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ELECTRON MICROSCOPY ANALYSIS
OF IRRADIATED SNAP 8 FUEL
(Title Unclassified)

AEC Research and Development Report

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ELECTRON MICROSCOPY ANALYSIS
OF IRRADIATED SNAP 8 FUEL
(Title Unclassified)

By

K. J. MILLER

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ABSTRACT

The irradiated fuel for this study was obtained from the SNAP 8 Experimental Reactor and the NAA 115-1 Capsule Irradiation Experiment. The fuel burnup achieved in these experiments was approximately 0.2 and 0.4 met. at. %, respectively.

During the postirradiation examinations, physical measurements and metallography were performed on the fuel to determine volume changes and fuel microstructure. Metallography revealed the fuel surface features up to 500X magnification; however, it was felt that further characterization of the fuel structure beyond that possible with optical metallography could be made through the use of an electron microscopic examination at selected fuel rod locations. Fuel samples were chosen both from regions of large volume change and excessive hydrogen loss.

Examination of the replica photographs revealed alpha uranium sites, carbide particle sites, phase boundaries, epsilon banding, and fission gas bubbles. Fuel microcracking was observed in some samples but was not excessive. Bubble density measurements on the order of 10^{13} bubbles/cc were typical, with an average bubble diameter of 900 Å.

Fission gas bubble formation accounted for approximately 50% of the observed hydrogen-corrected volume change in the peak distortion region from samples analyzed from two S8ER fuel rods. Analysis of the S8ER data indicated a possible dependence of fission gas bubble size and density on temperature. However, there did not appear to be a correlation with fuel burnup.

Based on the fuel samples examined, there was no evidence of fission gas bubble sweeping during phase change. No correlation was found between the location of fission gas bubble sites and epsilon banding or microcracks.

Bubble depletion regions observed around large particle sites are thought to be a result of the interaction of the gas bubbles with the larger uranium and carbide particles.

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I. INTRODUCTION

The irradiated fuel for this study was obtained from the SNAP 8 Experimental Reactor (S8ER) and the NAA 115-1 Capsule Irradiation Experiment.

The S8ER, first in a series of SNAP 8 nuclear tests, was operated to determine reactor performance characteristics and to demonstrate sustained operation at design power. SNAP 8 is a NaK-cooled, highly enriched uranium-zirconium hydride fueled, compact nuclear power system designed to operate at 600 kwt with a 1300°F reactor coolant outlet temperature. Nuclear testing of the reactor was conducted over the period from November 22, 1963 to April 15, 1965. The reactor experienced a total of 12,000 hr of operation with 5×10^6 kwh energy release, of which 8,760 hr (1 year) were between 400 and 600 kwt with coolant temperatures at 1300°F. The peak burnup achieved by the fuel was approximately 0.25 met. at. %.

The S8ER reactor core was shipped as a unit to the AI Hot Laboratory in the summer of 1965 for a postirradiation examination. Detailed analysis of the fuel from selected elements revealed localized swelling which was primarily the result of hydride fuel phase transformation.⁽¹⁾

The NAA 115-1 irradiation experiment consisted of twelve sub-length fuel elements which were irradiated simultaneously in a single facility of the Hanford K - East Reactor to design conditions of 0.4 met. at. % burnup in 10,000 hr. The fuel elements contained U-Zr hydride, and were typical of the S8ER design except for their lengths, internal clearances, and the range of their fuel compositions. Design fuel compositions included 1.60, 1.70, and 1.85 H/Zr ratios, and 0.02, 0.1, and 0.4 wt % carbon grain refiner additions. The elements were contained in six separate experimental units, which provided a means of controlling the maximum fuel temperatures at different levels ranging from 1300 to 1600°F. This was done to establish the effect of temperature and major compositional variables on the dimensional stability of the fuel.

Following discharge from the reactor, the experiment was shipped to the AI Hot Laboratory (AIHL) for a postirradiation examination, where 11 of the 12 fuel elements were revealed to be in excellent condition. For the temperatures, grain refiner additions, and hydrogen concentrations covered, fuel swelling was in the range of 2 to 4 vol %.⁽²⁾

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During both the S8ER and NAA 115-1 postirradiation examinations, fuel metallography samples were obtained from the fuel rods at selected locations which were representative of the various observed fuel swelling characteristics. Metallographic examination of the fuel revealed the microstructure and surface features, such as cracks and NaK reaction effects, at magnifications up to 500X. Further characterization of the fuel structure was made through the use of an electron microscopic examination at selected fuel rod locations.

Irradiation of fuel materials typically produces swelling, with a major cause being the formation of fission gas bubbles. The distribution of these bubbles in the lattice of the fuel material can have profound effects on the swelling behavior of the fuel. Precipitates, grain and phase boundaries, cracks and imperfections affect the distribution of fission gas bubbles and the macroscopic behavior of the fuel. By performing an electron microscopic examination of irradiated SNAP 8 fuel, it was possible to detect the microstructural details not observed by optical microscopy and, in particular, to determine fission gas bubble density.

A total of nine S8ER fuel samples, three NAA 115-1 fuel samples, and three unirradiated samples had their surfaces replicated for electron microscope examination. The samples were chosen from the following areas of fuel behavior:

- 1) Regions of suspected delta-to-delta plus beta transformation as evidenced by large volume changes and low final hydrogen contents.
- 2) Regions of suspected delta-to-epsilon transformation as evidenced by small volume changes and hydrogen contents equal to or greater than the preirradiation values.
- 3) Unusual phenomena, such as very large volume changes and unexpected postirradiation hydrogen contents.

Each sample, and its reason for selection, is listed in Table 1.

Prior to this time, replication at AIHL had not been demonstrated as routine. Therefore, technique development performed remotely on irradiated samples was necessary before successful replicas could be obtained. The procedures developed at the Hot Lab are discussed briefly in the following section of this report.

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TABLE 1
SAMPLE SELECTION FOR ELECTRON MICROSCOPIC ANALYSIS

Experiment	Element No.	Core Location	Axial Location on Rod (Distance from top, in.)	Reason for Selection As Listed in Text
S8ER	432	VIII-29	3	1
S8ER	332	VIII-25	1-1/2	3
S8ER	188	VIII-6	5-1/2	3
S8ER	378	IX-28	3	2
S8ER	378	IX-28	11-1/2	2
S8ER	111	IX-42	11-1/2	2
S8ER	232	VI-24	5-3/4	2
S8ER	519	IV-1	6-1/2	2
S8ER	398	IV-13	11	2
115-1	IS-4	-	2	2
115-1	IS-7	-	2	1
115-1	IS-8	-	2	3

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II. REPLICATION TECHNIQUES

Radioactive materials emitting high levels of beta and gamma activity present a major handling problem and cannot be directly examined in the electron microscope. Examination of thin foils of a radioactive material is possible only if the radioactivity rapidly decays to a low level. However, replication provides a means of transferring the surface characteristics of a highly radioactive sample to a low-level sample that can be examined in a nonshielded area.

Plastic, carbon, and oxide replicas are commonly used to prepare surfaces for examination with the electron microscope.⁽³⁾ Plastic replicas were selected due to apparent advantages in ease of remote operations. Use of plastic replication techniques involves careful metallographic processing and cleaning of the specimen surface. The replica, which is a negative of the specimen surface, can be stripped from the sample with a suitable backing material. The contrast on such replicas can be improved by shadowing the plastic with heavy metals (gold, platinum, chromium) or carbon,⁽⁴⁾ producing ultimate resolution of 200 Å.

Replication techniques were first attempted with unirradiated fuel material out-of-cell to conserve expensive in-cell development time. Techniques successful with unirradiated fuel were then tested in-cell with irradiated fuel.

A. SURFACE PREPARATION

It was not possible to successfully replicate as-etched fuel due to the pitting and extreme relief produced during electrolytic etching. However, the presence of hard zirconium carbide inclusions in SNAP fuel caused relief effects in as-polished samples that led to tearing of replicas. Consequently, attach-polishing and vibratory polishing techniques were evaluated.

1. Vibratory Polishing

Specimens were ground on 240-, 320-, and 600-grit SiC papers using copious amounts of water as the lubricant to prevent accumulation of material previously removed. Papers were changed after five minutes use, and the specimens were ultrasonically cleaned in water following each grinding step to remove all traces of abrasive.

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Figure 1. Equipment Used for Replication of SNAP Fuel In-Cell

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Rough polishing was accomplished using a rayon cloth and water slurries of 6- and 2- micron diamond paste for 15 minutes each. The final polishing took two hours on a Buehler Vibromet, using a slurry of Linde-B in a 10% oxalic acid solution on rayon cloth.

Replication of surfaces of unirradiated materials prepared by vibratory polishing out-of-cell showed resolution down to 200 to 300 Å. Replication of surfaces of irradiated materials prepared by vibratory polishing in-cell commonly showed scratches and disturbed metal. This was attributed to variation in abrasive cake depth (Linde-B), accentuated by the high in-cell air flow, which caused accelerated evaporation of the carrier. Attempts to replicate surfaces prepared by vibratory polishing in-cell were terminated.

2. Attack-Polishing

Grinding operations were identical to those described in the preceding section. Specimens were polished on rayon cloth, using alternate applications of polishing media. A Linde-B/H₂O slurry was applied to the polishing wheel and allowed to remain for five minutes. This was followed by the addition of a mixture containing 200 ml H₂O, 25 ml concentrated HNO₃, and 4 to 6 drops of 48% HF. After 30 seconds, a 10% oxalic acid solution was added in sufficient quantity to completely wash away the acid mixture. These operations continued for 1-1/2 hours, when the grain structure was faintly discernible, indicative of a faint chemical etch.⁽⁵⁾ Replications of surfaces prepared by attack-polishing with unirradiated and irradiated fuel were equally successful.

B. REPLICATION

The replicating solution was prepared by dissolving one gram of Faxfilm (cellulose acetate) in 50 ml of acetone. A squeeze bottle held by a master-slave manipulator was used to apply four drops of this solution to the specimen surface. A small strip of Faxfilm, 3.0 in. long by 1.0 in. wide by 0.005 in. thick, was immediately pressed onto the specimen surface. Approximately ten pounds of pressure was applied for 30 seconds by means of a spherical rubber tip (see Figure 1). After a three-minute wait to allow curing, the replica so obtained represented a negative of the entire specimen surface. This operation was repeated three to six times, depending on the surface condition of the specimen, to remove large particles of loose fuel and contaminants. The final replica was ultrasonically cleaned in water and removed from the cell.

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C. DECONTAMINATION

The cellulose acetate plastic used cannot be cleaned in concentrated acids such as HNO_3 or HF without being destroyed. Cleaning was accomplished by placing the replica in a 10% NH_4OH solution and shaking approximately 100 times. This was followed by three similar rinses in distilled H_2O . These operations removed much of the particulate contamination, lowering the activity of the replica to below 5 m-rad/hr (beta plus gamma). The replica was taped to a glass slide, holding up the face that contacted the specimen.

D. ELECTRON MICROSCOPY

The Faxfilm replicas were shadowed in a vacuum evaporator with B_2O_3 at an angle of 90 deg, chromium at 30 deg, and carbon at 90 deg. The entire assembly was placed in water to dissolve the B_2O_3 and plastic from the chromium-carbon secondary replica, which was then picked up with grids and inserted into the electron microscope for examination.

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III. ELECTRON MICROSCOPY ANALYSIS

A. UNIRRADIATED SNAP 8 FUEL

The unirradiated fuel examined consisted of three test samples heated to 1400°F with a temperature gradient that probably caused the H/Zr ratio to vary from 0.9 to 1.5. Optical metallography of the fuel revealed epsilon banding and microcracking. Differences between alpha uranium sites and carbide particles cannot be definitely determined from the electron microscope photographs; however, as a rule, the uranium sites are circular in shape while the carbide particles are more angular in nature.

Figure 2 is a typical example of the fuel surface showing microcracks, alpha uranium sites, coarse and fine twinning which are probably epsilon ghosts, and surface debris or artifacts. Figures 3, 4, and 5 show typical surface features on the second fuel sample. Figure 3 shows typical uranium and/or carbide sites along with banding. Whether the banding observed in this or subsequent photographs is real or ghosts can only be ascertained from the actual hydrogen content. Figure 4 indicates wide and fine banding. A phase boundary can be seen along one corner of the photograph. A large carbide particle can be seen in Figure 5. Gamma prime particles on a replica tend to be long and thin, as shown in Figure 6. Banding at right angles along with fine sub-banding is shown in Figure 7. This phenomenon could easily occur in fuel which had been repeatedly thermal cycled resulting in hydriding and dehydriding of the fuel. The features observed in these replicas of unirradiated fuel were used for comparison with the irradiated fuel replicas.

B. S8ER FUEL ANALYSIS

The first replicas to be discussed will be those taken from fuel regions of large volume changes (greater than 4% swelling based on hydrogen corrected data). These were regions of suspected delta-to-delta plus beta phase transformation during reactor operation. The postirradiation hydrogen content of these fuel samples ranged from 1.54 to 1.61 H/Zr. All of the samples contained a high carbon content ranging from 0.34 to 0.54 wt %.

The replicated fuel samples came from elements 432, 378, 232, 519, and 398. The location of the fuel sample from element 398 was a substitute sample,

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Figure 2. Typical Microcracks in Unirradiated U-ZrH Fuel
After Being in a Temperature Gradient

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Figure 3. Cross Banding in Unirradiated U-ZrH Fuel
After Being in a Temperature Gradient

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Figure 4. Fine and Coarse Banding in Unirradiated U-ZrH Fuel
After Being in a Temperature Gradient

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Figure 5. Large Carbide Particle in Unirradiated U-ZrH Fuel

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Figure 6. Possible Gamma Prime Particle in Unirradiated U-ZrH Fuel

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Figure 7. Cross Banding in Unirradiated U-ZrH Fuel

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unfortunately not in the desired location, and had to be taken from the lower portion of the fuel rod. The other four samples were obtained from the upper portion of the fuel rods in the region of maximum fuel temperature during reactor operation.

In all of the photographs, alpha uranium sites, epsilon banding and sub-banding, carbide particles, and phase boundaries were a common occurrence. Fission gas bubbles were observed in the fuel from elements 519, 432, and possibly 232. However, with insufficient magnification of the photographs on the replicas from element 232, it was not possible to positively resolve whether these particles were fission gas bubbles or surface debris, which was quite common on all replicas. It was of interest to note that there were bubble depletion regions around the large particles, particularly carbide and uranium sites. Also, grain or phase boundaries were noticeably void of fission gas bubbles. Figures 8, 9, and 10 show the surface features of the fuel from element 432. Figures 11 through 16 show typical surface features on the fuel from elements 378, 519, 232, and 398. Figures 14 and 15 show carbide and uranium particles lined up along a phase boundary. No evidence of fission gas bubbles was observed in the fuel structure of element 378, which had a large volume change. However, replication of the fuel from this element was poor, and no high magnification photographs were obtained which may have revealed the presence of bubbles.

Fission gas bubble density measurements were made at selected locations to determine the number of bubbles per cc. Also, distances were measured between the particles and the fission gas bubbles to determine the width of the bubble void areas around the particle sites. These results are discussed later in this report. A summary of the electron microscopy observations is presented in Table 2: Listed with these observations are the fuel rod diameter, percent diameter change, uncorrected and hydrogen corrected volume changes, carbon content, fuel burnup, hydrogen content, and optical metallography observations at the locations each of the replicas were taken.

Replicas were taken from two fuel surfaces which exhibited a large post-irradiation hydrogen content equal to or greater than the preirradiation values. Two elements which exhibited this characteristic were 111 and 378, 11-1/2 in. from the top of the element. These regions were suspected of crossing the

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Figure 8. Phase Boundary and Fission Gas Bubble Sites in the Element 432 Fuel Rod (3 in. from top)

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Figure 9. Bubble Depletion Region Around Large Particles
in the Element 432 Fuel Rod
(3 in. from top)

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Figure 10. Microcrack in the Element
432 Fuel Rod
(3 in. from top)

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Figure 11. Surface Features in the Element 378 Fuel Rod
(3 in. from top)

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Figure 12. Fission Gas Bubble Distribution in the Element 519 Fuel Rod
(6-1/2 in. from top)

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CRD (GP-1)
63-6-2

PHOTOGRAPHED AT 5,513X
NO REDUCTION

Figure 13. Fission Gas Bubble Depletion Regions Around Large Particle Sites in the Element 519 Fuel Rod (6-1/2 in. from top)

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CRD (GP-1)
63-7-1

PHOTOGRAPHED AT 5,624X
NO REDUCTION

Figure 14. Phase Boundary in the Element 232 Fuel Rod
(5-3/4 in. from top)

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CRD (GP-1)
63-2-2

PHOTOGRAPHED AT 6,660X
NO REDUCTION

Figure 15. Possible Phase Boundary in the Element 398 Fuel Rod
(11 in. from top)

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CRD (GP-1)
63-2-3

PHOTOGRAPHED AT 6,660X
NO REDUCTION

Figure 16. Surface Features in the Element 398 Fuel Rod
(11 in. from top)

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Element No.	Core Location	Axial Location on Rod (Dist from Top) (in.)	Fuel Rod Diam		Volume Change		Carbon Content (wt %)	Fuel Burnup (met.at.%)	Optical Metallography Observations and Post H/Zr Content	Remarks on Reason for Choice of Sample	Electron Microscopy Observations		
			Diam (in.)	$\Delta D/D$ (%)	$\frac{\Delta V}{V}$ (%)	$\frac{\Delta V}{V} H_2$ Corr (%)					Micro-cracks	Fission Gas Bubbles	Miscellaneous Observation
IP3, 4, 5	Unirradiated								ϵ + microcracks H/Zr = 0.0 to 1.5	Control sample to determine difference between irradiated and unirradiated.	Yes	No	α U sites, ϵ twinning ghosts, debris, carbide particles, possible γ' , ϵ banding + fine sub-banding, boundary line.
	S8ER												
E-432 (intact)	VIII-29	3	0.5395	+1.35	+3.94	+4.25	0.54	0.14	δ , longit. mount H/Zr = 1.61	Sample taken from peak distortion area of fuel rod — large $\Delta V/V$	Yes	Yes	α U sites, carbide particles, ϵ banding, phase boundary with bubble depleted region in the boundary and around C or U particles.
E-332 (weld failure)	VIII-25	1-1/2	0.5210	-2.15	-5.83	-0.39	0.55	0.10	α + δ eutectoid H/Zr = 0.92	Gross dehydriding to observe the structure	Yes	No	α U sites, possible two phase region, possible carbide particles.
E-188 (intact)	VIII-6	5-1/2	0.5332	+0.17	-0.47	+1.00	0.01	0.19	δ + γ' columnar structure, circum. cracks at 250X, H/Zr = 1.47, ϵ ghost bands	Large H_2 loss for intact element	No	No	α U sites, carbide particles, surface scratches, phase boundary, ϵ band ghosts.
E-378 (cracked)	IX-28	3	0.5430	+1.99	+4.56	+4.98	0.34	0.15	δ , longit. mount, faint banding at 500X, crack in fuel, H/Zr = 1.58	Sample taken from peak distortion area of fuel rod	No	No	α U sites, carbide particles. Poor replicas, insufficient detail for analysis.
E-378 (cracked)	IX-28	11-1/2	0.5362	+0.71	+0.91	+1.72	0.34	0.14	ϵ , H/Zr = 1.72	Region of suspected δ to ϵ transformation, high final H/Zr			Poor replicas, insufficient detail for analysis.
E-111 (cracked)	IX-42	11-1/2	0.5376	+0.99	+1.45	+1.50	0.01	0.14	ϵ , large voids H/Zr = 1.73	Sample taken from peak distortion area of fuel rod, region of suspected δ to ϵ transformation, high final H/Zr	No	No	α U sites, carbide particles, ϵ banding.
E-232 (intact)	VI-24	5-3/4	0.5376	+1.01	+3.25	+4.02	0.54	0.21	δ phase with banding, H/Zr = 1.56	Region of suspected δ to δ + β transformation, large $\Delta V/V$	No	Possible	α U sites, carbide particles, ϵ banding, phase boundary, possible bubble depletion zone around phase boundary and large particles (magnif. too low for positive identification).

Table 2 (Sheet 1 of 2)
Electron Microscopy Observations
of Irradiated SNAP 8 Fuel Samples

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Element No.	Core Location	Axial Location on Rod (Dist from Top) (in.)	Fuel Rod Diam		Volume Change		Carbon Content (wt %)	Fuel Burnup (met.at.%)	Optical Metallography Observations and Post H/Zr Content	Remarks on Reason for Choice of Sample	Electron Microscopy Observations		
			Diam (in.)	$\Delta D/D$ (%)	$\frac{\Delta V}{V}$ (%)	$\frac{\Delta V}{V} H_2$ Corr (%)					Micro-cracks	Fission Gas Bubbles	Miscellaneous Observations
E-519 (cracked)	IV-1	6-1/2	0.5415	+1.76	+3.72	+4.58	0.50	0.23	$\delta + \gamma'$ with faint ϵ banding ghosts, circum. cracks at 250X, H/Zr = 1.54	Sample was taken from peak distortion area of rod, region of suspected δ to $\delta + \beta$ trans., large $\Delta V/V$	No	Yes	α U sites, carbide particles, bubble depletion zone around large particles, ϵ banding ghosts, possible former grain boundary.
E-398 (cracked)	IV-13	11	0.5360	+0.73	+0.54	+0.91	0.40	0.17	δ with faint ϵ banding, H/Zr = 1.60	Substituted for requested sample which could not be obtained	No	No	α U sites, carbide particles, ϵ banding, phase boundaries.
	NAA 115-1												
IS-4 (intact)		2	0.5327	+0.98	+4.16	+4.30	0.40	0.41	ϵ , H/Zr = 1.64	Large $\Delta V/V$, only fuel rod with 0.4 w/o C in expt.	Yes	Yes	α U sites, ϵ banding, boundary lines, bubble depletion region around large particles, sample may be at point of incipient gross swelling, carbide particles.
IS-7 (intact)		2	0.5312	+0.73	+2.94	+4.11	0.10	0.41	δ phase with γ' at center, faint banding at edge, cracks from center to edge, H/Zr = 1.50	Large $\Delta V/V$	No	Yes	α U sites, ϵ banding, carbide particles, bubble depletion region around large particles, poor replicas.
IS-8 (intact)		2	0.5315	-0.43	+0.74	+2.08	0.10	0.43	Center and mid-radius area was $\delta + \gamma'$, edge was δ with faint banding, large cracks from edge to center, H/Zr = 1.45	Large $\Delta V/V$, large H_2 loss of -0.37 H/Zr	No	Yes	α U sites, ϵ banding and fine sub-banding, carbide particles, phase boundary, γ' particle, no apparent bubble depletion around large particles as was observed on other samples.

Table 2 (Sheet 2 of 2)
Electron Microscopy Observations
of Irradiated SNAP 8 Fuel Samples

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delta, delta plus epsilon, epsilon phase boundary with moderate fuel swelling of approximately 1.50%. Both samples examined had a postirradiation hydrogen content greater than 1.70 H/Zr. Replicas obtained from the element 378 fuel at 11-1/2 in. from the top were not of good quality and gave insufficient detail for analysis.

Optical metallography on the fuel from element 111 revealed epsilon phase with voids in the fuel. The replica was taken from the peak distortion area of the rod. No evidence of microcracking or fission gas bubbles was observed (at 6000X magnification, the limit of resolution is 1000 Å). Figures 17 and 18 show the surface features of the fuel. Epsilon cross banding, uranium sites, and carbide sites were common.

The remaining two fuel wafers replicated were chosen as a result of high hydrogen loss from the fuel. Element 332 exhibited gross dehydriding due to a weld failure. Element 188 was examined as it was the only intact element to exhibit dehydriding down to 1.47 H/Zr. Metallography of this fuel showed delta plus gamma prime with banding ghosts at 5-1/2 in. from the top. Metallography of rod 332, 1-1/2 in. from the top, showed an alpha-delta eutectoid composition with an H/Zr of 0.92.

Replication of the element 188 fuel revealed the normal particle sites and banding. No microcracks or fission gas bubbles were observed (see Figure 19). Replication of the fuel surface from element 332 showed the normal particle sites with some microcracking. Typical surface features are shown in Figures 20 and 21. A summary of the data is presented in Table 2.

C. NAA 115-1 FUEL ANALYSIS

The three fuel surfaces replicated were from elements IS-4, IS-7, and IS-8. All three surfaces were from regions of large volume changes ranging from 2.08 to 4.30% (H₂ corrected). These samples experienced a fuel burnup of 0.41 to 0.43 met. at. %.⁽⁶⁾ Optical metallographic examination revealed delta plus gamma prime phases in IS-7 and -8 with predominantly delta plus epsilon phase in IS-4. Also, radial cracks were observed in IS-7 and -8. The post-irradiation hydrogen content ranged from 1.45 in IS-8 to 1.64 in IS-4.

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CRD (GP-1)
64-1-2

PHOTOGRAPHED AT 17,760X
NO REDUCTION

Figure 17. Carbide Particles in the Element 111 Fuel Rod
(11-1/2 in. from top)

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CRD (GP-1)
64-1-3

PHOTOGRAPHED AT 5,624X
NO REDUCTION

Figure 18. Surface Features in the Element 111 Fuel Rod
(11-1/2 in. from top)

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CRD (GP-1)
64-8-2

PHOTOGRAPHED AT 5,976X
NO REDUCTION

Figure 19. Surface Features in the Element 188 Fuel Rod
(5-1/2 in. from top)

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CRD (GP-1)
64-6-2

PHOTOGRAPHED AT 14,400X
NO REDUCTION

Figure 20. Possible Two-Phase Region in the Element 332 Fuel Rod
(1-1/2 in. from top)

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CRD (GP-1)
64-6-3

PHOTOGRAPHED AT 9,000X
NO REDUCTION

Figure 21. Surface Features of the Element 332 Fuel Rod
(1-1/2 in. from top)

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Electron microscopy indicated the presence of fission gas bubbles in all three fuel rods; however, microcracks were observed only in IS-4. Alpha uranium and carbide sites, phase boundary lines, and banding were common in all three replicas. Fission gas bubble depletion regions were observed around the large particle sites and phase boundaries in IS-4 and IS-7. This depletion region was not found in IS-8, which experienced half the volume change of the other two fuel samples.

Surface features of IS-4 fuel are shown in Figures 22 to 24. A typical phase boundary is shown in Figure 24.

Replication of the fuel surface from IS-7 was generally poor; however, Figure 25 shows some of the surface features from this fuel rod.

Typical surface features of IS-8 are shown in Figures 26 to 29. Very fine banding can be seen in Figure 27. Figure 28 shows what may be some fission gas bubble agglomeration, but this cannot be positively ascertained from the photograph. A similar phenomenon has been observed in lower magnification photographs on irradiated fuel from other samples as shown in Figures 15 and 16. Examination of the unirradiated fuel replicas did not show this same characteristic. This possible agglomeration may also be the result of chemical attack by the etchant during sample preparation. Irradiated fuel is more susceptible to chemical attack than unirradiated fuel. A typical phase boundary and a gamma prime particle are shown in Figure 29. A summary of the electron microscope observations are tabulated in Table 2.

Fission gas bubble size and bubble density measurements were taken from the replica photographs. The results of these measurements are discussed in the next section of the report.

D. FISSION GAS BUBBLE ANALYSIS

Representative photographs were selected from those fuel surfaces which exhibited fission gas bubbles. From these photographs, the bubble volume fraction and bubble density in terms of bubbles/cc were calculated. Also, representative length measurements were taken of the distance between particles and the start of fission gas bubbles with the larger uranium and carbide particles. These large particles may act as trapping sites for the fission gas bubbles.

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CRD (GP-1)
63-5-3

PHOTOGRAPHED AT 5,513X
NO REDUCTION

Figure 22. Microcracks in the IS-4 Fuel Rod

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CRD (GP-1)
63-5-4

PHOTOGRAPHED AT 17,020X
REDUCED TO 15,062.7X

Figure 23. Fission Gas Bubbles in the IS-4 Fuel Rod

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CRD (GP-1)
63-5-5, -6

PHOTOGRAPHED AT 17,020X
REDUCED TO 14,467X

Figure 24. Phase Boundary in the
IS-4 Fuel Rod

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CRD (GP-1)
63-4-2

PHOTOGRAPHED AT 6,660X
NO REDUCTION

Figure 25. Surface Features of the IS-7 Fuel Rod

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CRD (GP-1)
63-8-2

PHOTOGRAPHED AT 17,760X
NO REDUCTION

Figure 26. Carbide Particle in the IS-8 Fuel Rod

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CRD (GP-1)
63-8-3

PHOTOGRAPHED AT 14,800X
REDUCED TO 12,580X

Figure 27. Fine Banding in the IS-8 Fuel Rod

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CRD (GP-1)
63-8-5

PHOTOGRAPHED AT 44,400X
REDUCED TO 37,296X

Figure 28. Fission Gas Bubbles in the IS-8 Fuel Rod

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CRD (GP-1)
63-8-7, -8

PHOTOGRAPHED AT 6,290X
REDUCED TO 5,346.5X

Figure 29. Phase Boundary and
Gamma Prime Phase in the
IS-8 Fuel Rod

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The bubbles were counted at various locations on a photograph by inscribing a square on the photograph and physically counting the number of bubbles within the square. This was done by using a quantitative metallographic particle size counter. To simplify the subsequent calculation, the assumptions were made that all particles on any one photograph were of uniform diameter,⁽⁶⁾ the bubble distribution in the inscribed square was uniform, and a cube formed by projecting the square into the fuel surface allowed calculation of the bubble volume fraction by assuming the volume fraction was equal to area fraction. From this information, the contribution of the formation of fission gas bubbles to the observed fuel swelling can be determined. The number of fission gas bubbles per unit volume was also calculated in a manner similar to that reported by Kramer.⁽⁷⁾

All bubble counts were taken in areas unaffected by void depletion zones around particle sites. From 4 to 10 squares were counted in each photograph measured. The location and identification of each counting square is shown in Figures 8 through 29 on those figures which had fission gas bubbles. Some of the measurements were made on photographs not reproduced in the report. Table 3 is a tabulation of the fission gas bubble volume fractions and bubble density for each counting square. The volume fractions are tabulated as percent of the volume due to bubbles. Measurements were taken on the five fuel samples which indicated positively the presence of fission gas bubbles.

A summary of the average bubble volume fraction as compared to the measured volumetric increases on the fuel is presented in Table 4. Due to the bubble depletion regions around particle sites and grain boundaries, the bubble volume fractions determined from the counting squares are not necessarily representative of volume fractions for the fuel since the counting squares were selected in regions away from particle sites and grain boundaries. Thus, a visual estimate was made of the fractional area covered by the bubbles in each photograph measured. The last column in Table 4 is the estimated contribution of the fission gas bubbles to the total fuel volume change due to irradiation.

The volume change due to fission gas bubble formation in the two S8ER samples examined accounted for approximately 60% of the observed uncorrected volume change. If it is assumed that rehydrating the fuel to its preirradiation level does not affect the bubble size or density, then a comparison can be made

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TABLE 3
FISSION GAS BUBBLE MEASUREMENTS

Element No.	Figure No.	Counting Square No.	Bubble Volume Fraction (%)	Bubble Density (bubbles/cc) (x 10 ¹³)	Element No.	Figure No.	Counting Square No.	Bubble Volume Fraction (%)	Bubble Density (bubbles/cc) (x 10 ¹³)		
519 (S8ER)	12	1	5.63	3.52	IS-4 (115-1)	23	8	2.00	2.75		
		2	5.11	3.19			9	0.81	1.11		
		3	3.20	2.00			10	0.98	1.34		
		4	3.62	2.26		24	1	0.84	1.24		
		5	4.28	2.68			2	0.78	1.16		
	13	1	3.58	2.24			3	0.71	1.06		
		2	4.52	2.82			4	0.97	1.43		
		3	3.00	1.88			5	0.96	1.43		
		4	2.88	1.80		6	0.91	1.47			
		5	2.63	1.64		7	1.07	1.73			
		Avg	3.85	2.40		8	0.91	1.47			
						9	1.13	1.82			
432 (115-1)	8	1	2.27	4.33	IS-7 (115-1)	25	10	1.44	2.32		
		2	2.74	5.23			11	1.27	2.04		
		3	2.38	5.20				Avg	1.13	1.69	
		4	3.88	8.50			IS-8 (115-1)	27	1	0.35	4.22
		5	5.47	11.98					2	0.43	5.27
		6	4.05	8.87					3	0.57	6.86
		7	7.03	15.38					4	0.43	5.22
		8	4.56	9.97					Not repro-duced	1	0.41
		9	2.82	6.17			2	0.38		3.07	
		10	2.36	5.17			3	0.36		2.90	
		11	3.54	7.76	4	0.49	3.93				
		12	3.02	6.61	5	0.53	4.26				
	9	1	1.50	7.62	28	1	0.11	1.58			
		2	1.48	7.48		2	0.22	3.26			
						3	0.25	3.65			
	10	1	1.31	3.55		4	0.52	7.68			
		2	1.47	4.01		5	0.36	5.32			
		3	1.65	4.48		6	0.36	5.32			
		Avg	3.03	7.19		7	0.39	5.77			
IS-4 (115-1)	Not repro-duced	1	0.63	1.21		8	0.25	3.65			
		2	0.63	1.21		29	1	0.68	3.18		
		3	0.47	0.89			2	0.78	3.64		
		4	0.88	1.69			3	0.98	4.54		
		5	0.19	0.36			4	1.00	4.65		
	23	1	1.40	1.92			5	0.61	2.85		
		2	1.78	2.44		Avg	0.47	4.27			
		3	1.39	1.92							
		4	2.04	2.80							
		5	2.28	3.13							
		6	1.40	1.92							
		7	1.60	2.20							

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TABLE 4
VOLUME CHANGE DUE TO FISSION GAS BUBBLE FORMATION

Element No.	Fuel $\Delta V/V$ (%)	Fuel $\Delta V/V$ (%) (H ₂ Corrected)	Fission Gas Bubble Volume (%)	Estimated Volume Corr Factor	Fuel $\Delta V/V$ Due to Fission Gas Bubbles (%)
519	3.72	4.58	3.85	0.6	2.31
432	3.94	4.25	3.03	0.7	2.12
IS-4	4.16	4.30	1.13	0.9	1.01
IS-7	2.94	4.11	6.78	0.6	4.06
IS-8	0.74	2.08	0.47	0.9	0.42

with the observed hydrogen-corrected volume change. For the two S8ER samples, the fission gas bubbles accounted for approximately 50% of the hydrogen-corrected volume change.

Microcracks observed in the fuel from element 432 may also have added a significant contribution to the fuel swelling.

Based on the measured fuel volume changes in the S8ER samples replicated, the fuel sample examined from element 378 near the top should have revealed microcracks or fission gas bubbles. The measured fuel volume change was greater than either of the 432 or 519 samples. The poor replication results on this sample may not have allowed sufficient resolution to identify the surface features of the fuel. Also, the sample from element 232 which had a large volume change did not give sufficient resolution to detect bubbles.

The contribution of fission gas bubbles to observed hydrogen-corrected volume change in two of the NAA 115-1 fuel specimens examined was less than that observed in the S8ER samples. About 25% of the observed volume change (uncorrected and corrected) in IS-4 was due to bubble formation. Bubble formation from IS-8 accounted for 57% of the uncorrected fuel volume change and only 20% of the hydrogen-corrected volume change.

The IS-7 data in Table 4 may not be representative of the volume change due to bubble formation. Replication of this sample was poor and only two

counting squares were obtained from one photograph shown in Figure 25. The bubble population distribution was quite varied, and the counting squares were taken from high bubble density areas to avoid the effects of the bubble depletion zones around large particle sites. Assuming the data are representative, however, it is interesting to note that the volume change due to bubble formation accounted for all of the observed hydrogen-corrected volume change. It is unreasonable to assume that all the volume change was due to bubble formation, but it may have accounted for 80% of total swelling. This is not unrealistic since IS-7 was believed to have operated in beta phase during irradiation due to high hydrogen loss.⁽²⁾ This would allow for additional bubble formation with phase change during irradiation.

Annealing studies performed on the fuel from elements 432 and IS-4 have indicated no gross swelling at temperatures up to 1700°F. Similar studies on IS-7 fuel revealed gross swelling, indicating beta phase fuel annealing characteristics.⁽⁸⁾

Length measurements were taken on the distance between large particle sites and the start of the fission gas bubble sites. This was done to determine the width of the bubble depletion zones. Bubble depletion regions on approximately 14 particles were measured on each of seven photographs. The average values and the range of data for each photograph measured are listed in Table 5.

TABLE 5
BUBBLE DEPLETION ZONE WIDTH MEASUREMENTS ABOUT
URANIUM AND CARBIDE PARTICLE SITES

Element No.	Figure No.	Number of Particle Zones Measured	Average Zone Width (Microns)	Zone Width Range (Microns)
519	11	13	0.83	0.58 - 1.22
	12	9	1.29	0.78 - 2.07
432	7	26	0.57	0.33 - 1.13
	8	17	0.63	0.40 - 1.07
	9	19	0.64	0.40 - 0.87

Only the S8ER samples were analyzed since the zones observed on the NAA 115-1 samples were not easily defined as to fission gas boundary lines. Depletion zones on the S8ER samples were more easily resolved. The average bubble depletion zone width was 1.07 and 0.61 microns for fuel rods 519 and 432, respectively.

An average bubble diameter was calculated to be 0.90×10^{-5} mm (900 Å) with a range from 0.51 to 1.45×10^{-5} mm in diameter. An estimate of the relative bubble size in the fuel can be obtained by dividing the average volume fraction due to bubble formation by the average number of bubbles/cc as listed in Table 3. The bubbles in IS-7 were seven times larger than the other NAA 115-1 samples examined. This may indicate a relationship between bubble size and phase or hydrogen content during irradiation.

About ten measurements were taken of the distance between bubbles. These data indicated an average distance of 0.212 microns (2.12×10^{-4} mm) between bubbles.

Measurements were made of the uranium particle density in five of the irradiated samples and the unirradiated fuel to determine if any change occurred due to irradiation. Uranium particle measurements on the unirradiated fuel ranged from 1.1×10^{10} to 3.4×10^{11} precipitates per cc. Measurements on the three NAA 115-1 fuel samples and elements 432 and 398 from the S8ER fuel indicated a uranium precipitate density ranging from 1.9×10^{10} to 1.0×10^{11} particles per cc. No measurable decrease in precipitates after irradiation was observed. The order of magnitude of uranium precipitate density found in these samples is in agreement with limited data reported by Taketani⁽⁶⁾ from 3 to 6×10^{10} particles per cc.

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IV. SUMMARY AND CONCLUSIONS

Resolution of the irradiated U-ZrH fuel was generally of high quality and allowed identification of such features as alpha uranium sites, carbide particle sites, phase boundaries, grain boundaries, banding, microcracks, and fission gas bubbles.

Microcracking in the fuel did not appear to be a major contributor to fuel swelling in two of the fuel rods examined from peak distortion areas. Fission gas bubble formation accounted for approximately 50% of the observed hydrogen-corrected volume change in this peak distortion region. Two of the NAA 115-1 fuel specimens with a burnup of ~ 0.4 met. at. % showed a lower percentage contribution of fission gas bubbles to observed volume change than that indicated by the S8ER fuel with a burnup of ~ 0.2 met. at. %.

In the S8ER fuel rods examined, no evidence of fission gas bubbles was found in the lower or cooler end of the rods. The peak distortion areas of the rods occurred from 3 to 5 in. from the top in the peak fuel temperature region. Samples with similar postirradiation hydrogen content were taken from peak distortion areas on some rods and the lower end of other rods. This indicates a possible dependence of fission gas bubble size and density on temperature. However, there did not appear to be a correlation with fuel burnup.

Only sample IS-7 gave indication that most of the observed volume change could be attributed to bubble formation. This sample had significantly larger bubbles (although the data are limited) than the other specimens and was believed to have operated in or near beta phase based on annealing studies and post-irradiation hydrogen analyses. Hence, there may be a correlation between bubble size and phase. Studies with other irradiated fuels have shown that changes in structural properties through phase change or thermal cycling can affect fuel swelling.⁽⁹⁾ Phase change from delta to beta in the U-ZrH system may allow larger bubbles to be formed.

Based on the fuel samples examined, there was no evidence of fission gas bubble sweeping during phase change. No correlation was found between the location of the fission gas bubble sites and epsilon banding or microcracks.

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Bubble depletion regions observed around large particle sites are thought to be a result of interaction of the gas bubbles with the larger uranium and carbide particles. The bubbles probably exist in this region but are too small to be detected. Similar observations were noted by Kramer et al.⁽¹⁰⁾ in their work on fission gas bubble studies in U-Mo alloys.

Fission gas bubble density measurements on the order of 10^{13} bubbles/cc were typical. The average diameter of these bubbles was $900 \text{ \AA}$ with a mean distance between bubbles of $2.1 \times 10^{-4} \text{ mm}$.

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