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Metallographic Examination of Damaged N Reactor Spent Nuclear Fuel Element SFEC5,4378

S. C. Marschman
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August 1997

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Executive Summary

N-Reactor spent nuclear fuel (SNF) is currently residing underwater in the K Basins at the Hanford site, in Richland, Washington. An Integrated Process Strategy was developed to package, condition, transport, and store this SNF in an interim storage facility. Information required to support the development of the conditioning process and to support the required safety analyses is being obtained from characterization tests conducted on SNF elements from the K Basins. This report presents results of the metallographic examination of specimens cut from an SNF element (Mark IV-E) with *breached* cladding. The element had resided in the Hanford K-West (KW) Storage Basin for at least 10 years after it was discharged from the N Reactor. The storage containers in the KW Basin were nominally closed, isolating the SNF elements from the open pool environment.

Seven specimens from this Mark IV-E outer fuel element were examined using an optical metallograph. Included were two specimens that had been subjected to a conditioning process recommended by the Independent Technical Assessment (ITA) Team, two specimens that had been subjected to a conditioning process recommended in the Integrated Process Strategy (IPS) Report, and three that were in the as-received, as-cut condition. One of the as-received specimens had been cut from the damaged (or breached) end of the element. All other specimens were cut from the undamaged mid-region of the fuel element.

The specimens were visually examined to 1) identify uranium hydride inclusions present in the uranium metal fuel, 2) measure the thickness of the oxide layer formed on the uranium edges and assess the apparent integrity and adhesion of the oxide layer, and 3) look for features in the microstructure that might provide an insight into the various corrosion processes that occurred during underwater storage in the KW Basin. These features included, but were not limited to, the integrity of the cladding and the fuel-to-cladding bond, obvious anomalies in the microstructure, excessive pitting or friability of the fuel matrix, and obvious anomalies in the distribution of uranium hydride or uranium carbide inclusions. Also, the observed metallographic features of the conditioned specimens were compared with those of the as-received (unconditioned) specimens.

Uranium hydride inclusions, uranium carbide inclusions, and pitting (fuel pull out) were observed in each specimen (Figure S.1). The hydride inclusions were identified using a heat-tint technique that caused a halo to form in the uranium matrix surrounding a hydride inclusion. As expected, the most extensive hydride was noted in the specimen from the damaged end of the element. The largest hydride inclusion measured about 250 μm in diameter. This specimen also displayed a large network of macro-cracks. The larger hydride inclusions were somewhat more prevalent in the macro-crack region. Smaller hydrides were distributed throughout the fuel matrix, but they were slightly more prevalent in the vicinity of the larger hydrides and in areas between cracks. Additionally, the fuel matrix adjacent to many of the cracks exhibited a notable halo effect when heat-tinted (Figure S.2). This result indicated that uranium hydride was also distributed in the adjacent fuel matrix and that cracks acted as conduits by carrying hydrogen from the corroded surface deeper into the fuel matrix. Hydride inclusions in the unconditioned specimens from the undamaged area were small, typically less than about 25 μm in diameter. No pattern was evident for the hydride distribution in the undamaged specimens.

Several macro-cracks in the damaged specimen extended a short distance ($\leq 180 \mu\text{m}$) into the Zircaloy-2 fuel cladding (Figure S.3). Nothing indicated the cracks had initiated in the cladding. It appeared the cracks began in the fuel region as a result of local corrosion processes and then propagated into the cladding. An initial breach in the cladding is suspected to have occurred when the fuel element was discharged from the reactor. Water ingress to the fuel core during subsequent storage likely initiated the fuel corrosion process. As the corrosion reaction proceeded, the fuel swelled. Thus, the combination of corrosion-induced fuel swelling and stress-induced crack propagation into the cladding resulted in the cladding becoming distorted and subsequently rupturing to relieve the stress.

Portions of a very thin oxide layer were observed along the fuel edges of most specimens. The layer appeared to be porous and was obviously nonadherent. Typically, it had pulled away from the fuel and appeared to be flaking off (Figure S.4). In all but one case, the maximum thickness of the layer was $3 \mu\text{m}$. The one exception was a very short fragment about $10 \mu\text{m}$ thick on an ITA-conditioned specimen.

The fuel-to-cladding bond appeared to be intact along its full length in each of the specimens. Uranium hydride inclusions, uranium carbide inclusions, and local areas of fuel pull out (shallow pits) were observed immediately adjacent to the bond on several specimens at a number of locations (Figure S.5). However, the integrity of the bond did not appear to be affected. No stringers or extensive void networks were found along the exposed length of the bond region, indicating the fuel-to-cladding interface did not serve as a preferential path for the migration of hydrogen or the ingress of corrosion.

The microstructure of the uranium metal fuel in the as-etched condition had a *swirled* appearance, and its grains were not distinct (Figure S.6). Thus, the grain size of the fuel was not measured.

Only a few subtle differences were apparent between the two undamaged and unconditioned specimens, the IPS-conditioned specimens, and the ITA-conditioned specimens. The fuel surface of the IPS-conditioned specimens was more friable than the others, and some of the pull out areas appeared to be hydrided areas with diffuse halos (Figure S.7). These halos indicated that some of the hydrides near the surface decomposed during the conditioning process, leaving a friable uranium structure. The ITA-conditioned specimens typically exhibited smaller hydride inclusions with less distinct halos than did the IPS-conditioned specimens, indicating that some hydride decomposition also occurred during the ITA-recommended conditioning process. Although indications were present that hydride decomposition occurred during both conditioning processes, obvious uranium hydride inclusions remained in each of the conditioned specimens (Figure S.8). Thus, neither conditioning process completely decomposed the uranium hydride phase.

Figure S.1. Fuel-to-Cladding Bond Area with Typical Hydride Inclusions, Carbide Inclusions, Minor Fuel Pull Out, and Pitting; After Heat Tint; Specimen 5-S2-C. Specimen 5-S2-C was removed from the as-received undamaged mid-length of an N-Reactor Fuel Element (see also Section 3.1).

Figure S.2. Small Portion of Large Crack Network Terminating at Cladding, Hydrided Areas Along Cracks, Hydride Inclusions, and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A. Specimen 5-S1A-A1A was removed from the as-received undamaged mid-length of an N-Reactor Fuel Element (see also Section 3.1).

Figure S.3. Tip of Macro-Crack Penetrating into the Cladding; After Heat Tint; Specimen 5-S1A-A1A (see also Section 3.1).

Figure S.4. Portion of Thin, Nonadherent, Porous Uranium Oxide Layer ($\leq 3 \mu\text{m}$ Thick); After Heat Tint; Specimen 5-S2-B. Specimen 5-S2-B was removed from the undamaged mid-length of an N-Reactor outer fuel element and subjected to an oxidizing conditioning process (see also Section 3.2).

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Abbreviations and Acronyms

ID	inside diameter
IPS	Integrated Process Strategy
ITA	Independent Technical Assessment Team
kPa	kilo Pascals
KW	K West
MCO	multi-canister overpack
m/s	meters per second
OD	outside diameter
ppm	parts per million
PNNL	Pacific Northwest National Laboratory
PTL	Postirradiation Testing Laboratory
SFEC	single fuel element canister
sfm	surface feet per minute
SNF	spent nuclear fuel

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1.0 Introduction

1.1 Background

Data on the extent of uranium oxide and hydride formation in Hanford spent nuclear fuel (SNF) are needed to support the design, transportation, and interim storage of the multi-canister overpack (MCO), as well as to address questions related to SNF behavior during fuel staging and stabilization (or conditioning). Data related to corrosion characteristics and corrosion rates of the fuel and cladding are necessary for making decisions on wet versus dry packaging, gas inventory, and vented or sealed containers. Also, knowledge of the corrosion characteristics is directly applicable to making decisions related to MCO materials compatibility, dimensional considerations, and degradation during staging in the MCO, if they are water-filled (Lawrence et al. 1995).

To obtain some of this data, three Mark IV-E SNF elements from the Hanford K West (KW) Basin were selected for examination at the Pacific Northwest National Laboratory (PNNL). Two of these (designated as SFEC5,4378 and SFEC10,4366) were fuel assembly outer elements. The third (SFEC8,1990) was a fuel assembly inner element. Each of the selected fuel elements was transferred from the storage basin to the PNNL hot cells in its own single fuel element canister (SFEC) filled with KW Basin water. Each fuel element was uniquely identified by the SFEC in which it was shipped and the multi-element storage container from which it was removed at the KW Basin. For example, SNF element SFEC5,4378 was transferred in SFEC number 5 after it was removed from storage container number 4378.

The three elements exhibited damage that began upon their discharge from the reactor and progressed in varying degrees while the elements were in long-term underwater storage. Obvious visible damage was located at one end of each fuel element and included the end-cap region. The cladding of element SFEC5,4378 appeared to have burst open, and the cladding of SFEC10,4366 was missing from the damaged area. Consequently, a significant amount of the uranium metal fuel in each of these outer elements had been exposed to the container water during storage in the basin. Fuel was obviously missing from the damaged area of each, and the exposed surface of the remaining metal fuel was severely corroded. Element SFEC8,1990 appeared to be intact; its visible damage consisted only of an obvious chip in its end cap. Neither the underlying metal fuel nor any swelling of the cladding was clearly visible on this inner element.

The three elements selected represented a spectrum of the damage exhibited by the fuel elements currently stored in the KW Basin. Specimens for optical metallographic examination were taken from SNF element SFEC5,4378 because it most visibly displayed the corrosion damage that was expected to result in the formation of uranium hydride (UH_3) and the oxidation of the uranium metal in oxygen-depleted water.

1.2 Purpose

Metallographic examination of selected SNF specimens was performed to help characterize the irradiated fuel material of SNF elements with breached cladding that had been in underwater storage in the Hanford KW Basin for more than 10 years. The primary purpose was to identify uranium hydride inclusions in the uranium metal matrix. Additional purposes were to 1) measure the thickness and identify the physical characteristics of the uranium oxide layer, 2) examine the fuel microstructure for other visible changes that might be attributable to long-term storage, 3) identify morphological features or anomalies that might warrant additional examination, and 4) compare the metallographic characteristics of pre- and post-conditioned SNF samples.

1.3 Scope

A total of seven specimens from the N-reactor outer fuel element SFEC5,4378 were examined using an optical metallographic microscope. These specimens included two that had been subjected to a conditioning process recommended by the Independent Technical Assessment Team (ITA 1994), two specimens that had been subjected to a conditioning process recommended in the Integrated Process Strategy Report (IPS 1995), and three that were in the as-received, as-cut condition. One as-received specimen had been cut from the damaged (or breached) end of the element. The remaining six had been cut from the unbreached mid-region of the fuel element.

The specimens were visually examined to 1) verify whether or not uranium hydride inclusions were present in the uranium metal fuel and, if present, assess their distribution; 2) measure the thickness and assess the apparent integrity and adhesion of the oxide layer formed on the uranium edges; and 3) look for features in the microstructure that might provide insight into the various corrosion processes that occurred during underwater storage in the KW Basin. These features included, but were not limited to, the integrity of the cladding and the fuel-to-cladding bond, obvious anomalies in the microstructure, excessive voids, pitting or friability of the fuel matrix, and obvious anomalies in the distribution of uranium hydride and carbide inclusions.

2.0 Material Examination and Specimen Preparation

2.1 Material Examination

Fuel element SFEC5,4378 is a Mark IV-E outer fuel element that was discharged from the 105 N Reactor during normal reactor operations. A brief description of the N-Reactor fuel element prior to irradiation is given in the following paragraphs. A more detailed description is provided in Appendix A of Abrefah et al. (1994).

2.1.1 Description of the Mark IV-E Fuel Element

The typical Mark IV-E fuel assembly consists of two concentric tubular fuel elements as shown in Figure 2.1. The outer element has a nominal outer and inner diameter of 6.15 cm (2.42 in.) and 4.31 cm (1.7 in.), respectively. The corresponding diameters of the inner element are 3.25 cm (1.28 in.) and 1.29 cm (0.51 in.), respectively. Both elements have a nominal length of 66.3 cm (26.1 in.), and an enrichment of 0.947% ^{235}U .

The Mark IV fuel elements were fabricated by co-extruding a tube of uranium Alloy 601 fuel (beta heat treated) with Zircaloy-2 cladding material (fully annealed), thereby forming a solid-state diffusion bond between the metal uranium core and its cladding. After extrusion, the clad fuel was cut to the desired length, and the ends were machined; Zircaloy-2 end-caps were brazed in place and then fusion-welded to form a hermetically sealed fuel element. The braze ring material, which was placed between the uranium core and the Zircaloy-2 end cap, was an alloy of zirconium and beryllium (5 wt %). The primary alloying constituents of uranium Alloy 601 are aluminum (700-900 ppm), carbon (365-735 ppm), and iron (300-400 ppm).

A system of four inner supports and two W springs were spot-welded to the outside of the inner element. This system ensured proper alignment of the inner annulus and locking with the outer element when the two elements were combined to form the fuel assembly. Eight outer supports were welded to the outside of the outer element to ensure proper alignment of the fuel assembly when inserted into the pressure tube of the reactor. The supports and springs were made of Zircaloy-2. Support shoes made of low-carbon steel were crimped into the outer supports to protect the fuel assembly during insertion and discharge from the reactor and to provide a wear couple that was compatible with the pressure tube. Finishing operations included heating the element in an autoclave to give the cladding surface a thin, adherent, black layer of zirconium oxide.

2.1.2 Fuel Specimens

The specimens identified in Table 2.1 were selected from SNF element SFEC5,4378 for optical metallographic examination. Of the seven specimens, two (5-S2-B and 5-S2-J) had been subjected to an ITA-recommended conditioning process, two (5-S2-F1 and 5-S2-G) had been subjected to an IPS-recommended conditioning process, and three remained in the as-received, as-cut (unconditioned)

condition. One of the unconditioned specimens (5-S1A-A1A) was from the damaged (breached) end of the element. The remaining six of the seven specimens had been cut from the undamaged mid-length region of the outer fuel element.

An illustration showing fuel element SFEC5,4378 and the location in the element from which each metallographic specimen was cut is provided in Figure 2.2. The 0° orientation and the top end of the element were arbitrarily designated as such for use during the hot cell examination. Their relation to the actual orientation of the element when it was in the reactor core is not known.

Each specimen was a segment of uranium metal (fuel) bonded on both sides to its Zircaloy-2 cladding. The specimens ranged in sector width from about 20° to 25° and were initially either ~2.5 cm (0.98 in.) or 5.0 cm (1.97 in.) long. The specimens were shortened to either ~1.25 cm (0.49 in.) or 2.5 cm (0.98 in.) prior to being mounted for metallographic examination. Specimen 5-S2-J was cut in half for metallographic examination of one transverse face (cross-section) and one longitudinal face (lengthwise). The upper half of this specimen (designated 5-S2-J1) was mounted transversely, and its top face was examined. The lower half (designated 5-S2-J2) was mounted longitudinally, and its 0° face was examined. Typical transverse and longitudinal metallographic mounts are illustrated in Figure 2.3.

The ITA-recommended conditioning process consists of drying the fuel elements in a flowing inert gas, followed by passivating them in a flowing gas (ITA 1994). The IPS-recommended conditioning process consists of drying the fuel elements in a vacuum, followed by passivating them in a flowing gas (IPS 1995). A summary of the conditioning process for each specimen is presented in Table 2.2. These processes were performed in an instrumented furnace system to assess the effectiveness of each process in passivating the specimens and in removing hydrogen from them. The furnace system was located in a hot cell and was operated remotely.

2.2 Specimen Preparation

2.2.1 Sectioning

The specimens were first cut from the element on a milling machine using a 15.2-cm (6-in.) diameter, silicon carbide, slitting-saw blade. Blade speed was maintained at about 0.76 surface m/s (150 surface feet per minute [sfm]). Each specimen was cooled during cutting by flowing argon over the cutting area through a vortex cooling tube (Chilcut-II). The argon was supplied at a gage pressure of about 100 psi (690 kPa) and a flow rate of about 7080 cm³/s (15 scfm). A K-type thermocouple was attached to the specimen to monitor specimen surface temperature during cutting. The feed rate of the milling machine was manually adjusted to maintain indicated specimen temperatures at no more than about 15°C above the hot cell ambient air temperature. Upon removal from the element, the specimens were stored in individual glass vials which, in turn, were stored within a canister of ultra high purity argon (≥99.999% argon) to minimize moisture absorption and corrosion.

Specimens 5-S1A-A1, 5-S2-J, 5-S2-F1, and 5-S2-G were further subdivided for metallographic mounting. Each was cut transversely into two sections of approximately equal length (~1.25 cm). A low-speed precision cut-off saw (Buehler Isomet or LECO VC-50) with a diamond wafering blade was used at blade speeds in the range of 0.66 to 1.02 surface m/s (130 to 200 sfm). Specimen 5-S2-J was cut wet by slowly dripping Hyprez O.S. Type IV Lubricant on the blade during cutting. The resulting subsections, 5-S2-J1 and 5-S2-J2, were cleaned with isopropyl alcohol and deionized water. The other specimens were cut dry in a flowing stream of pre-purified argon ($\geq 99.998\%$ argon). Prior to being mounted for metallographic examination, the specimens were stored within a canister of ultra high purity argon. After being mounted and when not being prepared or examined, the specimens were temporarily stored under roughing pump vacuum (~0.15 Pa) to reduce the surface oxidation rate until their examination was completed.

2.2.2 Metallographic Preparation

Each specimen was mounted in a cold curing epoxy or fiberglass resin within a stainless steel ring. Stainless steel coupons were typically included to improve edge retention for the measurement of oxide layer thickness. The specimen was ground progressively on 180 through 1200 grit silicon carbide paper with Hyprez O.S. Type IV Lubricant, and typically rough polished with a 1- μm diamond compound suspended in the Hyprez lubricant. The depth of grinding ranged from about 2.1 to 3.1 mm. To reduce fuel pull out, two specimens (5-S2-G1 and 5-S1A-A1A) were rough polished with 0.3 μm alumina in a 2% chromic acid solution, instead of the 1- μm diamond compound and Hyprez Lubricant.

Immediately prior to examination, the specimen was attack polished using a solution of 2% chromic acid with either 0.3- μm or 0.05- μm alumina, examined in the as-polished condition, and then heat tinted (partially oxidized) for at least 15 minutes under a halogen lamp to highlight the uranium hydride inclusions. When the specimen was successfully heat tinted, the hydride inclusions (dark) were surrounded by a silver/yellow glow or halo in the fuel matrix. An example of this is shown in Figure 2.4. This halo effect resulted from the lower oxidation rate of the uranium in or immediately adjacent to the area affected by hydride. The method used is similar to that of Gardner and Riches (1956). Specimen preparation is described in detail in Appendix A. An overview of the preparation given each specimen is included in Table 2.1.

Two specimens (5-S2-A and 5-S2-C) were also electrolytically polished and etched in an effort to examine the grain structure and to determine the grain size of their uranium matrix. The method of preparation and the electrolytic solutions used are given in Appendix B (Gardner and Riches 1956; ASM Handbook 1985).

Table 2.1. Specimens Examined on the Optical Metallograph

Metallographic Specimen	Location in Fuel Element	Surface Examined	Comments
SFEC5,4378-S1A-A1A ^(a)	Damaged end, 0° - 22° sector	Top half, 0° face of 5-S1A-A (longitudinal mount)	<p><u>Condition:</u> as-received and subsectioned from 5-S1A-A, ~1.25 cm (0.49 in.) long.</p> <ol style="list-style-type: none"> 1. Subsectioned under argon; mounted in Buehler Epo-Thin epoxy with metal coupons for edge retention. 2. Ground through 1200 grit SiC paper to a depth of ~2.67 mm (0.11 in.), attack polished with 0.3 µm and then 0.05 µm alumina (did not rough polish with diamond compound), and heat tinted.
SFEC5,4378-S2-A	Mid-length, 0° - 22° sector	Top end of 5-S2-A (transverse mount)	<p><u>Condition:</u> as-received and sectioned, ~2.5 cm (0.98 in.) long.</p> <ol style="list-style-type: none"> 1. Used initially to refine the metallographic technique; mounted in Titan Fiberglass Finishing Resin without metal coupons. 2. Ground through 1200 grit SiC paper, rough polished with 1-µm diamond compound, attack polished with 0.3-µm alumina, and heat tinted. 3. Electro-polished and electro-etched for grain structure.
SFEC5,4378-S2-B	Mid-length, 23° - 45° sector	45° face of 5-S2-B (longitudinal mount)	<p><u>Condition:</u> after ITA-recommended conditioning process (Furnace Run #2), ~2.5 cm (0.98 in.) long.</p> <ol style="list-style-type: none"> 1. Mounted in Buehler Epo-Thin epoxy with metal coupons for edge retention. 2. Ground through 1200 grit SiC paper, rough polished with 1-µm diamond compound, attack polished with 0.3-µm alumina, and heat tinted.
SFEC5,4378-S2-C	Mid-length, 46° - 68° sector	68° face of 5-S2-C (longitudinal mount)	<p><u>Condition:</u> as-received and sectioned, ~2.5 cm (0.98 in.) long.</p> <ol style="list-style-type: none"> 1. Used initially to refine the metallographic technique; mounted in Titan Fiberglass Finishing Resin without metal coupons. 2. Ground through 1200 grit SiC paper, rough polished with 1-µm diamond compound, attack polished with 0.05-µm alumina, and heat tinted. 3. Electro-polished and electro-etched for grain structure.
(a) This reduces to the abbreviated specimen number "5-S1A-A1A."			

Table 2.1. Continued

Metallographic Specimen	Location in Fuel Element	Surface Examined	Comments
SFEC5,4378-S2-F1A	Mid-length, 180° - 202° sector	Top end of 5-S2-F1 (transverse mount)	<p><u>Condition:</u> after IPS-recommended conditioning process (Furnace Run #6) and subsequent subsectioning of 5-S2-F1, ~1.25 cm (0.49 in.) long.</p> <ol style="list-style-type: none"> 1. Subsectioned under argon; mounted in Buehler Epo-Thin epoxy with metal coupons for edge retention. 2. Ground through 1200 grit SiC paper to a depth of ~2.13 mm (0.08 in.), rough polished with 1-μm diamond compound, attack polished with 0.3-μm alumina, and heat tinted.
SFEC5,4378-S2-G1	Mid-length, 270° - 298° sector	Top end of 5-S2-G1 (transverse mount)	<p><u>Condition:</u> after IPS -recommended conditioning process (Furnace Run #5) and subsequent subsectioning of 5-S2-G1, ~1.25 cm (0.49 in.) long.</p> <ol style="list-style-type: none"> 1. Subsectioned under argon; mounted in Buehler Epo-Thin epoxy with metal coupons for edge retention. 2. Ground through 1200 grit SiC paper to a depth of ~3.07 mm (0.12 in.), rough polished with 1-μm diamond compound, attack polished with 0.3-μm alumina, and heat tinted. 3. Re-ground from 600 through 1200 grit SiC paper, attack polished with 0.3-μm and then 0.05-μm alumina (did not rough polish with 1-μm diamond compound), and heat tinted.
SFEC5,4378-S2-J1	Mid-length, 338° - 360° sector	Top end of 5-S2-J1 (transverse mount)	<p><u>Condition:</u> after ITA-recommended conditioning process (Furnace Run #1) and subsequent subsectioning of 5-S2-J1, ~1.25 cm (0.49 in.) long.</p> <ol style="list-style-type: none"> 1. Subsectioned using Hyprez O.S. Type IV Lubricant; mounted in Buehler Epo-Thin epoxy with metal coupons for edge retention. 2. Ground through 1200 grit SiC paper, rough polished with 1-μm diamond compound, attack polished with 0.3-μm alumina, and heat tinted.
SFEC5,4378-S2-J2	Mid-length, 338° - 360° sector	Bottom half, 360° face of 5-S2-J2 (longitudinal mount)	<p><u>Condition:</u> after ITA-recommended conditioning process (Furnace Run #1) and subsequent subsectioning of 5-S2-J2, ~1.25 cm (0.49 in.) long.</p> <ol style="list-style-type: none"> 1. Subsectioned using Hyprez O.S. Type IV Lubricant; mounted in Buehler Epo-Thin epoxy with metal coupons for edge retention. 2. Ground through 1200 grit SiC paper, rough polished with 1-μm diamond compound, attack polished with 0.3-μm alumina, and heat tinted.

Table 2.2. Summary of ITA and IPS Test Conditions

Conditioning Process	Specimen	Test Conditions
ITA ^(a)	SFEC5,4378-S2-J1	Dried in two stages, first at 100°C for 10 hours in argon flowing at 100 cm ³ /min, and then at 300°C for 24 hours in argon flowing at 100 cm ³ /min. Passivated at 250°C for 10 hours in an atmosphere consisting of 2% oxygen and 98% argon at a pressure of 101 kPa (1 atm).
	SFEC5,4378-S2-B	Dried in two stages, first at 100°C for 10 hours in argon flowing at 100 cm ³ /min, and then at 300°C for 24 hours in argon flowing at 100 cm ³ /min. Passivated at 250°C for 10 hours in an atmosphere consisting of 2% oxygen and 98% argon at a pressure of 101 kPa (1 atm).
IPS ^(b)	SFEC5,4378-S2-G1 [with free water added]	Cold vacuum dried at 8.0 kPa (60 torr) and 50°C, followed by hot vacuum drying at 8.0 kPa and 300°C. Passivated at 200°C for 10 hours in an atmosphere consisting of 2% oxygen and 98% argon at a pressure of 101 kPa (1 atm).
	SFEC5,4378-S2-F1 [with free water added]	Cold vacuum dried at 8.0 kPa (60 torr) and 50°C, followed by hot vacuum drying at 8.0 kPa and 300°C. Passivated at 250°C for 10 hours in an atmosphere consisting of 2% oxygen and 98% argon at a pressure of 101 kPa (1 atm).
(a) ITA-recommended process: dried in flowing inert gas, followed by passivation in flowing gas.		
(b) IPS-recommended process: dried under vacuum, followed by passivation in flowing gas.

Figure 2.1. Mark IV-E Fuel Assembly

SG96120047.1

Figure 2.3. Illustration of Transverse and Longitudinal Metallographic Mounts

SNF-127

17μm

Figure 2.4. Uranium Hydride (UH_3) Inclusion with Characteristic Halo after Heat Tint

3.0 Results and Discussion

The specimens were visually examined using a remotely operated Bausch & Lomb Research Metallograph. Bright-field illumination and polarized light were used at magnifications ranging from about 35x to 750x. Photographs were taken at various magnifications to document typical features, selected areas of interest, and apparent anomalies. These features or areas of interest included fuel and cladding oxide layers, anomalies in the fuel-to-cladding bond region, cracks in the microstructure, uranium hydride and uranium carbide inclusions in the fuel region, areas of pitting or pull out in the fuel region, and the apparent grain structure of the fuel matrix.

Selected photomicrographs displaying representative or salient features of each specimen are presented and described in this section. For presentation, the specimens are grouped by the conditioning process to which they were subjected prior to examination, that is 1) unconditioned (as-received and sectioned), 2) ITA-recommended process, and 3) IPS-recommended process. The specimens and the associated conditioning process for each are listed in Tables 2.1 and 2.2. Features of the unconditioned specimens are described in Section 3.1. Features of the ITA-conditioned specimens and the IPS-conditioned specimens are described in Sections 3.2 and 3.3, respectively. The results are compared and discussed in Section 3.4.

3.1 Unconditioned Specimens 5-S2-A, 5-S2-C, and 5-S1A-A1A

Specimens 5-S2-A (transverse) and 5-S2-C (longitudinal) are considered to be representative of the undamaged mid-length region of the fuel element after removal of the element from wet storage, prior to being conditioned or passivated. Their features are described in Sections 3.1.1 and 3.1.2, respectively.

Specimen 5-S1A-A1A (longitudinal) is considered to be representative of the mildly damaged area at the breached end of the fuel element after removal of the element from wet storage, prior to being conditioned or passivated. This specimen displayed several interesting features, which are described in Section 3.1.3.

3.1.1 Specimen 5-S2-A

Typical features on the top face of Specimen 5-S2-A are shown in Figures 3.1 through 3.7 and are described in the following paragraphs.

- The mid-fuel region of the as-polished, transverse surface is shown in Figures 3.1 and 3.2 before and after heat tinting, respectively. Uranium carbide inclusions (light colored, angular) and small pits or local oxidation spots (dark, near-circular specks) are apparent in both figures. Pitting of this nature was shown in subsequent specimens to result from pull out of the fuel primarily while rough polishing with 1- μ m diamond compound. Uranium hydride inclusions (dark pits or spots highlighted by the characteristic bright halos) are obvious in the latter figure, along with some streak-like stains resulting from minor surface contamination. The hydride inclusions observed were small, typically less than about 25 μ m in diameter. Their distribution over the surface is similar to that displayed in Figure 3.2. No distribution gradient is apparent.

- Higher magnification views of very small ($\leq 10\text{ }\mu\text{m}$), but also very common, hydride inclusions are shown in Figures 3.3 and 3.4. The very small angular white chips that are apparent in these two figures are fragments of uranium carbide inclusions.
- This specimen was electrolytically etched to measure the grain size of the uranium matrix. However, the grains were not discernable for measurement at magnifications in the range of 50x to 750x. The typical as-etched surface appearance in the mid-fuel region is shown in Figures 3.5, 3.6, and 3.7.
- No oxide layer was observed on the edges of this specimen. This condition was expected because the specimen was not mounted to preserve its edges.

3.1.2 Specimen 5-S2-C

Features typical of the longitudinal surface (67° face) of the specimen are shown in Figures 3.8 through 3.16 and are described as follows.

- The appearance of the as-polished surface in the mid-fuel region is shown in Figure 3.8. The distribution of uranium carbide inclusions and the amount of pitting are similar to those seen on the transverse face of 5-S2-A.
- Figures 3.9, 3.10, and 3.11 display the same area before and after the specimen was heat tinted. Several very small hydride inclusions ($< 5\text{ }\mu\text{m}$ diameter) are located immediately adjacent to three uranium carbide inclusions. A few even smaller hydride inclusions (with halos) are evident elsewhere in the fuel matrix. Small dark specks of local surface oxide are also visible in the fuel matrix. Overall, the hydride inclusions seen throughout the surface of this specimen were small (typically less than about $25\text{ }\mu\text{m}$ in diameter) and similar in morphology to those seen in specimen 5-S2-A. The inclusions appeared to be randomly distributed over the surface, and no concentration gradient was obvious.
- Different areas along the fuel-to-cladding bond are shown before heat tinting in Figure 3.12, and after heat tinting in Figures 3.13 and 3.14. Carbide inclusions located adjacent to the bond region, similar to that shown in Figure 3.12, were common and had no visible effect on the integrity of the bond. No separation of fuel from its cladding was observed along the bond, and there was no visible indication that the integrity of the fuel-to-cladding bond had been degraded. Hydride inclusions, carbide inclusions, and minor fuel pull out and pitting are apparent in Figures 3.13 and 3.14. The irregularly shaped dark region that is shown extending into the fuel from the bond interface was present at various lengths in all specimens that were in the heat-tinted condition. This dark region is believed to be a region where submicron-sized fuel particles were pulled from the matrix while the specimen was being polished. In other specimens that were viewed after they were heat tinted, less extensive regions with a similar appearance occasionally exhibited the halos characteristically displayed by uranium hydride inclusions (for example, see Figures 3.76 through 3.78). The thin, black line immediately adjacent to the cladding and the wider, light-colored band ($\sim 4\text{ }\mu\text{m}$ wide) were common in the bond area of all

specimens and were visibly enhanced by the heat tint. They are believed to be artifacts of polishing which are attributed to the difference in hardness between the uranium metal fuel and its Zircaloy-2 cladding.

- The surface appearance in the mid-fuel region after electrolytic etching is shown in Figure 3.15. The grains of the uranium metal could not be discerned for measurement at magnifications in the range of 50x to 750x. The appearance of the microstructure was similar to that of specimen 5-S2-A (compare Figure 3.5).
- A very short remnant of an oxide layer was observed on the edge of the fuel, as shown in Figure 3.16. This layer appeared to be porous, in the process of flaking off, and less than 2 μm thick. The presence of this remnant was unexpected because this specimen was not mounted with the specific intent of preserving its edges.

3.1.3 Specimen 5-S1A-A1A

The longitudinal surface (0° face) of this specimen displayed several significant features. These features included an extensive network of macro-cracks, many large uranium hydride inclusions, the largest hydride inclusion ($\leq 250\text{-}\mu\text{m}$ rough diameter) seen among the specimens examined, and only a partial oxide layer on the damaged end of the fuel. The thickness of this oxide was less than 2 μm . These features are shown in Figures 3.17 through 3.34, which are described in the following paragraphs. All the figures, except Figure 3.34, show the specimen in the heat-tinted condition.

- Two major cracks in the fuel matrix, one near the element ID side of the specimen and the other near the OD side, apparently originated near the cladding at the breached end of the specimen. The cracks branched down and across the mid-length of the fuel, forming a moderately extensive network of macro-cracks in the fuel. Several of the cracks penetrated into the cladding; the maximum observed depth of penetration was about 180 μm . Within the fuel matrix, the visible crack network appeared to end at or near the bottom of this specimen ~1.25-cm (0.49 in.) in length. After being heat tinted, the fuel along the sides of many cracks displayed a bright glowing band with a diffuse edge that was similar in appearance to the halo that highlighted uranium hydride inclusions. This indicated the fuel adjacent to the cracks was affected by the hydride and suggests the cracks acted as conduits by carrying hydrogen from the corroded surface region deeper into the fuel matrix. Figures 3.17 through 3.25 are part of a series of photomicrographs taken at low magnification showing the major areas of interest.^(a) Figures 3.17, 3.18, and 3.19 show the upper half of the specimen near the OD side of the element. Figures 3.20 and 3.21 show the lower third on the OD side. Figures 3.22, 3.23, and 3.24 display the top corner, approximate mid-length, and bottom corner areas, respectively, near the ID side of the element. Figures 3.19, 3.23, and 3.25 form a rough composite image of the specimen from the OD to ID side, just above mid-length.

(a) The white, and sometimes dark, gum-like material having distinct edges and located within the macro-cracks is the epoxy material that was used to vacuum impregnate the specimen prior to polishing.

- Most of the larger hydride inclusions were located at or above the mid-length region of the specimen. Two hydrides that are typical of those in the crack branches are shown in Figures 3.26 and 3.27. One hydride located on the top edge of the specimen and one located about 2 mm from the bottom edge are shown in Figures 3.28 and 3.29, respectively. The fine structure of the hydride-affected material is apparent in these figures.
- Smaller hydride inclusions (less than about 25 μm in size) were slightly more numerous here than in the undamaged specimens. They were distributed over the surface, but were found more commonly in the vicinity of larger hydrides and crack branches. A typical small inclusion located near the bottom edge of the specimen is shown in Figure 3.30.
- Higher magnification views of three representative macro-cracks penetrating into the cladding are shown in Figures 3.31, 3.32, and 3.33. These crack tips correspond to the fuel cracks shown in Figures 3.20 and 3.21. No indication showed the cracks had initiated in the cladding. Instead, it appeared they originated in the fuel region and propagated into the cladding.
- Only a very short segment of oxide was observed at the exposed edge of the fuel, even though this specimen was prepared using standard metallographic methods to improve edge retention. The thickness of this oxide segment was less than 2 μm . The segment is barely visible in Figure 3.34. The fuel immediately under the oxide appeared to be porous, but no positive indication was found that the dark, porous region contained hydrides. When heat tinted, a few potential small hydride inclusions were seen in the area; however, they were surrounded by an accumulation of very small white chips, instead of the expected halos. The porosity was attributed to degradation resulting from corrosion. The white chips were fragments of uranium carbide inclusions that fractured during polishing; they are similar to those shown in Figure 3.31. The oxide segment was located on the breached end, top edge shown in Figure 3.17.
- The fuel-to-cladding bond appeared intact along its full length on both the ID and OD sides of the fuel.

3.2 ITA-Conditioned Specimens 5-S2-B, 5-S2-J1, and 5-S2-J2

Specimens 5-S2-B (longitudinal), 5-S2-J1 (transverse), and 5-S2-J2 (longitudinal) are considered representative of the undamaged mid-length region of the fuel element after completing the ITA-recommended conditioning process. No significant anomalies were seen. Each specimen exhibited nearly the same amount of, or perhaps slightly fewer, hydrides than the unconditioned specimens from the mid-length of the element. All of the hydrides were small (less than about 50 μm in size) and, with a few notable exceptions in 5-S2-J2, most had very diffuse halos. The diffuse halos suggest that some hydride decomposition may have happened during the conditioning process. Specimen 5-S2-J2 turned darker, and darkened faster, than the others when heat tinted. It also exhibited notably more fuel pull out or pitting than the others. Oxide layers at the fuel edges were observed only on 5-S2-B and 5-S2-J1. The thickness of the oxide layers was typically less than 2 or 3 μm , but one very short, nonadherent segment on 5-S2-B was about 10 μm thick. The features of specimens 5-S2-B, 5-S2-J1, and 5-S2-J2 are described in Sections 3.2.1, 3.2.2, and 3.2.3, respectively.

3.2.1 Specimen 5-S2-B

Features of the longitudinal surface (45° face) of the specimen are shown in Figures 3.35 through 3.44 and are described below.

- The heat-tinted surfaces at the OD side and near the mid-fuel region are shown in Figures 3.35 and 3.36, respectively. Compared to the others, this specimen oxidized (darkened) very slowly during its heat tint and did not darken significantly. The reason for this is not known. The hydride inclusions are seen to be very small, typically less than about 10 μm in size, and their halos are very diffuse. Uranium carbide inclusions and a small amount of pitting and fuel pull out are also obvious in the figures. Some pull out is apparent in the fuel-to cladding interface region.
- Higher magnification views of the hydride inclusions typically seen in this specimen are shown in Figures 3.37 and 3.38. One hydride inclusion is also apparent at the residual carbide inclusion shown in the former figure.
- Portions of an oxide layer were present along the edges of the fuel at the top and bottom ends of the specimen. These portions appeared to be reasonably adherent in some areas, but flaking in others. In general, the oxide layer was broken or flaking, and was less than 3 μm thick. An adherent, porous section of the oxide layer along the top edge of the specimen is shown in Figures 3.39 and 3.40. Its thickness ranged from about 1.5 to 3 μm . Along the bottom edge, the layer was less adherent and apparently in the process of separating and flaking off, as shown in Figures 3.41, 3.42, and 3.43. These thicknesses ranged from about 1.2 to 3 μm , equivalent to those at the top edge. One short segment of oxide as thick as about 10 μm , but less than about 0.2 mm long, was also observed at the bottom edge of the specimen (Figure 3.44). This segment was the thickest one seen on any of the specimens examined.
- Oxide thickness measured at one location on the cladding OD surface was less than 1.3 μm .

3.2.2 Specimen 5-S2-J1

Overall, relatively few hydrided areas with large distinct halos were seen in this specimen. Some fuel pull out occurred during polishing, but it was not excessive. Relatively few dark inclusion or pits were observed; all were small, measuring less than about 50 μm in diameter. Typical features of the top surface of this specimen are shown in Figures 3.45 through 3.53 and are described in the following paragraphs.

- The heat-tinted surface at the ID side and near the mid-fuel region is shown in Figures 3.45 and 3.46, respectively. The hydride inclusions are small, less than about 35 μm in size. With few exceptions, their halos are either very narrow and distinct or somewhat broad and diffuse. Uranium carbide inclusions and a very small amount of pitting (dark) and fuel pull out are also apparent in the figures. The unusually bright hydride inclusion near the center of Figure 3.46 is shown at higher magnification in Figure 3.47. Other typically small hydrides, either isolated or at carbide inclusions, are shown in Figure 3.48.

- The fuel-to-cladding bond appeared intact along its full length on both the ID and OD sides. Several carbide inclusions were present at the fuel interface, but they had no apparent effect on the integrity of the bond. Two such areas on the ID side are shown in Figures 3.49 through 3.52. Areas of hydride were also present in the fuel matrix at the carbide inclusions, as indicated by the light halo near the carbides. These halos were more distinct at lower magnifications. The dark hydride inclusion in Figure 3.49 is displayed under polarized light in Figure 3.50. The dark particles on the cladding surface shown in Figures 3.51 and 3.52 are believed to be oxidized or hydrided fuel residue resulting from polishing, indicating the friability of the fuel surface.
- Portions of an oxide layer were seen flaking off at both edges of the fuel (the 338° and 360° sides). The flaking layers appeared to be porous and had a maximum thickness of about 2 μm . The small fragment of a second, slightly more adherent, underlayer of similar thickness was also present on the 360° edge. This fragment is displayed in Figure 3.53. No indication exists of a hydride phase immediately beneath the oxide layer.

3.2.3 Specimen 5-S2-J2

The longitudinal surface (0° face) of this specimen displayed no new features of significance. However, notably more fuel pull out and pitting (dark inclusions) in the as-polished condition was seen than on previous specimens, and the heat-tinted surface was much darker than the others. Relatively few hydride halos were observed around uranium carbide particles, and relatively few areas with large, distinct, hydride halos were seen. All of the hydride inclusions were small, measuring less than about 50 μm in diameter, and most displayed only faint halos. No oxide layer was visible at the fuel edges. Typical features on the surface of this specimen are shown in Figures 3.54 through 3.57 and are described below.

- The heat-tinted surface near the ID side and near the mid-section of the specimen is shown in Figures 3.54 and 3.55, respectively. Several small hydride inclusions, typically less than 35 μm in size, with thin halos are apparent in the matrix, along with the usual amount of uranium carbide particles. These features and their distribution are representative of those seen elsewhere on this specimen.
- One unusually bright hydride inclusion and several less distinct ones at the bottom edge of the specimen are shown in Figure 3.56. The structural detail of this bright inclusion is shown at higher magnification in Figure 3.57. Although it is small, the presence of such a distinct hydride at the edge of a conditioned specimen is surprising.

3.3 IPS-Conditioned Specimens 5-S2-F1A and 5-S2-G1

Specimens 5-S2-F1A (transverse) and 5-S2-G1 (transverse) are representative of the undamaged mid-length region of the fuel element after completing the IPS-recommended conditioning process. The conditioning temperature of the former specimen was 250°C, that of the latter was 200°C. These specimens were unique in that they exhibited a large amount of fuel pull out during polishing; more pull-out occurred on 5-S2-G1 than on 5-S2-F1A. Subjectively, it appeared the hydride content of each was

similar to that of the unconditioned specimens cut from the same mid-length region of the fuel element. However, the excessive pull out and friability of the surface of these IPS-conditioned specimens suggests that some of the hydrides near the surface might have decomposed during the conditioning process, leaving a friable uranium residue near the surface. Several large inclusions were present in the conditioned specimens (more so than in the unconditioned ones), but their characteristic halos were usually thin and not always distinct. Fragments of an oxide layer were observed on one fuel edge of 5-S2-F1A. The layer was flaking off and was less than 3 μm thick. No oxide layer was visible on the fuel edges of 5-S2-G1. The features of specimens 5-S2-F1A and 5-S2-G1 are described in Sections 3.3.1 and 3.3.2, respectively.

3.3.1 Specimen 5-S2-F1A

The fuel material at the top surface of this transverse specimen displayed a moderate amount of pull out or pitting, and an assortment of hydride shapes. Most of the hydride inclusions were small, less than about 35 μm in size. However, several hydrided areas and inclusions as large as 150 μm were also present. Subjectively, it appeared the hydride content was comparable to that seen in the unconditioned specimens 5-S2-A and 5-S2-C. A partial oxide layer, slightly less than 3 μm thick and flaking off, was observed on one fuel edge. Salient features of specimen 5-S2-F1A are shown in Figures 3.58 through 3.67 and are described as follows.

- Typical features in the mid-fuel region, after heat tinting, are shown in Figures 3.58, 3.59, and 3.60. These illustrations include hydride inclusions ranging in size from about 10 μm to 90 μm , along with several pits in the same size range but without halos. Some hydride inclusions are adjacent to carbide particles, others are not. No distribution pattern was evident. A higher magnification view of Figure 3.60 is shown in Figure 3.61, which displays very small hydride inclusions (dark), less than 5 μm in size, and located in what appears to be a small network of fine hydride "whiskers" (light).
- Several large hydride inclusions, along with several small ones, were present along the bottom edge of the specimen. These are shown in Figure 3.62. The inclusions encompassing the carbide particles are about 150 μm in size. The fine structure of these hydrides, observed before heat tinting, is shown in Figure 3.63.
- As with the other specimens, the fuel-to-cladding bond appeared intact along its full length on both the ID and OD sides. Several carbide and hydride inclusions were present in the fuel near the fuel-to-cladding interface. One large hydride inclusion near the interface is shown before and after heat-tinting in Figures 3.64 and 3.65, respectively. The fine structure in the hydrided area at the interface is shown at greater magnification in Figure 3.66. No effect on the overall integrity of the bond was evident.
- One of several flaking layers of oxide on the fuel edge (202° side) is shown in Figure 3.67. Similar fragments were present elsewhere along the same edge. The maximum oxide layer thickness measured was 3 μm . No oxide layer was visible on the fuel edge of the 180° side.

- Portions of an oxide layer were present on the waterside surfaces (ID and OD) of the cladding. The maximum thickness observed was about 1 μm .

3.3.2 Specimen 5-S2-G1

This specimen was the most friable of the specimens examined, in that it exhibited excessive pull out of fuel when using the same rough polishing method (1- μm diamond compound suspended in Hyprez lubricant) that was used on all specimens except 5-S1A-A1A. Pull out was reduced significantly after the surface was re-ground and then rough polished using 0.3- μm alumina in a 2% solution of chromic acid, as was used on specimen 5-S1A-A1A. Prior to re-grinding and polishing, many relatively large hydride inclusions and apparently hydrided pull out areas were visible, suggesting that these friable areas consisted of the residual fuel material from decomposed hydrides. A few of these regions are shown in Figures 3.68 through 3.79. After the final polish and heat tint, considerably fewer pits were visible on the fuel surface and most of the observed hydride inclusions were less than about 25 μm in diameter, as shown in Figures 3.80 and 3.81. These features are described in the following.

- Typical features located near the OD side, mid-fuel region, and ID side are shown after heat tinting in Figures 3.68, 3.69, and 3.70, respectively. A large number of small pits (dark spots) and a few large and small hydride inclusions with their characteristic halos are apparent in each figure. The largest hydride shown in the latter figure was about 200 μm wide.
- A typical hydride inclusion (~50 μm) near the 202° edge and a smaller one (~20 μm) at the 225° edge are shown after heat tinting in Figures 3.71 and 3.72, respectively.
- A large, apparently hydrided, pull out area on the ID side, near the fuel-to-cladding bond, is shown as-polished in Figures 3.73 and 3.74. A few small pits (dark specks) are also apparent nearby. The same area after heat tinting is shown in Figure 3.75. A diffuse halo is visible around the area, indicating that some uranium hydride is still present in the area.
- A significantly larger pull out area at the fuel-to-cladding bond on the ID side is shown as-polished in Figure 3.76 and heat tinted in Figure 3.77. The fine structure of a portion of this area is shown in Figures 3.78 and 3.79 before heat tinting and at a higher magnification.
- The typical appearance of the surface after re-grinding and polishing through 0.05- μm alumina is shown in Figure 3.80. Only a few dark inclusions, some as large as 50 μm but with very thin halos, are apparent in this figure. Hydrides with more distinct halos observed elsewhere on the specimen were typically less than about 25 μm in diameter. A typical, very small hydride (less than about 5 μm in diameter) is shown in Figure 3.81.
- No oxide layer was visible on the fuel edges.

3.4 Discussion

3.4.1 Inclusions and Pitting

Uranium carbide inclusions, uranium hydride inclusions, and dark areas of localized pitting and pull out of fuel particles were commonly observed in the uranium matrix of each specimen. The carbide inclusions (angular, equiaxed, white/grey) were visible in the as-polished and the heat tinted (partially oxidized) conditions of a specimen. The hydride inclusions were identified by a characteristic silver/yellow halo in the surrounding uranium matrix. With experience it was possible to identify hydride inclusions individually or in groups as small, dark pits in the as-polished specimen different from pits due to pull out or other pitting mechanisms. Pitting commonly resulted from the pull out of oxidized uranium fuel during polishing, and to a lesser extent after heat tinting, because of the friable nature of the oxide. Pitting was differentiated from hydride inclusions by the absence of a halo around the pitted area after heat tinting. Localized areas and former inclusions of decomposed uranium hydride might also appear as microscopic pits in the uranium surface. Pitting of this origin was expected to be present in the conditioned specimens. An alignment of pits in the direction of extrusion would signal the presence of carbide stringers formed during fuel fabrication; however, we did not observe this type of pitting in our specimens.

3.4.2 Uranium Hydride Inclusions

Most of the hydride inclusions appeared to be randomly distributed throughout the uranium matrix, some were located at only a few of the many carbide inclusions. They did not appear to be preferentially co-located with carbide inclusions. The size of the hydride inclusions and the extent of the hydrided areas varied among the specimens; the size and the extent of the hydrides were notably greater in the macro-crack areas of the specimen from the breached end of the element (Specimen 5-S1A-A1A). Hydride inclusions are expected to form at major discontinuities within the uranium microstructure, such as around carbide inclusions and carbide stringers, on grain boundaries, and on the surface of voids or micro-cracks (Gardner and Riches 1956; Waber 1958; Robinson and Thomas 1996). However, grain boundaries and micro-cracks were not clearly identified within the uranium metal on either the attack-polished or the electrolytically etched surfaces of our specimens. The microstructure in the as-etched condition had a "swirled" appearance, the grains were not distinct, and the size of the grains could not be measured. The appearance of the microstructure was very similar to that reported by Leggett et al. (1963, p.25) for as-extruded, high purity uranium specimens irradiated at temperatures in the range of 400 to 450°C.

3.4.3 Fuel-to-Cladding Bond Region

In the region of the fuel-to-cladding bond, irregularly shaped areas extended into the fuel from the bond interface at various lengths on most specimens. These areas were more distinct in the heat-tinted condition. They appeared to be either stains (polishing artifacts) or a region where submicron-sized fuel particles had been pulled from the matrix. In some of the heat-tinted specimens, less extensive areas with a similar appearance occasionally exhibited the halos characteristically displayed by uranium hydride inclusions. A thin, dark line immediately adjacent to the cladding and a wider, light-colored band (~4- μ m-wide) were also commonly observed in the bond area of all specimens. Both features were visibly

enhanced by the heat tint. They are believed to be artifacts of polishing, resulting from a difference in hardness between the fuel and its cladding. Carbide inclusions located adjacent to the bond region were also common and had no visible effect on the integrity of the bond. No separation of fuel from its cladding was observed, and no visible indication that the integrity of the fuel-to-cladding bond had been degraded. No indication was observed in any of the specimens that the fuel-cladding interface served as a preferential path for the migration of hydrogen or the ingress of corrosion to the uranium fuel core.

3.4.4 Uranium Oxide Layer

Short segments of a very thin uranium oxide layer were observed along the fuel edges of most specimens. The layer appeared to be porous and was obviously nonadherent. Typically, the layer appeared to be in the process of pulling away from the fuel and flaking off. In all but one case, the maximum thickness of the layer was 3 μm . The one exception was a very short segment that was about 10 μm thick, which was observed on the ITA-conditioned specimen 5-S2-B.

The fuel immediately under the oxide layer appeared to be porous in only one small segment. This segment was on the unconditioned specimen from the breached end of the element (Specimen 5-S1A-A1A). There was no positive indication that the dark, porous region contained a hydride phase. The porosity was attributed to degradation resulting from corrosion. The thickness of the oxide on this segment was less than 2 μm .

Waber (1958) reported that coherent layers of uranium oxide became unstable and began flaking or spalling from the uranium metal at oxide layer thicknesses of about 0.075 μm or greater. The layers were porous and were aligned parallel to the metal surface. When water vapor was the principal oxidant, Waber observed that a UH_3 phase with finger-like projections formed along the oxidation front in the metal, immediately below the oxide layer. Wilkinson (1962, p.764) noted that oxidation of uranium metal in air or high humidity accelerated markedly at temperatures above 75°C to form a nonadherent oxide. We observed the thin, porous, nonadherent, oxide layer similar to, but somewhat thicker than, that observed by Waber. We did not detect a hydride phase that was clearly associated with the oxide layer because the existing oxide layer had apparently formed in a dry atmosphere during specimen preparation or conditioning.

3.4.5 Comparison of Unconditioned vs. Conditioned Specimens

Only a few subtle differences were apparent between the two undamaged and unconditioned specimens, the IPS-conditioned specimens, and the ITA-conditioned specimens. The fuel surface of the IPS-conditioned specimens was more friable than the others, as evidenced by the large amount of fuel pull out or pitting that was observed during polishing. Some of the pull out areas appeared to be large hydride inclusions or hydrided areas with diffuse halos. This, in conjunction with the excessive pull out and friability of the surface, indicated that some of the hydrides near the surface decomposed during the IPS-recommended conditioning process, leaving a friable uranium residue near the surface. The ITA-conditioned specimens typically exhibited smaller hydride inclusions (less than about 50 μm in diameter) than did the IPS-conditioned specimens. Many of their hydrides had diffuse halos, indicating that some hydride decomposition also occurred during the ITA-recommended conditioning process. Although

indications were that some hydride decomposition was achieved during both conditioning processes, obvious uranium hydride inclusions remained in each of the conditioned specimens. Thus, neither conditioning process completely decomposed the uranium hydride phase.

3.4.6 Damaged Fuel Region, Crack Propagation into the Cladding, and Hydrogen Sources

Macro-cracks were observed only on the unconditioned specimen cut from the corroded, breached end of the fuel element (Specimen 5-S1A-A1A). An extensive crack network was present in the fuel matrix of this specimen. Several of the cracks penetrated a short distance into the cladding. The maximum observed depth of penetration was about 180 μm . Within the fuel matrix, the last vestige of a crack appeared to end near the bottom of this specimen, about 1.2 cm (0.47 in.) from the corroded surface. After being heat tinted, the fuel along the sides of many cracks displayed a bright glowing band with a diffuse edge that was similar in appearance to the distinct halos that highlighted uranium hydride inclusions. This glow indicated the fuel adjacent to the cracks was hydrided and the cracks acted as conduits by carrying hydrogen from the exposed corroded surface region deeper into the fuel matrix. Nothing indicated the cracks had initiated in the cladding. Instead, they appeared to originate in the fuel region and propagate into the cladding. Aside from the macro-cracks and the more extensive hydriding, no significant differences between this specimen and the others were observed. Thus, the combination of corrosion-induced fuel swelling and stress-induced crack propagation into the cladding apparently resulted in the cladding becoming distorted and subsequently rupturing to relieve the stress.

Troutner (1960) studied the corrosion of unirradiated uranium fuel elements and their rupture in water and steam in the temperature range of 200 to 300°C. The carbon composition of the ingot material ranged from 30 to 1625 ppm. The sample elements were fabricated by co-extruding beta heat treated ingots of natural uranium with Zircaloy-2 cladding material. The corrosion tests were performed on open-ended samples. The rupture tests were performed on small elements that had pinholes (0.635-mm [0.03-in.] diameter) pre-drilled through their cladding to permit water to enter the uranium core of the element.

In the corrosion tests, Troutner observed that nonuniform corrosion occurred in the open-ended samples. The corrosion pits contained UO_2 , UH_3 , and particles of uranium metal. A zone of UH_3 had formed on the uranium surface. Zirconium hydride (ZrH_2) had formed on the inner surface of the Zircaloy-2 cladding, extending to just ahead of the reaction site. These features indicated that corrosion proceeded behind a hydride front when water was restricted from the reaction site. The nonuniform corrosion occurred with penetration in the direction of extrusion, possibly along carbide stringers, and with the formation of uranium hydride within the metal. Troutner concluded the nonuniform attack was caused by penetration of hydrogen into the metal along stringers of inclusions, probably carbides. Expansion due to hydride formation disrupted the metal, which eventually permitted the entrance of water, followed by conversion of the hydride to oxide.

In the rupture tests, Troutner observed the cladding had hydrided some distance from the rupture site, resulting in brittle failure of the cladding before it had swelled. The uranium exhibited extensive swelling and had a spongy appearance, which clearly resulted from internal hydride penetration of the uranium as in the corrosion tests. Troutner concluded that water entered the defect and reacted with the uranium to form oxide and hydrogen. When the buildup of oxide blocked the defect, the hydrogen reacted rapidly with the

uranium to form UH_3 . The volume change resulting from the formation of uranium oxide and hydride caused the cladding to swell. Because the hydride penetrated into the uranium, the swelling of the cladding spread over a wide area, rather than being constrained to a small blister. Carbide stringers in the uranium probably provided an easy path for the diffusion of hydrogen and the formation of uranium hydride. Thus, hydriding proceeded faster along the length of the element than radially through the core. Swelling and tearing of the cladding also proceeded lengthwise. The extensive hydriding observed in the cladding of the ruptured elements was attributed to hydriding from within the element. The increase in rupture rate of irradiated samples was attributed to a more rapid attack down the fuel-to-cladding bond region, causing separation of the cladding or blistering.

In contrast, we did not observe a distinct zone of UH_3 on the corroded uranium (fuel) surface, nor did we see any indication that zirconium hydride (ZrH_2) had preferentially formed on the inner surface of the Zircaloy-2 cladding. We did not observe a network of carbide stringers in the fuel sufficient to provide an easy path for the diffusion of hydrogen. Furthermore, we found no indication that the bond region served as a preferential path for the migration of hydrogen or for the initiation and progression of hydriding. These results do not refute the extensive work of Troutner. Within a short time after the breached element was discharged into the K Basin, it is quite likely that corrosion proceeded in the manner that he described. As the residence time of the element in the basin increased, however, the incipient features of the corrosion process apparently dissipated. The hydride front and the area of initial corrosion eventually oxidized, and the residue fell away into the storage canister.

Sources of hydrogen for the formation of uranium hydride in the fuel matrix of a breached fuel element could include 1) tramp hydrogen resulting from the fuel fabrication process, 2) hydrogen generated from oxidation of the Zircaloy-2 cladding and its subsequent transport into the fuel while operating in the reactor, and 3) hydrogen produced as the fuel corroded during storage in the K Basin. The hydrogen content of the fuel and cladding was tightly controlled during material procurement and fuel element fabrication to avoid cladding embrittlement later in life, which could result if the concentration of zirconium hydride in the cladding became excessive. The hydrogen content of the procured fuel and cladding material was limited to 2 ppm and 24 ppm, respectively. The solubility limit of hydrogen in Zircaloy is about 200 ppm at cladding normal operating temperature. Only a fraction of the hydrogen (about 10% to 20%) generated from waterside oxidation of Zircaloy-2 cladding is picked up by the cladding. Based on thermodynamic principles, the hydrogen that is picked up is expected to remain within the cladding, either in solution or as a zirconium hydride (ZrH_2) precipitate, rather than diffuse into the uranium. The contribution of hydrogen to the fuel matrix from fabrication and from waterside corrosion of intact cladding is not considered significant. Thus, most of the hydrogen in the uranium fuel probably resulted from aqueous corrosion of the exposed uranium after the breached element was discharged from the reactor.

SNF-130

— 90 μ m

Figure 3.1. Typical Surface Features Before Heat Tint; Mid-Fuel Region; Specimen 5-S2-A

SNF-138

|—| 90 μ m

Figure 3.2. Typical Surface Features After Heat Tint; Mid-Fuel Region; Specimen 5-S2-A. Same area as Figure 3.1.

SNF-127

|—| 18 μ m

Figure 3.3. Small Uranium Hydride Inclusions (<10 μ m) with Halo and Uranium Carbide Inclusions;
After Heat Tint; Specimen 5-S2-A

SNF-128

— 6 μm

Figure 3.4. Magnified View of Figure 3.3 Showing Hydride Structure; After Heat Tint; Specimen 5-S2-A

SNF-416

—| 115 μm

Figure 3.5. Typical Surface Appearance After Electrolytic Etch for Grain Enhancement; Mid-Fuel Region; Specimen 5-S2-A

SNF-455

—| 23 μ m

Figure 3.6. Typical Surface Appearance After Electrolytic Etch for Grain Enhancement; Mid-Fuel Region; Specimen 5-S2-A

SNF-456

—| 12 μm

Figure 3.7. Typical Surface Appearance at Higher Magnification After Electrolytic Etch for Grain Enhancement; Mid-Fuel Region; Specimen 5-S2-A). Same area as Figure 3.6.

SNF-144

85 μm

Figure 3.8. Typical Surface Features Before Heat Tint; Mid-Fuel Region; Specimen 5-S2-C

SNF-178

—| 23 μ m

Figure 3.9. Uranium Hydride Inclusion, Uranium Carbide Inclusion, and Fuel-to-Cladding Bond Region;
Before Heat Tint; Specimen 5-S2-C

SNF-182

—| 21 μ m

Figure 3.10. Hydride Inclusion, Carbide Inclusion, and Fuel-to-Cladding Bond Region; Before Heat Tint; Specimen 5-S2-C. Same area as Figure 3.9.

SNF-184

— 11 μ m

Figure 3.11. Hydride Inclusion Collocated at Carbide Inclusion; After Heat Tint; Specimen 5-S2-C.
Same area as Figure 3.10.

Figure 3.12. Fuel-to-Cladding Bond Area with Typical Carbide Inclusions, Possible Hydride Inclusions, and Local Oxidation; Before Heat Tint; Specimen 5-S2-C

SNF-152

18 μ m

Figure 3.13. Fuel-to-Cladding Bond Area, OD Side, with Typical Hydride Inclusions, Carbide Inclusions, Minor Fuel Pull Out, and Pitting; After Heat Tint; Specimen 5-S2-C

SNF-153

— 9 μ m

Figure 3.14. Fuel-to-Cladding Bond Area, OD Side, Mid-Length with Typical Hydride Inclusions, Carbide Inclusion, Minor Fuel Pull Out, and Pitting; After Heat Tint; Specimen 5-S2-C

SNF-419

—| 115 μ m

Figure 3.15. Typical Surface Appearance After Electrolytic Etch for Grain Enhancement; Mid-Fuel Region; Specimen 5-S2-C

Figure 3.16. Small Fragment of Uranium Oxide Layer ($\geq 2 \mu\text{m}$ Thick); Specimen 5-S2-C

Figure 3.17. Corroded Surface, Breached End, OD Side Showing Fuel Cracks, Carbide Inclusions, and Hydride Inclusions; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.18. Continuation of Figure 3.17 Showing Portion of Crack Network Terminating at Cladding, Hydrided Areas Along Cracks, Hydride Inclusions, and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.19. Continuation of Figure 3.18 Showing Portion of Crack Network Terminating at Cladding, Hydrided Areas Along Cracks, Hydride Inclusions, and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

SNF-645

—| 315 μ m

Figure 3.20. OD Side, Below Mid-length, Showing Portion of Crack Network Terminating at Cladding, Hydrided Areas Along Cracks, Hydride Inclusions, and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

SNF-646

—| 333 μ m

Figure 3.21. Continuation of Figure 3.20 to Bottom Edge, OD Side Showing Portion of Crack Network Terminating at Cladding, Hydrided Areas Along Cracks, Hydride Inclusions, and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

SNF-639

—| 330 μ m

Figure 3.22. Corroded Surface, Breached End, ID Side Showing Fuel Cracks Terminating in Cladding, Carbide Inclusions, and Hydride Inclusions; After Heat Tint; Specimen 5-S1A-A1A

SNF-639

—| 330 μ m

Figure 3.23. ID Side, Mid-Length, Showing Continuation of Crack Network Terminating at Cladding, Hydrided Areas Along Cracks, Hydride Inclusions, and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.24. ID Side, Bottom End, Showing Large Hydride Inclusion and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.25. Mid-Fuel Region Between Figures 3.19 and 3.23 Showing Continuation of Crack Network, Hydrided Areas Along Cracks, Hydride Inclusions, and Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

SNF-663

—| 40 μm

Figure 3.26. Magnified View of Figure 3.23 Showing Large Hydride Inclusion Typical of Those in the Crack Region; After Heat Tint; Specimen 5-S1A-A1A

SNF-669

—| 22 μm

Figure 3.27. Magnified View of Figure 3.25 Showing Fine Structure of Large Hydride Inclusion Typical of Those in the Crack Region; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.28. Magnified View of Figure 3.22 Showing Large Hydride Inclusion in the Crack Region at Breached End; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.29. Magnified View of Figure 3.24 Large Hydride Inclusion Encompassing Carbide Inclusions; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.30. Small Hydride Inclusion and Carbide Inclusions Along Bottom Edge; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.31. Magnified View of Figure 3.20 Showing Tip of Macro-Crack Penetrating into the Cladding; After Heat Tint; Specimen 5-S1A-A1A

SNF-636

—| 15 μ m

Figure 3.32. Magnified View of Figure 3.21 Showing Tip of Macro-Crack Penetrating into the Cladding; After Heat Tint; Specimen 5-S1A-A1A

SNF-638

—| 45 μ m

Figure 3.33. Magnified View of Figure 3.21 Showing Tip of Macro-Crack Penetrating into the Cladding; After Heat Tint; Specimen 5-S1A-A1A

Figure 3.34. Magnified View of Specimen 5-S1A-A1A Showing Fragment of Thin Oxide Layer Above Porous Fuel Substructure; Before Heat Tint. Figure 3.17 shows a lower magnification of this area following the heat tint.

SNF-319

—| 85 μ m

Figure 3.35. Typical Surface Appearance After Heat Tint, OD Side; Specimen 5-S2-B

SNF-322

—| 85 μm

Figure 3.36. Typical Surface Appearance After Heat Tint, Mid-Fuel Region; Specimen 5-S2-B

SNF-314

— 8 μ m

Figure 3.37. Small Hydride Inclusions ($< 10 \mu\text{m}$) Collocated at Carbide Inclusion and in Fuel Matrix; After Heat Tint; Specimen 5-S2-B

SNF-316

8 μm

Figure 3.38. Small Hydride Inclusions ($< 10 \mu\text{m}$) and Carbide Inclusion in Fuel Matrix; After Heat Tint; Specimen 5-S2-B

Figure 3.39. Adherent Portion of Thin, Porous Uranium Oxide Layer ($\leq 2 \mu\text{m}$ Thick) at Top Edge; Before Heat Tint; Specimen 5-S2-B

Figure 3.40. Adherent Portion of Thin, Porous Uranium Oxide Layer ($\leq 3 \mu\text{m}$ Thick) and Carbide Inclusions at Top Edge; Before Heat Tint; Specimen 5-S2-B

Figure 3.41. Nonadherent Portion of Thin, Porous Uranium Oxide Layer ($\leq 3 \mu\text{m}$ Thick) and Carbide Inclusions at Bottom Edge; After Heat Tint; Specimen 5-S2-B

Figure 3.42. Magnified View of Figure 3.41 Showing Portion of Nonadherent, Thin, Porous Oxide Layer; After Heat Tint; Specimen 5-S2-B

Figure 3.43. Nonadherent Portion of Thin, Porous Uranium Oxide Layer ($\leq 2 \mu\text{m}$ Thick) Flaking from the Bottom Edge; After Heat Tint; Specimen 5-S2-B

SNF-325

—| 22 μm

Figure 3.44. Abnormally Thick Fragment of Uranium Oxide ($\leq 10 \mu\text{m}$ Thick) at the Bottom Edge; After Heat Tint; Specimen 5-S2-B

Figure 3.45. Typical Surface Appearance After Heat Tint, ID Side; Specimen 5-S2-J1

SNF-265

|—| 85 μm

Figure 3.46. Typical Surface Appearance After Heat Tint, Mid-Fuel Region; Specimen 5-S2-J1

SNF-274

—| 21 μm

Figure 3.47. Magnified View of Figure 3.46 Showing Small Bright Hydride Inclusion and Carbide Inclusions; After Heat Tint; Specimen 5-S2-J1

SNF-261

—| 22 μm

Figure 3.48. Small Hydride Inclusions in Fuel Matrix and Collocated at a Carbide Inclusion; After Heat Tint; Specimen 5-S2-J1

Figure 3.49. Fuel-to-Cladding Bond on ID Side, Hydride Inclusion Collocated with Carbide Inclusion; After Heat Tint; Specimen 5-S2-J1

SNF-263

17 μm

Figure 3.50. Area of Figure 3.49 Shown with Polarized Light Illumination; After Heat Tint; Specimen 5-S2-J1

SNF-280

—| 17 μ m

Figure 3.51. Fuel-to-Cladding Bond on ID Side, Hydride Inclusions Collocated with Carbide Inclusions, and Minor Pitting; After Heat Tint; Specimen 5-S2-J1

SNF-281

7 μ m

Figure 3.52. Magnified View of Figure 3.51 Showing Collocated Carbide Inclusions, Hydride Inclusions on the Fuel-to-Cladding Bond; After Heat Tint; Specimen 5-S2-J1

Figure 3.53. Small Fragment of Uranium Oxide Layer Flaking from a More Adherent Layer; Before Heat Tint; Specimen 5-S2-J1

SNF-287

85 μm

Figure 3.54. Typical Surface Appearance After Heat Tint, ID Side, Mid-Length; Specimen 5-S2-J2

SNF-290

85 μ m

Figure 3.55. Typical Surface Appearance After Heat Tint, Mid-Fuel Region; Specimen 5-S2-J2

Figure 3.56. Unusually Bright Hydride Inclusion at Bottom Edge; After Heat Tint; Specimen 5-S2-J2

Figure 3.57. Magnified View of Figure 3.56 Showing Fine Structure of the Small Bright Hydride Inclusion; After Heat Tint; Specimen 5-S2-J2

Figure 3.58. Typical Surface Appearance with Carbide Inclusions, Residual Hydrides, and Pitted Areas; Mid-Fuel Region; After Heat Tint; Specimen 5-S2-F1A

Figure 3.59. Mid-Fuel Region Below Figure 3.58 After Heat Tint; Specimen 5-S2-F1A

SNF-510

135 μm

Figure 3.60. Mid-Fuel Region Below Figure 3.59 After Heat Tint; Specimen 5-S2-F1A

SNF-517

40 μm

Figure 3.61. Magnified View of Figure 3.60 (Left of Center) Showing Network of Small Hydride Inclusions and Whiskers; After Heat Tint; Specimen 5-S2-F1A

Figure 3.63. Magnified View of Figure 3.62 Showing Large Hydride Inclusions Encompassing Carbide Inclusions; Before Heat Tint; Specimen 5-S2-F1A

Figure 3.62. Assortment of Hydride Inclusions, Carbide Inclusions, and Pitting Along the Bottom Edge; After Heat Tint; Specimen 5-S2-F1A

SNF-502

40 μ m

Figure 3.64. Fuel-to-Cladding Bond on OD Side, Hydride Inclusions and Pitting; Before Heat Tint; Specimen 5-S2-F1A

SNF-504

135 μ m

Figure 3.65. Same Area as Figure 3.64 Showing Residual Hydride Inclusions, Carbide Inclusions, and Pitting Near the Bond Area; After Heat Tint; Specimen 5-S2-F1A

SNF-516

—| 20 μ m

Figure 3.66. Magnified View of Figure 3.65 Showing Hydride and Carbide Inclusions in the Bond Area; After Heat Tint; Specimen 5-S2-F1A

SNF-494

—| 16 μ m

Figure 3.67. Fragment of Porous, Nonadherent Uranium Oxide Layer ($\leq 3 \mu$ m Thick) Along the 202° Edge; Before Heat Tint; Specimen 5-S2-F1A

SNF-567

—| 135 μ m

Figure 3.68. Typical Surface Appearance; OD Side Near Fuel-to-Cladding Bond; After Heat Tint; Specimen 5-S2-G1

SNF-569

—| 135 μ m

Figure 3.69. Typical Surface Appearance, Mid-Fuel Region; After Heat Tint; Specimen 5-S2-G1

SNF-573

135 μm

Figure 3.70. Typical Surface Appearance, Near ID Side; After Heat Tint; Specimen 5-S2-G1

SNF-565

40 μm

Figure 3.71. Partially Decomposed Hydride Inclusion, Carbide Inclusions, and Small Pits, Near 202° Edge; After Heat Tint; Specimen 5-S2-G1

SNF-564

40 μm

Figure 3.72. Small Hydride Inclusion at 225° Edge; After Heat Tint; Specimen 5-S2-G1

SNF-552

40 μm

Figure 3.73. Partially Decomposed Hydride Inclusion, Carbide Inclusions, and Pitting at Fuel-to-Cladding Bond on ID Side; Before Heat Tint; Specimen 5-S2-G1

SNF-551

13 μ m

Figure 3.74. Magnified View of Area in Figure 3.73 Showing Friable Surface of Hydride Inclusion; Before Heat Tint; Specimen 5-S2-G1

SNF-563

40 μ m

Figure 3.75. Partially Decomposed Hydride Inclusion, Carbide Inclusions, and Pitting at Fuel-to-Cladding Bond on ID Side; After Heat Tint; Specimen 5-S2-G1. Same area as Figure 3.73.

SNF-561

40 μm

Figure 3.76. Large Partially Decomposed Hydride and Pitting at Fuel-to-Cladding Bond on ID Side; Before Heat Tint; Specimen 5-S2-G1

SNF-562

40 μm

Figure 3.77. Large Partially Decomposed Hydride and Pitting at Fuel-to-Cladding Bond on ID Side; After Heat Tint; Specimen 5-S2-G1. Same area as Figure 3.76.

SNF-557

13 μm

Figure 3.78. Magnified View of Figure 3.76 Before Heat Tint; Specimen 5-S2-G1

SNF-558

13 μm

Figure 3.79. Continuation of Figure 3.78 Before Heat Tint; Specimen 5-S2-G1

SNF-592

140 μm

Figure 3.80. Fuel-to-Cladding Bond on ID Side After Repolishing (0.05-μm alumina); After Heat Tint; Specimen 5-S2-G1

SNF-594

14 μm

Figure 3.81. Very Small Hydride Inclusions After Repolishing (0.05-μm alumina); After Heat Tint; Specimen 5-S2-G1

4.0 Recommendations for Further Metallography

1. Polish and etch the Zircaloy-2 cladding of a specimen from the breached end of fuel element SFEC5,4378 to display the distribution of zirconium hydrides within the cladding, particularly near the crack tips. This process will provide additional qualitative information to assess whether or not the cladding acts as a sink for the hydrogen generated in the corrosion process. For comparison, similarly prepare and examine at least one specimen from the undamaged mid-length region of this element to characterize the typical distribution of zirconium hydrides.
2. Examine three specimens from the breached area of SFEC5,4378; these being one as-cut, one ITA-conditioned, and one IPS-conditioned specimen. This examination will provide information concerning the degree to which the processes remove uranium hydrides from the fuel, and the nature of the uranium oxide layer formed on the exposed uranium surfaces.
3. Examine a specimen from the damaged end of SFEC10,4366 to compare its uranium and zirconium hydride morphologies with those in the specimens from SFEC5,4378.

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Appendix A

Specimen Preparation for Detection of Uranium Hydride and Measurement of Oxide Layer Thickness

Appendix A

Specimen Preparation for Detection of Uranium Hydride and Measurement of Oxide Layer Thickness

The specimens were polished and heat-tinted, as described in the following paragraphs. The methods used are similar to those of Gardner and Riches (1956). The operations were performed remotely in a hot cell.

The specimen was mounted in a stainless steel ring [typically 3.2 cm (1.3 in.) diameter by 1.6 cm (0.63 in.) high]. If measuring the oxide layer, stainless steel coupons were placed within the ring, immediately adjacent to the edges of the specimen, to assist in edge retention during the subsequent grinding and polishing. The ring was filled with Buehler Epo-Thin epoxy mounting material or Titan Fiberglass Finishing Resin, and then allowed to cure. Titan resin was used to prepare the first two specimens (5-S2-A and 5-S2-C). Buehler Epo-Thin epoxy was used for the other specimens. This epoxy was preferred because of its low viscosity, low shrinkage, and minimal softening during the subsequent heat-tinting process.

The mounted specimen was ground progressively on 180, 240, 400, and 600 grit silicon carbide abrasive papers, vacuum impregnated with Buehler Epo-Thin epoxy, and allowed to cure overnight. Grinding was then continued on 600, 800, and 1200 grit silicon carbide abrasive papers. All of the grinding was done in the hot cell on a rotary grinder/polisher using Hyprez O.S. Type IV Lubricant. The specimen was cleaned with isopropyl alcohol and de-ionized water.

Rough polishing was performed using one of the following methods:

1. Polished with Hyprez Diamond Compound Grade 1 OS MA 47 (1- μ m diamond compound) suspended in Hyprez O.S. Type IV Lubricant on either a Glennel Gold Label, Buehler Texmet, or LECO Pan K polishing cloth. Either a rotary or a vibratory polisher was used; the rotary was preferred. The specimen was cleaned using isopropyl alcohol and de-ionized water. Rotary polishing times were approximately 35 minutes per cycle. One cycle was needed to adequately polish the fuel region, and either three or four cycles were needed to polish the cladding. This method was used to satisfactorily polish the cladding surface, but it often caused pull-out of material (pitting) from the uranium fuel surface.
2. *Attack* polished using a saturated solution of 0.3- μ m alumina (Al_2O_3 ; Buehler Micropolish II) in 2% chromic acid [2 g CrO_3 in 100 mL (3.4 fl oz) of de-ionized water] on Buehler Texmet polishing cloth. A vibratory polisher was used. The polishing typically was performed in 2 cycles of about 35 minutes each for a total rough polishing time of about 70 minutes. The specimen was cleaned with de-ionized water. This method resulted in less pull-out of the fuel material and a good polish of the fuel region

when followed by a final attack polish with 0.05- μm alumina. However, the polishing times were too short to obtain a good polish of the cladding surface. (This method was used on specimens 5-S1A-A1A and 5-S2-G1 to minimize pull-out.)

The final polish was performed on a vibratory polisher with a Wendt Dunnington, Final B polishing cloth. An attack polish was used, which consisted of a saturated solution of either 0.3- μm alumina or 0.05- μm alumina (Al_2O_3 ; Buehler Micropolish II) in 2% chromic acid. The final polishing times were typically in a range from 2 to 5 minutes.

The specimen was heat-tinted (partially oxidized) by exposing its polished surface to a 500 W halogen lamp for 15 to 45 minutes. The distance between the lamp and the surface of the specimen was kept at about 6 to 7 cm (2.4 to 2.8 in.) to avoid softening the Epo-Thin mounting material. When the specimen was successfully heat-tinted, discrete uranium hydride inclusions (dark) were surrounded by a silver/yellow glow or halo resulting from the lower oxidation rate of the uranium immediately adjacent to the hydride inclusion.

Appendix B

Specimen Preparation for Examination of Uranium Grain Structure and Fine Uranium Hydride Networks

Appendix B

Specimen Preparation for Examination of Uranium Grain Structure and Fine Uranium Hydride Networks

Selected specimens were prepared for examination of their grain structure in order to facilitate a more detailed search for fine networks of uranium hydride. These fine networks of very small hydrides are expected to be preferentially located in the grain boundary and triple-point regions of the fuel micro-structure. Typically, the specimens were electrolytically polished and etched for follow-on examination after they were heat-tinted and examined for the presence of larger hydrides. A minimal amount of grinding and rough polishing was necessary to expose a fresh surface of the specimen. This method was used on specimens 5-S2-A and 5-S2-C.

Each of the following preparatory steps gave an equivalent surface appearance of the fuel after electro-polishing and electro-etching.

1. Ground the specimen through 800-grit silicon carbide paper on a rotary grinder/polisher with Hyprez O.S. Type IV Lubricant. Rough polished for about 30 minutes on a rotary polisher using Hyprez Diamond Compound Grade 1 OS MA 47 (1- μ m diamond compound) suspended in Hyprez O.S. Type IV Lubricant on Glennel Gold Label polishing cloth.
2. Ground the specimen through 1200-grit silicon carbide paper on a rotary grinder/polisher with Hyprez O.S. Type IV Lubricant. Attack polished on a vibratory polisher using a saturated solution of 0.3- μ m alumina (Al_2O_3 ; Buehler Micropolish II) in 2% chromic acid [2 g CrO_3 in 100 mL (3.4 fl oz) of de-ionized water] on Buehler Texmet polishing cloth.

The electrolytic polishing and etching methods used are described in the following paragraphs. The methods are similar to those of Gardner and Riches (1956). The chemical solutions are given in Table 1 (Solution 2) and Table 5 (Solution 7), pages 478 and 480 respectively, of the ASM Handbook (1985).

- Electro-polished the specimen for 30 to 35 seconds at 18 to 20 V(dc) in a solution of 50 mL (1.7 fl oz) ethylene glycol, 50 mL (1.7 fl oz) ortho-phosphoric acid (sp.gr. = 1.70 g/cm³), and 80 mL (2.7 fl oz) ethyl alcohol using a platinum cathode.
- Electro-etched the specimen for 30 to 35 seconds at 1.5 to 2.0 V(dc) in a solution of 10 g (0.32 oz) citric acid, 215 mL (7.3 fl oz) nitric acid (sp.gr. = 1.42 g/cm³), and 490 mL (16.6 fl oz) H_2O using either a platinum or a stainless steel cathode.

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