI, using both natural UO₂ and 2.59% enriched UO₂. The double-dished pellets were produced with a 12.5° chamfer, but approximately four stack lengths, two of each diameter, were made with a 45° chamfer. This feature was expected to reduce pellet ridging at high burnup, and these pellets were included in selected elements in three bundles.

The sintered density of the pellets was depressed by using controlled additions of a carbohydrate pore former to produce residual porosity of a suitable size (<10 μ m) and distribution. The final sintered density of the enriched pellets was 10.51-10.56 g/cm³ and that of the natural UO₂ pellets was 10.64-10.71 g/cm³. The hydrogen content of the pellets was typically 150-250 μ g per stack.

BUNDLE FABRICATION

The five bundles were assembled and welded at CRL, using sub-assemblies (Zr-4 endcaps, endplates, sheaths and UO₂ pellets) provided by various suppliers. Two of the bundles, AHN and AHP, contained enriched UO₂ only in the inner elements, with natural UO₂ in the intermediate and outer elements. This allowed the inner elements to operate at higher power than achievable in the three all-enriched bundles. Selected elements in bundles AHK (22 and 36), AHL (22) and AHP (36) were also loaded with pellets containing a 45° chamfer.

IRRADIATION CONDITIONS

The five bundles, identified as AHM, AHL, AHK, AHP and AHN, were loaded in the vertical test section, in axial positions 1 (top) to 5 (near bottom), respectively. A Bruce-type fuel bundle, identified as ZK, was loaded in position 6 (bottom). The bundles were irradiated in the inlet (downflow) leg of the U2 loop from 1990 November 5 to 19. Table 1 lists the typical operating conditions during the irradiation and immediately prior to the trip. The light-water coolant was typically maintained at pH 10 with the addition of LiOH. Table 2 lists the element linear power ratings and surface heat flux during the irradiation. There was a flux tilt (~ 4%) across the loop site due to an adjacent, empty (withdrawn) control-rod site at NRU lattice position F21. Elements facing this D₂O-filled site were exposed to a higher flux and thus ran at higher powers than elements on the opposite side of the bundle. A total of approximately six full-power-days had accumulated during the short irradiation, resulting in a maximum element (mid-plane) burnup of 10.8 MWh/kgU.

Table 3 summarizes the pertinent events, as recorded on the NRU sequential logger, following the loss of pump power. The records show that the power outage in NRU lasted for approximately 4 min; however, the main U2 loop pumps were not switched back on for another 6 min. When the pumps are off, decay heat and stored energy of the loop fuel is removed by thermal syphoning; i.e., natural convective cooling. However, the normal flow direction in the inlet leg of the U2 loop is downward, so in order for thermal syphoning to begin, the coolant flow must first stop, and then reverse itself. Cooling of the fuel was severely impaired during this transition from pumped flow to natural circulation, and the fuel bundles, which were running at high power immediately before the trip, overheated during the brief stagnation period that preceded full thermal syphoning. Subsequent analysis indicates that the fuel was exposed to a more-or-less stagnant steam atmosphere above 11.2 MPa for at least 8-10 s.

POST-IRRADIATION EXAMINATIONS

The post-irradiation examinations consisted of: visual inspections and sipping tests in the NRU bays; disassembly and visual examinations in the universal cells; neutron radiographic examinations; leak testing via pressurization with helium; metallographic and ceramographic examinations in the hot cells; and, hydrogen analyses of the fuel samples.

RESULTS

NRU Bay Examinations and Sipping Tests

Underwater inspection revealed extensive sheath collapse on the outer elements of bundles AHK and AHL. The Zr-4 cladding had collapsed into gaps between the end cap and pellet stack, and between adjacent pellets within the stack. All other bundles appeared to be in good condition (subsequent examinations showed that the inner elements in bundle AHP had collapsed sheaths, but these were not visible underwater). Gamma activity in the water samples from underwater sipping tests indicated that bundles AHK and AHL had possibly defected.

Visual Examinations

Bundle AHM from position 1 (top), and AHN from position 5 (near bottom) were essentially undamaged, and hence were not destructively examined. However, bundles AHL (#2), AHK (#3) and AHP (#4), which operated in the higher flux sites in the loop, had extensive sheath collapse and evidence of hydride blisters. Figure 1 shows the location of the damaged elements in bundles AHL and AHK. Figure 2 shows the typical collapse features observed on several elements.

The damage in bundle AHL was mainly located in the segment of the bundle circumscribed by elements 8 through 16. Within individual elements most of the damage occurred near the reference end (RE) which was at the top during irradiation; however, there were also several instances of collapse near the bottom, or non-reference ends (NRE). In the outer ring (maximum element linear power 38-48 kW/m), 8 of the 21 elements contained areas with varying degrees of deformation, ranging from slight localized indentations to complete circumferential grooves. In the intermediate ring (maximum element linear power 24-31 kW/m), elements 27, 28 and 31 had crevices where the sheath collapsed into the gap between the pellet stack and the end caps at the reference end. The inner ring of 13.5 mm diameter elements (maximum element linear power 25-31 kW/m) contained two elements that had through-wall defects with fission-product stains flowing from the blisters.

The damage in bundle AHK extended to a larger segment of the bundle, compared to bundle AHL. In the outer ring (maximum element linear power 53-61 kW/m), 15 of the 21 elements were visibly damaged, some with severe collapse zones; i.e., deep circumferential grooves that were clearly visible through the cell window. In the intermediate ring (maximum element linear power 34-39 kW/m), six elements showed evidence of sheath collapse, and three had artifacts that may have been possible hydride blisters. In the inner ring of elements (maximum element linear power 35-40 kW/m), whitish oxide bands were observed at the edges of the braze heat-affected zones, and a possible hydride blister defect was observed.

Bundle AHP contained natural UO₂ in the outer and intermediate ring of elements, and 2.59% enriched UO₂ in the inner ring of seven elements. The outer and intermediate rings operated at maximum element linear powers of 28 and 21 kW/m, respectively, while the innermost ring operated at 65 kW/m. The outer and intermediate ring had no damaged elements, but five of the seven inner elements had collapsed zones.

Neutron Radiography

Fourteen elements from bundles AHK and AHL were radiographed to determine whether hydrogenous material could be detected in elements that were severely damaged or suspected of having through-wall defects. Hydrogenous material was found in elements AHL 36 and 39, but conclusive evidence was not found in either elements AHK 32 or 36, both of which were suspected of being defective.

Helium Leak Testing

Ten elements from bundles AHK, AHL and AHP were leak tested using high-pressure (13.8 MPa) helium. Two leaking elements were found in bundle AHL. Element AHL 36 contained two defects and element AHL 39 contained one defect, at the locations indicated on the radiographs. The suspect elements from bundle AHK (32

and 36) were retested, but no leaks were discovered. None of the elements with severe deformation from sheath collapse showed any evidence of a leak.

Metallographic Examinations

Elements from bundles AHL, AHK and AHP were selected for metallographic examination.

<u>Sheath Collapse.</u> Typical micrographs of the collapsed regions on representative high-power elements are shown in Figure 3.

The outer elements of bundle AHL operated at a maximum linear power of 48 kW/m before the incident. The sheath collapsed into the gap between the first and second pellets under the RE bearing pad of AHL 12, as shown in Figure 3. The measured gap between the pellets at the collapsed zone was approximately 0.84 mm, but the depth to which the sheath inner diameter intruded was approximately 1.3 mm from the outer surface. The equivalent diametral strain would be -16.5% if the depth of the crevice was uniform around the circumference; however, the support provided by the bearing pad prevented complete collapse. There was minimal sheath wall thinning at the crevice. The polarized light image shows that the deformed sheath contained the large grains expected in the braze heat-affected zone, but there was a thin layer of smaller recrystallized grains on the inside surface of the sheath. The microstructure of the pellets at the collapse site revealed equiaxed grain growth at the centre of the fuel. Other than typical cracks and a missing pellet chip that may have resulted from sample preparation, there were no anomalous features.

Similar but more extensive sheath collapse was observed in bundle AHK, which operated at a maximum outer element linear power of 65 kW/m.

The inner element in bundle AHP operated at a maximum linear power of 65 kW/m. Element AHP 36 contained 45° chamfered pellets, and as shown in Figure 3, the sheath collapsed into the space formed by the chamfers, which was approximately 1.0-1.4 mm wide at the pellet surface. The calculated diametral strain was approximately -3%. The micrographs show the expected equiaxed grain growth at the centre of the pellet, but there was also evidence of the onset of fuel coalescence and infilling of the dish at the hot centre of adjacent pellets.

<u>Oxide Thickness Measurements.</u> The inside surface of the intact elements was generally free of oxides, but oxygen-enriched layers up to 1 μ m thick were observed on the sheath I.D. Typically, the O.D. was covered with an oxide layer 1-2 μ m thick remote from the collapse zones, but within the crevice up to 10 μ m patches were seen.

<u>Hydride Blisters.</u> Hydride blisters were observed in elements AHL 36 and 39. On AHL 39, the hydride concentration extended over approximately 1.3 mm (3%) of the inside circumference of the sheath. The periphery of the fuel in the region of the defects contained a thin, fragmented, oxidized layer. In element AHL 39, this outer layer appeared to be adhering to the sheath. There was also evidence of oxidation along the radial cracks in the pellets. This suggests that the pellets were exposed to an oxidizing atmosphere while at elevated temperatures. It is not clear whether this occurred during the overheating transient, or as a result of the fuel being returned to power during the subsequent startup.

<u>Grain Growth.</u> Fuel samples from AHK (8, 27 and 36), AHL (12, 28 and 36) and AHP (12, 36 and 40) were sectioned, mounted transversely and examined to determine the degree of grain growth within the pellets. Most of the samples exhibited no measurable grain growth, or equiaxed growth at the centre. However, one of the three samples from element AHK 8 showed evidence of columnar grain growth extending to approximately mid-radius of the pellet (r/a = 0.48 - 0.52).

<u>End-Cap Examination</u>. The end caps from element AHL 39 were sectioned and examined to determine whether the material and welds were sound. No signs of piping or connected porosity were detected. Concentrations of randomly oriented hydride flakes were observed in the spigot of the RE end cap, but otherwise the end cap was free of anomalies, and the end closure welds were intact.

Hydrogen Analysis

The hydrogen-content of the irradiated Zircaloy sheath samples ranged from 26-57 μ g/g. Samples of unirradiated tubing were also analyzed by ZPI and the reported hydrogen content was < 5 μ g/g.

DISCUSSION

Effect of Thin-Wall Cladding

There was initial concern that the collapse may have occurred because thin-wall cladding was used in the bundles. However, although the nominal wall thickness was less than that of 37-element fuel, the ratio of mean radius to minimum thickness (R/t), which determines the maximum hoop stress due to coolant pressure (pR/t), was comparable to that of current CANDU fuel. For example, the R/t for these prototype bundles (5.585/0.33 = 16.92 and 6.5725/0.355 = 18.51) falls within the range for CANDU-6 (6.35/0.38 = 16.71) and Pickering (7.41/0.38 = 18.51). Therefore, the hoop stress in the 43-element bundles would be comparable to that in current CANDU fuel, hence cladding collapse would not be expected under normal operating conditions.

There have been previous examples of sheath collapse into axial gaps in experimental bundles with cladding up to 0.41 mm thick, under off-normal conditions. However, no defects have been attributed to sheath collapse. The sheath deformation into unsupported plenum regions and interpellet gaps, observed in the previous experimental bundles, appeared almost identical to that observed in these prototype 43-element bundles. This suggests that the current damage must have occurred under similar off-normal conditions.

Effect of Cladding Annealing

The prototype bundles contained fully annealed cladding and the yield strength was lower than that of typical cold-worked and stress-relieved CANDU fuel cladding. Autoclave tests were performed on archive annealed samples and standard stress-relieved cladding samples, to assess the effect of prior heat-treatment on collapse behaviour. There was no sheath collapse into axial gaps up to 3 mm wide under typical CANDU operating conditions, 300°C and 11 MPa, but at temperatures above 600°C the collapse behaviour was reproduced and both materials behaved similarly. This suggests that prior heat treatment had little effect, and confirms that the observed damage must have occurred under off-normal conditions.

Transient Conditions after Pump Flow Stopped

Post-test assessments showed that the coolant flow stagnated following the loss of class IV power, and the fuel string was exposed to an almost stagnant steam atmosphere for at least 8-10 s. Under these conditions, the main heat-transfer process is radiation. Based on the assumption that the heat-transfer coefficient quickly changed from the initial forced convective cooling value (50 kW/m^2) to a value between that for cooling by radiation (0.2 kW/m^2) and poor steam flow (1.0 kW/m^2) , calculations show that sheath temperatures of 600°C were likely, and temperatures of the order of 850°C were possible. The NRU logs show that the surge tank relief valve opened and remained so for 7 s, indicating that the system pressure was greater than 11.2 MPa during this period. The second high-pressure trip shows that the system was pressurized to between 10.5 and 11.2 MPa for approximately 1.5 min following the outage.

Sheath Collapse Conditions

At temperatures between 600 and 850°C, and under a pressure of 11.2 MPa or greater, the Zr-4 cladding would be expected to plastically deform into the open gaps and missing pellet chips. The total sheath stress is the sum of (i) the compressive stresses due to mechanical or hydraulic loads, (ii) bending stresses due to load eccentricity, and (iii) hoop stresses due to coolant pressure,

$$\sigma = \frac{P}{A} + \frac{PRr_e}{I} + \frac{pR}{t}$$

where P is the applied load per element, A is the sheath cross-section area, R is the mean radius, r_e is the eccentricity of the load, I is the moment of inertia of the sheath, p is the coolant pressure and t is the sheath wall thickness. At 1+.2 MPa coolant pressure, the hoop stress alone is equal to 189 MPa in the 11.5 mm diameter elements and 207 MPa in the 13.5 mm diameter elements. However, at 600°C the yield strength of Zr-4 is only ~ 93 MPa. Clearly, the maximum sheath stress exceeded the Zr-4 yield strength, hence plastic flow would be expected.

Temperature Markers

The collapsed zone under the bearing pad in element AHK 12 contained a thin layer of small recrystallized grains on the inside surface of the sheath. Zirconium alloys start to recrystallize only when recovery is almost complete; i.e., after hours at 600° or minutes at 800°C. The presence of alpha grains remote from the collapse zone indicate that the sheath temperature did not exceed 815°C, the alpha-beta transformation temperature. Thus the recrystallized grains indicate that the sheath may have been in the 600 to 800°C range.

Oxide layers 1-3 μ m thick on the outside surface of the sheaths also indicate that high temperatures were achieved. Correlations for weight gain of Zircaloy as a function of time in steam show that for 100 s exposure (the duration of high-pressure excursion due to steam generation in the loop was ~ 1.5 min) a 0.5 μ m oxide layer would grow at 600°C and a 2 μ m layer would grow at 800°C (3). Assuming that the oxide layer prior to the incident was ~ 0.5 μ m, considering the short irradiation period, then temperatures of 600 to 800°C would be required to grow the oxide to 1-3 μ m. Given the uncertainty in the duration of the high-temperature excursion, the estimates based on oxide thickness seem consistent with those based on microstructural observations.

Most of the fuel pellets showed no evidence of abnormal fuel temperatures. However, one sample from AHK 8 showed columnar grain growth at the centre. The high temperature appeared to be localized in one region of the element, as two other samples from AHK 8 showed equiaxed growth. Some fuel samples also showed evidence of crack healing within the pellets, and dendritic grain growth at adjacent hot pellet interfaces.

Location of Damaged Elements in the Bundle

Most of the severe damage in the bundles was in the segment facing the empty (withdrawn) control-rod site at NRU lattice position F21, consistent with the known flux tilt across the loop. However, it is difficult to reconcile the localization of damage with such a reportedly small flux tilt.

Hydride Blisters

Hydride blisters were found in elements 36 and 39 from bundle AHL. Blisters are usually secondary manifestations of primary defects in CANDU fuel. However, the loop in NRU is light-water cooled, so the hydriding damage could be primary or secondary. Known primary defect mechanisms include incomplete or defective closure welds, porous end caps, stress-corrosion cracking and debris fretting. The metallographic examinations eliminated defective welds or porous end caps as likely causes, and fretting and stress corrosion cracking were ruled out, so by elimination the defects were attributed to primary hydriding. It is known that improperly baked CANLUB coatings may be a source of hydrogen, but the source of hydrogen in elements AHL 36 and 39 was not established.

Leak Testing of Collapsed Elements

The helium pressurization tests showed that none of the collapsed elements leaked. This was a significant finding since some of the elements contained deep grooves where the sheath collapsed plastically into gaps between the stack and end cap and between pellets. The maximum measured diametral strain was -16% where the sheath intruded 1.3 mm below the pellet surface. At such severe strains, sheath thinning and possible failure of the cladding were suspected, but the results showed no significant sheath thinning or breach of cladding due to collapse.

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CONCLUSION-

The prototype 43-element bundles overheated when, due to loss of electrical power to the NRU loop pumps, the coolant flow stagnated briefly in the down-flow leg of the U2 loop and then reversed direction as thermal syphoning was established. The stored energy and decay heat were sufficient to cause overheating and overpressurization of the bundles during the brief stagnation period associated with the transition from pumped flow to thermal syphoning.

The cladding collapsed into the gaps between the pellet stack and end caps, between adjacent pellets within the stack, and into missing pellet chips and cracks. The cladding flowed plastically into the gaps due to the reduction in Zr-4 strength at high temperature. However, no defects were attributed to sheath collapse.

Two inner elements in bundles AHL contained defects due to hydride blisters. The defects were attributed to primary hydriding, but the source of the hydrogen could not be conclusively linked to a breach in the cladding, welds or end caps, or to the CANLUB coating, sheath or pellets. The source of the hydrogen was not determined.

The post-irradiation examinations show that although the fuel cladding was subjected to abnormally high temperatures in the range 600-850°C, and pressures above 11.2 MPa, the bundles remained physically intact, and none of the collapsed elements leaked. The results show that thin-wall Zr-4 cladding behaves in a highly ductile manner during a short-term, high-temperature, high-pressure transient of the type encountered when the coolant flow stagnated.

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Parameter -	Mean	Min/Max	Std. Deviation	Before Trip
E-20 Power (MW)	3.382	1.610/3.842	0.442	3.725
Total Loop Power (MW)	5.558	2.044/6.202	0.710	5.9
Inlet Temperature (°C)	238.4	210.0/256.1	16.5	255
Outlet Temperature (°C)	286.1	260.7/292.4	6.5	291
Inlet Pressure (MPa)	11.384	7.499/11.692	0.064	11.49
Outlet Pressure (MPa)	10.871	7.185/11.130	0.192	10.93
Mass Flow (kg/s)	19.53	15.00/21.27	1.42	20.0

Table 1. Typical U-2 Loop Operating Conditions During the Irradiation

Table 2. Power Distribution and Surface Heat Flux in Fuel Bundles

Bundle (pos)	Element	Avg. Power RE - NRE (kW/m)	Max. Power RE - NRE (kW/m)	Flux Tilt Power ^a (kW/m)	Max. Surface Heat Flux ^b (kW/m ²)
AHL (2)	Outer	35.8 - 44.8	38.4 - 48.0	48.8	1350
	Intermediate	22.8 - 28.6	24.5 - 30.6	31.1	860
	Inner	23.3 - 29.2	25.0 - 31.3	31.8	750
AHK (3)	Outer	49.5 - 57 .1	53.1 - 61.2	62.2	1721
	Intermediate	31.6 - 36.4	33.9 - 39.0	39.7	1099
	Inner	32.3 - 37.2	34.6 - 39.9	40.6	957
AHP (4)	Outer	25.0 - 26.6	26.8 - 28.5	29.0	803
	Intermediate	18.4 - 19.5	19.7 - 21.0	21.3	598
	Inner	57.2 - 60.9	61.4 - 65.2	66.3	1563

^a Flux tilt calculated as 1.7% of maximum sustained element linear power from BURFEL.
^b Surface heat flux at power rating due to flux tilt.

Clock Time	Elapsed Time (s)	Event
09:53:56.7	0.0	Reactor trip due to power outage
09:53:57.1	0.4	U2 low-flow trip (10% loss of flow)
09:53:58.1	1.4	High-temperature alert
		(175°C in thermal syphon leg)
09:53:58.8	2.1	Surge tank high-pressure trip (1525 psig)
09:54:00.8	4.1	Surge tank relief valve open (1625 psig)
09:54:01.9	5.2	High-pressure alarm cleared
09:54:07.8	11.1	Relief valve closed
09:54:12.5	15.8	2nd high-pressure trip
09:55:27.4	90.7	High-pressure trip cleared
09:57:57.1	240.4	NRU Class IV power restored
09:58:30.6	273.9	High test-section temperature
10:01:43.3	457.6	Main flow controller cleared
10:03:49.0	592.3	Low-flow trip cleared (pumps on)

Table 3. Summary of Events Recorded on NRU Data Logger 90/11/19





BUNDLE AHL (2.59% Enriched UO₂) 48 kW/m Outer Ring 31 kW/m Intermediate Ring 31 kW/m Inner Ring

BUNDLE AHK (2.59% Enriched UO2) 61 kW/m Outer Ring 39 kW/m Intermediate Ring 40 kW/m Inner Ring

Figure 1. Schematic showing location of damage on bundles. C = complete collapse, c = partial collapse, H = confirmed hydride defect, h = possible hydride blister.



UCS-3137-6 AHK 8 (RE) 5.57X



UCS-3137-7 AHK 8 (Mid) 5.75X

Figure 2. Examples of sheath collapse observed on elements in bundles AHK.

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Figure 3. Micrographs showing sheath collapse under bearing pad of AHL 12, and into pellet chamfers in AHP 36. Top shows general fuel condition and bottom shows polarized light image of sheath at the corresponding collapse zones. Arrows show recrystallized grains on inside surface of collapsed sheath under bearing pad (braze heat-affected zone). Equiaxed grains in annealed tubing where sheath collapsed into pellet chamfers.