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REPORT TITLE **Electron Microscopy Analysis of Irradiated S8ER**
and NAA 115-1 fuel (title unclassified)

AUTHOR **K. J. Miller**

NAA-SR-MEMO 12368

GROUP 1

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ATOMICS INTERNATIONAL A Division of North American Aviation, Inc.		NAA-SR-MEMO XXXX 12368	APPROVALS <i>W.R. McCurm</i>
TECHNICAL DATA RECORD		PAGE 1 OF 51	<i>C. J. Kohl</i>
AUTHOR K. J. Miller		DEPT. & GROUP NO. 737-756	DATE 3-7-67
TITLE Electron Microscopy Analysis of Irradiated S8ER and NAA 115-1 Fuel (Title Unclassified)		GO NO 7695	S/A NO 35310 TWR
PROGRAM SNAP 8	SUBACCOUNT TITLE SNAP 8 Fuel Element Examination	SECURITY CLASSIFICATION (CHECK ONE BOX ONLY) AEC DOD UNCL. ☐ ☐ CONF. ☒ ☐ SECRET ☐ ☐ (CHECK ONE BOX ONLY) RESTRICTED DATA ☒ DEFENSE INFO. ☐	
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J.G. Asquith C02	STATEMENT OF PROBLEM		
K. R. Birney C02	Perform electron microscopy by replication of irradiated S8ER and NAA 115-1 fuel to develop remote replication techniques and to further characterize the fuel structure.		
C.D. Calkins C02	ABSTRACT:		
E.M. Chandler C01	Based on the results of the S8ER detailed examination and the NAA 115-1 post-irradiation examination, selected fuel samples were chosen from regions of interest for electron microscopy to develop remote replication techniques at AIHL and to further characterize the fuel structure beyond that possible with optical metallography. Fuel samples were chosen from regions of large volume changes as characterized by peak distortions in the fuel rods and from regions of excessive hydrogen loss. The samples were prepared and replicated by the Faxfilm technique at AIHL, and the replicas were sent to Dept. 742 at AI/Canoga for shadowing, examination, and photography.		
R. Courtright C01	Examination of the replica photographs revealed alpha uranium sites, carbide particle sites, phase boundaries, epsilon banding, and microcracks. Fission gas bubbles were observed in two S8ER samples from peak distortion areas and in all three NAA 115-1 samples examined. Bubble density measurements on the order of 10^{13} bubbles/cc were typical with an average bubble diameter of 900°A .		
H. Dieckamp C02	The major contribution to fuel swelling in the S8ER fuel from peak distortion regions of the fuel rod was fission gas bubble formation. Fuel microcracking was observed in some samples, but was not as extensive as expected. Fission gas bubble formation was noted in all the NAA 115-1 fuel samples examined, but was not excessive. Apparent bubble depletion regions were observed around large particle sites and phase boundaries. This is thought to be a result of a gas bubble interaction region with the particle sites. Also, no correlation was observed of epsilon banding with the location of gas bubbles, particle sites, or microcracks.		
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C.J. Engberg C01			
E.D. Falk C01			
R. Forrester SS20			
B.B. Gillies C01			
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The author is indebted to those personnel who helped to develop the replication techniques on irradiated fuel and in the subsequent electron microscope examinations and photography by A. G. Pard. I would particularly like to thank Dr. W. V. Johnston from the NAA Science Center for his help in the interpretation of the electron microscope photographs.

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ELECTRON MICROSCOPE REPORT

A. INTRODUCTION

Results of the SNAP 8 Experimental Reactor (S8ER) destructive examination revealed localized fuel swelling behavior at various axial locations in the fuel rods. This observed localized fuel instability was attributed to axial element restraint, hydrogen redistribution phenomena, fuel burnup, and thermal transient stresses in the elements during irradiation^(1,2). During the destructive examination, fuel metallography samples were obtained from the fuel rods at selected locations which were representative of the various observed fuel swelling characteristics. In addition to the S8ER fuel samples cut for evaluation, three samples from the NAA 115-1 fuel rods were included for analysis.

Metallographic examination of the fuel revealed the microstructure and surface features, such as cracks and NaK reaction effects at magnifications up to 500X. It was felt that further characterization of the fuel structure could be made through the use of an electron microscopic examination at selected fuel rod locations. From this information, it may be possible to detect microcracks not observed by optical microscopy and also to determine fission gas bubble density. The samples to be examined were chosen from the following areas of fuel behavior⁽³⁾.

1. Regions of suspected delta to delta + 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 pre-irradiation values.
3. Unusual phenomena, such as very large volume changes and unexpected post-irradiation hydrogen contents.

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. Each sample and its reason for selection as listed above are tabulated in Table 1.

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TABLE ISample Selection for EM Analysis

<u>Expt.</u>	<u>Element No.</u>	<u>Core Location</u>	<u>Axial Location on Rod (Dist. from Top, in.)</u>	<u>Reason for Selection as Listed in Text</u>
S8ER	432	VIII-29	3	1
S8ER	332	VIII-25	1½	3
S8ER	188	VIII-6	5½	3
S8ER	378	IX-28	3	1
S8ER	378	IX-28	11½	2
S8ER	111	IX-42	11½	2
S8ER	232	VI-24	5 3/4	1
S8ER	519	IV-1	6½	1
S8ER	398	IV-13	11	1
115-1	IS-4		2	1
115-1	IS-7		2	1
115-1	IS-8		2	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.

The replicas were pulled from the specimens at the Hot Lab, sent to Dept. 742 at AI/Canoga for shadowing and examination, and the examination negatives returned to the Hot Lab for printing and final evaluation. From the photographs, particles were identified and measurement of fission gas bubble densities was obtained.

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B. SAMPLE PREPARATION

1. AIHL

The fuel specimens were ground on 240, 320, and 600 grit silicon carbide paper using copious amounts of water as the lubricant to prevent accumulation of material previously removed. After grinding was completed, the samples were polished. The polishing media were added in the following manner: First, two applications of a Linde B-H₂O slurry were made over a five minute period. This was followed by the addition of a mixture of 200 ml. H₂O, 25 ml. conc. HNO₃, and 4 to 6 drops of 48 percent HF. After 30 seconds, a solution of 10% oxalic acid was added in sufficient quantity to completely wash away the HNO₃-HF mixture. The amount of Linde B added was decreased as polishing progressed. The relative amounts of H₂O, HNO₃, and HF in the attach polish solution were varied with each sample, depending on its burnup, thermal history, and surface condition. Each sample was examined prior to replication to insure the surface was properly polished.

Replication of the surface was initiated by applying a solution of Faxfilm and acetone to the surface by dipping a brush into the solution and lightly brushing the specimen. A small strip of Faxfilm (larger than the sample mount) was immediately pressed to the fuel surface and held in place by applying a force of five pounds for 30 seconds. After allowing the replica to cure for three minutes, it was removed by peeling the strip of Faxfilm from the mount. The replica was then taped, face up, to a glass slide.

The replicas were removed from the cell and decontaminated by washing the replicas first in distilled water, then ammonium hydroxide, and finally in distilled water again to remove all traces of the hydroxide prior to shipping to Dept. 742 for examination.

2. Electron Microscope Laboratory

Upon receipt of the replicas, they were placed in a vacuum evaporator and appropriately shadowed. A deposit of boron oxide (B₂O₃) was applied to the replica surfaces. Chromium was applied which effected the actual shadowing. Carbon was then deposited and supported by the chromium during its removal from the Faxfilm.

After removing the replicas from the evaporator, the actual replica area to be examined was cut away from the rest of the Faxfilm and held on a water surface in a large beaker. The water dissolved the B₂O₃ and the chromium-carbon deposit floated away from the plastic forming a second replica. This second replica was then picked up on electron microscope grids and placed in the microscope specimen chamber for examination.

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The replicas were examined to determine where on the fuel cross-section good replication occurred. This area was chosen for analysis. Generally, the area examined was obtained near the center of the fuel cross-section. Magnification of the final photographs ranged from 3000X to 44,000X. With this being a first attempt at replication of irradiated SNAP 8 fuel, approximately 60% of the replicas taken were suitable for final photographic reproduction. Improved techniques on future specimens based on the experience with the replicas described in this report should allow for a higher recovery factor.

C. DISCUSSION OF RESULTS

At this point, a few details should be noted about the electron microscopy analyses. A replica sample provides information only on topography. The sample is etched selectively to provide "mountains and valleys" which are distinguishable by shadowing effects. As a result, the interpretation is no better or worse than the initial preparation. Also, the size scale of resolvable defects is on the order of 300 Å (3×10^{-6} cm). This is probable limit of resolution using the Faxfilm replication technique.

With the replicas described in this report, the major interest has been to evaluate existing methods on unirradiated SNAP 8 fuel and to compare the results with replicas obtained remotely at the Hot Lab. A secondary effort has been expended on improving the resolution of the replica surfaces⁽⁴⁾.

The shadowing angle aids in evaluating the topological details such as whether a particle on the surface of the replica is raised or depressed into the fuel surface. A replica is also a negative of the fuel surface such that a depression on the replica is a raised particle in the fuel surface as shown below: A fission gas bubble, for example, which is actually a void or hole in the fuel surface would appear as a raised particle in the photograph. The front surface of this particle would appear dark and the back surface would appear light.

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(DIAGRAM 1 - OBTAINING A REPLICA)

(DIAGRAM 2 - REPLICA SURFACE)

(DIAGRAM 3 - POSITIVE PRINT DEVELOPED FROM REPLICA NEGATIVE)

1. 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. It cannot be definitely determined from the electron microscope photographs differences between alpha uranium sites and carbide particles. However, as a rule, the uranium sites are circular in shape while the carbide particles are more angular in nature.

Figure 1 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. Figure 2, 3, and 4 show typical surface features on the second fuel sample. Figure 2 shows typical uranium and/or carbide sites along with banding. Whether the banding observed in this or

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SHADOW
ANGLE

FIGURE 1 ♦ TYPICAL MICROCRACKS IN UN-IRRADIATED U-ZrH FUEL AFTER BEING IN A TEMPERATURE GRADIENT (6000X)

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IP4-5

SHADOW
ANGLE

FIGURE 2 ♦ CROSS-BANDING IN UNIRRADIATED
U-ZrH FUEL AFTER BEING IN A
TEMPERATURE GRADIENT
(15,000X)

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SHADOW
ANGLE

FIGURE 3 ♦ FINE AND COARSE BANDING IN UN-IRRADIATED U-ZrH FUEL AFTER BEING IN A TEMPERATURE GRADIENT (15,000X)

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SHADOW
ANGLE

FIGURE 4 ♦ LARGE CARBIDE PARTICLE IN UN-
IRRADIATED U-ZrH FUEL
(15,000X)

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subsequent photographs is real or ghosts can only be ascertained from the actual hydrogen content. Figure 3 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 4. Gamma prime particles on a replica tend to be long and thin as shown in Figure 5. Banding at right angles along with fine sub-banding is shown in Figure 6. This phenomena 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.

2. SEER

Some of the fuel samples were prepared for replication using the vibramet polishing techniques to compare this process with the normal Hot Lab polishing operations. Replication of these samples in many instances revealed surface scratches, flowed metal, and no true structure. Generally, the particles throughout the matrix were barely resolvable, indicating this technique was not suitable for replication procedures(5).

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 post-irradiation 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 w/o.

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, 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

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↑
1μ
↑

IP5-1

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SHADOW
ANGLE

FIGURE 5 ♦ POSSIBLE GAMMA PRIME PARTICLE IN
UNIRRADIATED U-ZrH FUEL
(6000X)

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IP5-2

FIGURE 6 ♦ CROSS-BANDING IN UNIRRADIATED
U-ZrH FUEL
(6000X)

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SHADOW
ANGLE

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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. See Figures 7, 8, and 9, which show the surface features of the fuel from element 432. Figures 10 through 15 show typical surface features on the fuel from elements 378, 519, 232, and 398. Figures 13 and 14 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 are listed 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 pre-irradiation values. Two elements which exhibited this characteristic were 111 and 378, $11\frac{1}{2}$ inches from the top of the element. These regions were suspected of crossing the delta, delta + epsilon, epsilon phase boundary with moderate fuel swelling of approximately 1.50 percent. Both samples examined had a post-irradiation hydrogen content greater than 1.70 H/Zr. Replicas obtained from the element 378 fuel at $11\frac{1}{2}$ inches 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, the limit of resolution is 1000°A). Figures 16, and 17 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 $5\frac{1}{2}$ inches from the top. Metallography of rod 332, $1\frac{1}{2}$ inches from the top, showed an alpha-

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63-10-2

63-10-3

1 μ

PHOTO CROPPED FOR REPORT

FISSION GAS BUBBLE COUNTING SQUARE

SHADOW
ANGLE

FIGURE 7 - PHASE BOUNDARY AND FISSION GAS
BUBBLE SITES IN THE ELEMENT 432
FUEL ROD, 3 INCHES FROM THE TOP
(9000X)

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SHADOW
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FIGURE 8 ♦ BUBBLE DEPLETION REGION AROUND
LARGE PARTICLES IN THE ELEMENT
432 FUEL ROD, 3 INCHES FROM TOP
(9000X)

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FISSION GAS BUBBLE COUNTING SQUARE

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ANGLE

FIGURE 9 ♦ MICROCRACK IN THE ELEMENT 432
FUEL ROD, 3 INCHES FROM THE TOP
(6000X)

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↑
1μ
↑

63-9V-1

FIGURE 10 ♦ SURFACE FEATURES IN THE
ELEMENT 378 FUEL ROD 3 INCHES
FROM THE TOP (4440X).

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ANGLE

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FISSION GAS BUBBLE COUNTING SQUARE

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↑
1 μ
↑

63-6-1

↑
SHADOW
ANGLE

FIGURE 11 ♦ FISSION GAS BUBBLE DISTRIBUTION
IN THE ELEMENT 519 FUEL ROD
6½ INCHES FROM THE TOP (5513X).

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↑
1μ
↑

↑
SHADOW
ANGLE

FIGURE 12 ♦ FISSION GAS BUBBLE DEPLETION
REGIONS AROUND LARGE PARTICLE
SITES IN THE ELEMENT 519 FUEL
ROD 6½ INCHES FROM THE TOP
(5513X).

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↑
1μ
↑

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ANGLE

FIGURE 13 ♦ PHASE BOUNDARY IN THE ELEMENT
232 FUEL ROD 5-3/4 INCHES
FROM THE TOP (5624X).

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CARBIDE PARTICLE SITES

STRINGER OF PARTICLES LINED UP ALONG
POSSIBLE PHASE BOUNDARY

SURFACE SCRATCH

ALPHA URANIUM OR CARBIDE PARTICLE SITES

↑
1μ
↑

FIGURE 14 ♦ POSSIBLE PHASE BOUNDARY IN THE
ELEMENT 398 FUEL ROD 11 INCHES
FROM THE TOP (6660X).

63-2-2

SHADOW
ANGLE

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↑
1 μ
↑

63-2-3

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SHADOW
ANGLE

FIGURE 15 ♦ SURFACE FEATURES IN THE
ELEMENT 398 FUEL ROD 11 INCHES
FROM THE TOP (6660X).

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↑ 1 μ ↑

→
SHADOW
ANGLE

64-1-2

FIGURE 16 ♦ CARBIDE PARTICLES IN THE
ELEMENT 111 FUEL ROD 11½ INCHES
FROM THE TOP (17,760X).

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↑
1μ
↑

SHADOW
ANGLE

FIGURE 17 ♦ SURFACE FEATURES IN THE ELEMENT
111 FUEL ROD 11½ INCHES FROM
THE TOP (5624X).

64-1-3

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SURFACE SCRATCHES REMAINING FROM SAMPLE
PREPARATION

1 μm

SHADOW
ANGLE

FIGURE 18 - SURFACE FEATURES IN THE ELEMENT
188 FUEL ROD 5½ INCHES FROM THE
TOP (5976X).

64-8-2

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↑
1 μ
↑

64-6-2

FIGURE 19 ♦ TWO PHASE REGION IN THE ELEMENT
332 FUEL ROD 1½ INCHES FROM THE
TOP (14,400X).

↑
SHADOW
ANGLE

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↑ 1 μ ↑
↑
SHADOW
ANGLE

FIGURE 20 ♦ SURFACE FEATURES OF THE ELEMENT
332 FUEL ROD 1½ INCHES FROM THE
TOP (9000X).

64-6-3

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TABLE 2

Electron Microscopy Observations of Irradiated SNAP 8 Fuel Samples

Element No.	Core Location	Axial Location on Rod (Dist. From Top)(in.)	Fuel Rod Diam.		Volume Change		Carbon Fuel Content Burnup		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 (%)	(w/o)	(ma/o)			Micro-cracks	Fission Gas Bubbles	Miscellaneous Observations
IP3,4,5	Unirradiated								$\epsilon +$ microcracks H/Zr = 0.9 to 1.5	Control sample to determine diff. between irradiated & 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	.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 $\frac{\Delta V}{V}$	Yes	Yes	α U sites, carbide particles, ϵ banding, phase boundary with bubble depleted region in the boundary & around C or U particles.
E-332 (weld failure)	VIII-25	1 $\frac{1}{2}$	.5210	-2.15	-5.83	-0.39	0.55	0.10	$\alpha + \delta$ 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 $\frac{1}{2}$	.5332	+0.17	-0.47	+1.00	0.01	0.19	$\delta + \gamma'$ 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	.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 $\frac{1}{2}$	.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 $\frac{1}{2}$	.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 $\frac{3}{4}$	.5376	+1.01	+3.25	+4.02	0.54	0.21	δ phase with banding, H/Zr = 1.56	Region of suspected δ to $\delta + \beta$ transformation, large $\frac{\Delta V}{V}$	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).

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Table 2 (Continued)

Element No.	Core Location	Axial Location on Rod (Dist. From Top)(in.)	Fuel Rod Diam.		Volume Change		Carbon Fuel Content Burnup		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 (%)	(w/o)	(ma/o)			Micro cracks	Fission Gas Bubbles	Miscellaneous Observations
E-519 (cracked)	IV-1	6 $\frac{1}{2}$	.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	.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	.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	.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 $\frac{\Delta V}{V}$	No	Yes	α U sites, ϵ banding, carbide particles, bubble depletion region around large particles, poor replicas.
IS-8 (intact)		2	.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 $\frac{\Delta V}{V}$, large H/Zr loss of -0.37	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.

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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 18. Replication of the fuel surface from element 332 showed the normal particle sites with some microcracking. Typical surface features are shown in Figures 19 and 20. A summary of the data is presented in Table 2.

3. NAA 115-1

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 percent (H_2 corrected). These samples experienced a fuel burnup of 0.41 to 0.43 ma/o. Optical metallographic examination revealed delta plus gamma prime phases in IS-7 and -8 with predominately 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.

Electron microscopy indicated the presence of fission gas bubbles in all three fuel rods; however, microcracks were observed only in IS-8. 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 21 to 23. The degree of microcracking and microtearing shown in Figure 23 indicates what may be the start of incipient gross swelling. A typical phase boundary is shown in Figure 23.

Replication of the fuel surface from IS-7 was generally poor; however, Figure 24 shows some of the surface features from this fuel rod.

Typical surface features of IS-8 are shown in Figures 25 to 28. Very fine banding can be seen in Figure 26. Figure 27 shows what may be some fission gas bubble agglomeration, but this cannot be positively ascertained from the photograph. A typical phase boundary and a gamma prime particle are shown in Figure 28. A summary of the electron microscope observations are tabulated in Table 2.

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FIGURE 21 ♦ MICROCRACKS IN THE IS-4 FUEL
ROD (5513X).

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4

5

6

FISSION GAS BUBBLES

7

8

9

POSSIBLE CARBIDE PARTICLE SITES

10

POSSIBLE PHASE BOUNDARY LINE

ARTIFACT

1 μ

63-5-4

FIGURE 22 • FISSION GAS BUBBLES IN THE IS-4
FUEL ROD (17,020X).

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FISSION GAS BUBBLE COUNTING SQUARE

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1μ

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FIGURE 23 ♦ PHASE BOUNDARY IN THE IS-4
FUEL ROD (5513X) (17020X)

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FIGURE 24 ♦ SURFACE FEATURES OF THE
IS-7 FUEL ROD (6660X)

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FIGURE 25 ♦ CARBIDE PARTICLE IN THE IS-8
FUEL ROD (17,760X).

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FISSION GAS BUBBLE COUNTING SQUARE

↑ 1μ ↑
FIGURE 26 ♦ FINE BANDING IN THE IS-8 FUEL
ROD (14,800X).

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FISSION GAS BUBBLE COUNTING SQUARE

FIGURE 27 ♦ FISSION GAS BUBBLES IN THE
IS-8 FUEL ROD (44,400X).

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FIGURE 28 ♦ PHASE BOUNDARY AND GAMMA PRIME PHASE IN THE IS-8 FUEL ROD (6290X) (14,800X).

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SHADOW ANGLE

SHADOW ANGLE

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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.

4. 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 bubble sites. This void area is thought to be an interaction of the gas bubbles with the larger uranium and carbide particles. These large particles may act as trapping sites for the fission gas bubbles.

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⁽⁸⁾.

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 7 through 28 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. The observed uncorrected volume changes on the two S8ER fuel samples agreed within 25% of that reported from the

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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)
519 (S8ER)	11	1	5.63	3.52 x 10 ¹³
		2	5.11	3.19
		3	3.20	2.00
		4	3.62	2.26
		5	4.28	2.68
	12	1	3.58	2.24
		2	4.52	2.82
		3	3.00	1.88
		4	2.88	1.80
		5	<u>2.63</u>	<u>1.64</u>
		Avg.	3.85	2.40
432 (S8ER)	7	1	2.27	4.33
		2	2.74	5.23
		3	2.38	5.20
		4	3.88	8.50
		5	5.47	11.98
		6	4.05	8.87
		7	7.03	15.38
		8	4.56	9.97
		9	2.82	6.17
		10	2.36	5.17
		11	3.54	7.76
		12	3.02	6.61
	8	1	1.50	7.62
		2	1.48	7.48
	9	1	1.31	3.55
		2	1.47	4.01
		3	<u>1.65</u>	<u>4.48</u>
		Avg.	3.03	7.19

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Table 3 (Continued)

Element No.	Figure No.	Counting Square No.	Bubble Volume Fraction (%)	Bubble Density (bubbles/cc)(x 10 ¹³)
IS-4 (115-1)	Not reproduced	1	0.63	1.21
		2	0.63	1.21
		3	0.47	0.89
		4	0.88	1.69
		5	0.19	0.36
	22	1	1.40	1.92
		2	1.78	2.44
		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
		8	2.00	2.75
		9	0.81	1.11
		10	0.98	1.34
	23	1	0.84	1.24
		2	0.78	1.16
		3	0.71	1.06
		4	0.97	1.43
		5	0.96	1.43
		6	0.91	1.47
		7	1.07	1.73
		8	0.91	1.47
		9	1.13	1.82
		10	1.44	2.32
		11	1.27	2.04
		Avg.	1.13	1.69
IS-7 (115-1)	24	1	4.47	0.92
		2	9.09	1.87
		Avg.	6.78	1.39

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Table 3 (Continued)

Element No.	Figure No.	Counting Square No.	Bubble Volume Fraction (%)	Bubble Density ¹³ (bubbles/cc)(x 10 ¹³)
IS-8 (115-1)	26	1	0.35	4.22
		2	0.43	5.27
		3	0.57	6.86
		4	0.43	5.22
	Not reproduced	1	0.41	3.31
		2	0.38	3.07
		3	0.36	2.90
		4	0.49	3.93
		5	0.53	4.26
	27	1	0.11	1.58
		2	0.22	3.26
		3	0.25	3.65
		4	0.52	7.68
		5	0.36	5.32
		6	0.36	5.32
		7	0.39	5.77
		8	0.25	3.65
	28	1	0.68	3.18
		2	0.78	3.64
		3	0.98	4.54
		4	1.00	4.65
		5	0.61	2.85
		Avg.	0.47	4.27

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fission gas bubble measurements. This indicated the bulk of observed volumetric changes in these elements was primarily the result of fission gas formation. The radial fuel burnup profiles in the S8ER fuel rods were relatively flat and uniform⁽¹⁾. Hence, the location of the replica on the fuel surface should be representative of fuel surface regardless of its actual location. The void areas in the S8ER fuel due to fission gas bubble formation was 3.85 and 3.03 percent for the samples from elements 519, and 432, respectively. Microcracks were observed in the fuel from element 432. An additional contribution to fuel swelling from the microcracks would bring the 3.03% volume change due to bubble formation closer to the observed volume change of 3.94% in this fuel sample.

TABLE 4

Volume Change Due to Fission Gas Bubble Formation

Element No.	Fuel $\Delta V/V$ (%)	Fuel $\Delta V/V$ (%) (H_2 Corrected)	$\Delta V/V$ Due to Fission Gas Bubble Formation (%)
519	3.72	4.58	3.85
432	3.94	4.25	3.03
IS-4	4.16	4.30	1.13
IS-7	2.94	4.11	6.78*
IS-8	0.74	2.08	0.47

* See test for qualification of this data.

A poorer correlation was noted of the observed volume changes for the NAA 115-1 fuel samples to the volume change contribution due to fission gas bubble formation. These fuel samples did not experience a uniform radial burnup profile⁽⁷⁾. The actual location of the replica photographs on the surface of the fuel cross-section is not known. Photographs of representative fission gas bubble sites over the surface of the fuel cross-section would be required for proper interpretation of the bubble volume contribution.

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 24. It can be seen from the figure that 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.

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Bubble formation in IS-8 accounted for 0.4% volume change compared to the observed volume change of 0.74%. It is interesting to note that the bubble density in IS-8 (as reported in Table 3) is over $2\frac{1}{2}$ times that reported for IS-4 or IS-7. This is to be expected since a greater bubble dispersion will result in restrained fuel swelling as evidenced by the low volume change in IS-8 compared to IS-4 and IS-7. A higher bubble density at the same burnup would indicate greater surface tension and higher gas pressure within the bubbles. For this phenomena to occur, more precipitation and particle sites were probably present in the fuel to allow additional bubble nucleation sites. The largest discrepancy between the two volume changes was noted in IS-4 where the volume contribution due to bubble formation was only 27% of the observed change.

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 is listed in Table 5. 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.

TABLE 5

Bubble Depletion Zone Width Measurements
About Uranium and Carbide Particle Sites

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

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An average bubble diameter was calculated to 0.90×10^{-5} mm (900°A) with a range from 0.51 to 1.45×10^{-5} mm in diameter. A few measurements were taken of the distance between bubbles. This 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 studies reported by Taketani⁽⁶⁾ in which he reported uranium precipitate densities ranging from 3 to 5×10^{10} particles per cc.

D. CONCLUSIONS

The ability to replicate remotely irradiated U-ZrH fuel has been demonstrated. Replicas could be satisfactorily decontaminated, shadowed, and photographed under the electron microscope. Distinguishing features, such as uranium sites, carbide particle sites, phase boundaries, grain boundaries, banding, microcracks, and fission gas bubbles were identified. In addition, fission gas bubble resolution was good on some samples to allow a quantitative count of the bubbles per unit volume.

Microcracking in the fuel did not appear to be a major contributor to fuel swelling in the peak distortion areas of the fuel rods based on the replicas evaluated. In the S8ER fuel samples replicated, fission gas bubble formation was the primary cause for the observed volume changes in elements 432 and 519 in the peak fuel rod distortion areas. Sizable contributions of bubble formation to fuel swelling in the NAA 115-1 fuel samples was also observed. Fission gas bubble density measurements on the order of 10^{13} bubbles/cc were typical. The average diameter of these bubbles were 900°A with a mean distance between bubbles of 2.12×10^{-4} mm.

If the size of gas bubbles remains small, approximately 400°A or less, the surface tension of the matrix permits retention of the gas at high pressures and swelling is greatly reduced.

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However, if the small bubbles agglomerate into larger bubbles, serious swelling results⁽⁸⁾. The average size of the bubbles in the S8ER and NAA 115 fuel were approximately twice the 400°A size.

Surface tension forces have a smaller significance with larger bubbles. When high excess pressures are built up, the matrix can deform plastically by dislocation generation. Reduction of creep strength by thermal cycling can allow enhanced bubble growth. Changes in structural properties can affect fuel swelling which is very sensitive to bubble spacing. Large swelling has been observed in irradiated fuel which experienced thermal cycling at low burnup⁽⁹⁾.

Other studies⁽⁹⁾ on irradiated fuel at a burnup of approximately 0.5 ma/o resulted in a volume change slightly less than 5%. Electron microscopy of the fuel revealed bubbles $\sim 10^{-5}$ cm in diameter with a spacing of $\sim 5 \times 10^{-5}$ cm. By comparison, the S8ER and NAA 115-1 fuel replicas revealed the same order of magnitude of bubble diameter, but the spacing was half as great.

In summation, it appeared that comparing the contribution to swelling from fission gas bubbles in S8ER fuel from peak distortion areas with other studies, indicated greater than predicted bubble formation at a burnup of ~ 0.2 ma/o. Thermal transients and external loading on the fuel rod^(1,2) may have contributed significantly to the observed additional fission gas agglomeration. Other fuel samples examined from S8ER fuel not in areas of peak distortion revealed no fission gas bubbles at the magnification employed. The bubbles may have been too finely dispersed in the matrix and of too small a diameter to be observed.

The NAA 115-1 fuel samples examined exhibited bubble formations comparable to other studies^(9,10,11) at the burnup levels for this fuel.

Bubble depletion regions 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⁽¹²⁾ et al. in their work on fission gas bubble studies in U-Mo alloys.

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There was no evidence of a correlation in either the S8ER or NAA 115-1 replicas of epsilon banding with the location of fission gas bubbles, particle sites, precipitates, or micro-cracks.

E. RECOMMENDATIONS FOR FUTURE WORK

Many SNAP fuel experiments, NAA 115-1 included, are capsule irradiations which are inserted into a large reactor such as Hanford for irradiation. The fuel in these capsules do not generally receive a uniform burnup due to the geometry of the core. Evaluation of the electron microscope photographs for fission gas bubble formation is dependent on burnup. Hence, it is important to identify the location of the photographs on the wafer cross-section. Multiple replicas on a fuel cross-section of non-uniform burnup should allow a correlation of bubble formation with burnup.

Fuel surfaces should be marked in the areas of interest prior to replication. When the replicas are transmitted for analysis, every effort should be made to obtain that portion of the replica in the area of interest. If this is not possible due to poor replication or other reasons, replicas which are taken should be identified as to location on the fuel cross-section.

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