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Examination of Irradiated EBWR Core-I Fuel Elements

by

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EXAMINATION OF IRRADIATED EBWR CORE-I FUEL ELEMENTS

by

C. F. Reinke and R. Carlander

ABSTRACT

Two fuel elements were removed from the Experimental Boiling Water Reactor at Argonne National Laboratory and examined in a hot cell. The elements had maximum burnups of 0.11 a/o and 0.39 a/o, respectively. Both were disassembled and sampled for the evaluation of the effects of in-pile operation and radiation damage to the fuel. The fuel elements were in good condition with no ruptured cladding, core-clad nonbonds, or excessive fuel plate swelling or warpage. Thin samples cut from the fuel plates in element ET-51 warped and cracked, suggesting a relieving of locked-in stresses and indicating that after 0.39 a/o burnup the fuel cores were hard, brittle and highly stressed. The rate of fuel plate volume increase due to the burnup of uranium was 6 to 7% ΔV per a/o burnup. Hydrogen was picked up by the fuel plates under reactor operating conditions with the probable formation of isolated areas of small amounts of zirconium hydride. Annealing studies on sections of fuel plate at 500°C and 550°C indicated bulk volume increases of 1 to 2% and 5 to 10%, respectively, after 500 hours. A 600°C anneal resulted in a bulk volume increase of 17% after 45 hours.

INTRODUCTION

The satisfactory operation of any reactor is intimately associated with the performance of its fuel. Irradiation experiments which are used to evaluate potential reactor fuels are difficult to perform, expensive, time-consuming, and do not always duplicate completely the actual reactor operating conditions. Often a choice of fuel must be made on the basis of incomplete information. Therefore, the examination of fuel elements from a reactor provides useful information on their performance which could not be obtained in any other way, and corroborates and expands on the initial information which formed the basis for the choice of fuel for the reactor. With these facts in mind the effects of in-pile operation and radiation damage on EBWR Core-I fuel were determined and are discussed in this report.

The Experimental Boiling Water Reactor (1) is a direct cycle boiling reactor which has been operating since December 1956, during which time power levels up to three times the design value of 20 Mw were attained. The reactor is cooled by boiling heat transfer to demineralized water, having a pH of 7, which circulates through the core by natural convection.

The reactor fuel is a mixture of natural and slightly enriched U-5 w/o Zr-1.5 w/o Nb alloy clad with Zircaloy-2. One of the reasons for choosing this alloy was the aqueous corrosion resistance afforded by the presence of the zirconium and niobium.(2) To obtain the maximum benefit from the alloying additions a suitable heat treatment had to be employed. However, a heat treatment which would produce both dimensional stability and corrosion resistance was not available.(3) Since dimensional stability under irradiation was judged more important than corrosion resistance, the heat treatment producing dimensional stability was chosen. This consisted of a gamma soak at 825°C, an isothermal quench and holding at 640°C for 23 hours, followed by air cooling. The Zircaloy-2 cladding was metallurgically bonded to the fuel core by roll cladding using the plate and frame process.(4,5) The finished fuel plates were assembled into a fixture with Zircaloy-2 side plates and spot welded into completed fuel elements.

Under initial reactor operating conditions at 20 Mw, the average fuel plate surface temperature was 260°C, with a corresponding centerline fuel core temperature of 280°C. With normal buildup of surface scale on the fuel plates during reactor operation, calculations indicate the maximum fuel temperature may reach 410°C or higher.

During the course of an examination which covered the period from June 1958 to November 1959, two fuel elements were disassembled and sampled. They were fuel elements T-23 and ET-51, which were in the reactor for a period of 1 year and 2½ years, respectively. The fuel plates in element T-23, which had a maximum burnup of 0.11 a/o, were composed of 0.170 inch of natural U-5 w/o Zr-1.5 w/o Nb alloy clad with 0.020 inch of Zircaloy-2. Element ET-51, which had a maximum burnup of 0.39 a/o, was identical to T-23, with the exception that the uranium used was 1.44% enriched instead of natural. The procedures used during the handling and disassembly have been described previously.(6) The details of the examination, which included visual inspections, dimensional measurements, gamma-activity scans, ultrasonic inspections, metallographic examinations, micro-hardness measurements, corrosion tests, analyses for hydrogen pickup and hydride formation, and annealing studies are discussed in the following sections of the report.

RESULTS AND DISCUSSION

Visual Examination

The two EBWR fuel elements were first examined visually through the windows of a hot cell. The elements appeared to be in good condition with no excessive fuel plate warpage or swelling. Figures 1, 2 and 3 show the fuel elements in the as-received condition. The Zircaloy-2 side plates and stainless steel end fittings were attached firmly; also, there was no indication of preferential corrosion at the spot weld or rivet areas.

A considerable quantity of surface scale was observed on the fuel plates of both elements; however, in the element with the longest exposure (ET-51) extensive portions of this scale were not adherent. Figures 3, 4 and 5 illustrate the condition of the scale on these plates. X-ray diffraction and spectrochemical analyses of the scale revealed aluminum as the major metallic constituent. Significant quantities of nickel and iron were present also. Based on these analyses, boehmite ($\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$) was one constituent of the scale. A variation in the thickness of the deposited scale resulted in a gradation in shading along the length of the plates, as can be seen in Figures 1 and 5. The lighter end in the figures is the lower end or high-flux end.

106-3932

Figure 1. EBWR Fuel Element T-23 in the As-received Condition

106-3934

Figure 2. Lower End of EBWR Fuel Element T-23 in the As-received Condition

25450

Figure 3. Lower End of EBWR Fuel Element ET-51 in the As-received Condition

25461

0.4X

Figure 4. Surface Scale on Fuel Plate E828 from Element ET-51

25449

0.12X

Figure 5. Surface Scale on Fuel Plate E803 from Element ET-51

Physical Dimensions

The fuel plates in elements T-23 and ET-51 exhibited good dimensional stability under EBWR reactor operating conditions.

Pre-irradiation measurements of width were made at a minimum of three random locations along the length of each plate, excluding the last six inches at each end. On the basis of these measurements and the post-irradiation values, which were accurate to $\pm \frac{1}{32}$ inch, there was no change in either the width or length of any of the plates checked.

Table I lists a comparison between the pre- and postirradiation measurements of thickness along the centerline of the plates. Since the pre-irradiation values were not measured at specific areas but at random locations along the length of each plate, and since all these values were within the reference design specifications of 0.214 ± 0.004 inch, a value of 0.214 inch was used as a basis for comparison with the postirradiation values.

The thickness of plate 606 from fuel element T-23 increased by a consistent 0.004 to 0.005-inch maximum along the bottom half of the plate.

TABLE I

Thickness Measurements^(a) on Fuel Plates from Fuel Elements T23 and ET51

Fuel Element		T23		ET51		ET51		ET51		ET51		ET51		ET51	
Fuel Plate		606		E 813		E 828		E 829		E 825		E 821		E 803	
Loca- tion ^(b)	Pre- irradiation Thickness ^(c)	Postirra- diation Thickness	Δ ^(d)	Postirra- diation Thickness	Δ	Postirra- diation Thickness	Δ	Postirra- diation Thickness	Δ	Postirra- diation Thickness	Δ	Postirra- diation Thickness	Δ	Postirra- diation Thickness	Δ
48	0.214	0.212	-0.002	0.216	0.002	0.212	-0.002	0.213	-0.001	0.212	-0.002	0.213	-0.001	0.219	0.005
45	0.214	0.220	0.006	0.216	0.002	0.214	0.000	0.217	0.003	0.217	0.003	0.217	0.003	0.219	0.005
42	0.214	0.215	0.001	0.216	0.002	0.216	0.002	0.217	0.003	0.217	0.003	0.219	0.005	0.218	0.004
39	0.214	0.214	0.000	0.217	0.003	0.217	0.003	0.217	0.003	0.218	0.004	0.219	0.005	0.221	0.007
36	0.214	-	-	0.219	0.005	0.217	0.003	0.219	0.005	0.218	0.004	0.223	0.009	0.218	0.004
33	0.214	0.215	0.001	0.219	0.005	0.218	0.004	0.223	0.009	0.221	0.007	0.223	0.009	0.221	0.007
30	0.214	0.215	0.001	0.222	0.008	0.220	0.006	0.225	0.011	0.224	0.010	0.226	0.012	0.223	0.009
27	0.214	0.216	0.002	0.220	0.006	0.220	0.006	0.225	0.011	0.217	0.003	0.227	0.013	0.223	0.009
24	0.214	0.218	0.004	0.217	0.003	0.218	0.004	0.223	0.009	0.217	0.003	0.227	0.013	0.223	0.009
21	0.214	0.219	0.005	0.217	0.003	0.216	0.002	0.223	0.009	0.218	0.004	0.225	0.011	0.218	0.004
18	0.214	0.218	0.004	0.219	0.005	0.216	0.002	0.223	0.009	0.218	0.004	0.224	0.010	0.218	0.004
15	0.214	0.218	0.004	0.219	0.005	0.216	0.002	0.223	0.009	0.218	0.004	0.224	0.010	0.219	0.005
12	0.214	0.218	0.004	0.221	0.007	0.216	0.002	0.223	0.009	0.219	0.005	0.228	0.014	0.221	0.007
9	0.214	0.218	0.004	0.225	0.011	0.218	0.004	0.220	0.006	0.219	0.005	0.227	0.013	0.220	0.006
6	0.214	0.212	0.002	0.222	0.008	0.216	0.002	0.216	0.002	0.216	0.002	0.215	0.001	0.216	0.002
Condition		No descaling		Descaling on one face 10 to 27 inches from the bottom		Descaling on both faces 10 to 25 inches from the bottom		No descaling		Descaling on one face 10 to 30 inches from the bottom		No descaling		Descaling on both faces 10 to 33 inches from the bottom	

(a) All measurements are in inches and were taken along the centerline of each plate.

(b) The distances are in inches from the bottom end of each plate.

(c) The pre-irradiation thicknesses at the center of the plates averaged 0.214 in. and were measured at a minimum of four places, excluding the first and last 6 in. of each plate. The reference thickness on each plate was 0.214 ± 0.004 in.(d) All postirradiation thickness measurements are ± 0.001 in.

This increase was due to the sum of any volume increase plus scale deposition during in-pile operation. It is significant to note that even with this increase the postirradiation dimensions fell within the specifications for the unirradiated plate. The plate had a maximum burnup of 0.11 a/o.

Thickness measurements on the plates from fuel element ET-51, which had a maximum burnup of 0.39 a/o, included an unknown thickness of scale. The thickness is unknown because (1) areas were present at which partial descaling occurred and (2) the plates were wire-brushed prior to the ultrasonic examination which preceded the dimensional measurements. Measurements on scale which had spalled from the fuel plate surfaces indicated a thickness of about 0.005 inch. It is estimated that the thickness of the plates in fuel element ET-51 increased by a consistent 0.010 to 0.015-inch maximum along the bottom half of each plate. Since 0.010 inch of this increase was attributed to surface scale deposit during operation, the remaining 0 to 0.005 inch was due to the volume increase accompanying the burnup of the uranium. On the basis of the 0.005-inch increase in thickness, if the length and width continue to remain constant, a volume increase of about 6 to 7% per a/o burnup can be expected due to burnup alone under normal EBWR operating conditions.

The postirradiation channel gap measurements on fuel element ET-51 are tabulated in Table II. The values varied widely and differed considerably from the reference design specification of 0.438 ± 0.005 inch. The measurements, which were reproducible to ± 0.002 inch, indicated considerable variation in channel gap along the length and width of each channel. As might be expected, the minimum spacing in a channel usually occurred midway between the side plates. On the basis of the visual examination and dimensional measurements on the individual plates, it was felt that the variations in the present channel gaps were not due primarily to in-pile operation. It is believed that essentially the existing channel gaps were present initially.

Gamma-activity Scan

Specimens were removed from the plates for metallographic examinations, microhardness measurements, corrosion tests, hydrogen analyses, and postirradiation annealing studies. It was obvious that burnup levels were a prime consideration in interpreting the results of the various tests. Therefore, before sectioning the plates, plots of the gamma-activity level along the length of each plate were made, and these plots, in conjunction with a radiochemical burnup analysis on a sample taken from the area of maximum gamma activity, were to be used to determine the specimen burnup values. The equipment used to measure the gamma-activity level along the length of each plate is shown in Figures 6 and 7. Figures 8 and 9 are plots of the relative gamma activities along the lengths of the plates in fuel elements T-23 and ET-51, respectively. The curves were based on a scan of an entire plate. Similar plots based on radiochemical analyses of burnup are included for comparison. It can be seen readily that significant differences

TABLE II

Channel Gap Measurements on Fuel Element ET-51^(a)

Distance from Bottom of Fuel Plates, in.	Channel 813/828			Channel 828/829			Channel 829/825			Channel 825/821			Channel 821/803		
	1 in. from Edge	Center of Channel	1 in. from Edge	1 in. from Edge	Center of Channel	1 in. from Edge	1 in. from Edge	Center of Channel	1 in. from Edge	1 in. from Edge	Center of Channel	1 in. from Edge	1 in. from Edge	Center of Channel	1 in. from Edge
48.5	0.443	0.420	0.454	0.461	0.407	0.462	0.397	0.373	0.400	0.483	0.446	0.486	0.425	0.410	0.422
45.5		0.437			0.448			0.368			0.462			0.412	
42.5		0.439			0.456			0.344			0.481			0.413	
39.5		0.433			0.466			0.345			0.466			0.411	
36.5		0.431			0.487			0.347			0.455			0.410	
33.5		0.428			0.454			0.358			0.476			0.403	
31.5		0.422			0.448			0.366			0.464			0.392	
30.5		0.421			0.451			0.363			0.480			0.392	
27.5		0.415			0.455			0.364			0.452			0.395	
24.5		0.423			0.465			0.345			0.454			0.395	
21.5		0.428			0.477		0.363	0.336			0.473			0.394	
18.5	0.439	0.416	0.435	0.471	0.485	0.477	0.361	0.337	0.364	0.473	0.471	0.481	0.404	0.396	0.405
15.5		0.415		0.468	0.489			0.339			0.475			0.397	
12.5		0.426			0.461			0.351			0.479			0.394	
9.5		0.425			0.453			0.354			0.469			0.393	
6.5		0.421			0.471			0.361			0.470			0.397	
5.5	0.439	0.443	0.441	0.460	0.408	0.458	0.385	0.362	0.388	0.470	0.432	0.483	0.411	0.401	0.413

(a) Reference design spacing is 0.438 ± 0.005 in. The postirradiation measurements were reproducible to within ± 0.002 in.

27557

Figure 6. Equipment Used to Obtain Gamma-activity Scan of EBWR Fuel Plate

Figure 7
Equipment for Gamma-activity Scanning with EBWR Fuel Plate in Position in Cave

106-3967

Figure 8. Variation in Gamma Activity and Burnup along the Length of Fuel Plate 606 in Element T-23

Figure 9. Variation in Gamma Activity and Burnup along the Length of Fuel Plates in Element ET-51

exist. It is believed that the curves based on radiochemical analyses represent more accurately the variation in activity level and burnup along the length of a plate. A plot of the gamma activity level along the length of plate 606 from fuel element T-23 based on the gamma activity of small samples⁽⁷⁾ removed from plate 606, verified the curve based on the burnup analyses. In scanning an entire plate it proved impossible to lower the high background level due to the remainder of the plate. This background affected the lower activity readings to a greater extent and thus tended to flatten the curve.

The results of radiochemical analyses for burnup on examined plates are tabulated in Table III.

Ultrasonic Inspection

After separating the fuel elements into individual plates, the integrity of the core-to-clad bonds was determined by an ultrasonic inspection. The details of the method have been previously described.⁽⁸⁾

TABLE III

Burnup Analyses on Examined Plates

Fuel Element	Plate	Distance From Bottom of Fuel Plates, in.	Location of Sample	Burnup ^(a)	
				a/o ^(b)	MWD/ton U
T-23	606	3	Width of Plate	0.065	650
T-23	606	18½	Width of Plate	0.11	1100
T-23	606	36½	Width of Plate	0.030	300
T-23	606	51	Width of Plate	0.005	50
ET-51	E828	6½	Center of Plate	0.23	2300
ET-51	E828	21½	Edge of Plate	0.39	3900
ET-51	E828	21½	Center of Plate	0.33	3300
ET-51	E828	30½	Center of Plate	0.25	2500
ET-51	E828	36½	Center of Plate	0.16	1600
ET-51	E828	42½	Center of Plate	0.094	940
ET-51	E828	48½	Center of Plate	0.20	2000
ET-51	E821	21½	Center of Plate	0.32	3200
ET-51	E803	23	Center of Plate	0.32	3200
ET-51	E813	22½	Center of Plate	0.32	3200
ET-51	E825	22½	Center of Plate	0.31	3100
ET-51	E829	25½	Center of Plate	0.30	3000

(a) The values are based on radiochemical analyses for Cs¹³⁷.

(b) a/o burnup is the ratio of the number of fissions to the total number of atoms present in the core times 100.

An ultrasonic inspection consisted of a scan across the width of each plate and along its length. These two-dimensional recordings of the irradiated plates when compared with pre-irradiation scans indicated that the cladding was firmly adherent to the core on all the plates tested.⁽⁹⁾

The initial scan of plate 606 from fuel element T-23 disclosed an area of poor ultrasonic transmission which coincided with a deposit of scale. The plate was removed from the scanner and wire brushed, after which it was scanned again. The second scan did not reveal any abnormalities. It is believed that the poor ultrasonic transmission observed in the first scan was due to a loosely adherent or porous scale deposit.

The ultrasonic scan of each plate also gave an indication of the condition of the core. Since porosity or cracks would be observed in an area of poor ultrasonic transmission and since such areas were not found, it is believed that no large cracks were present in the fuel plate cores.

Metallographic Examination

Samples were milled from the fuel plates of both elements with a slitting saw. Metallographic specimens were cut from these samples by electrical discharge machining and cold mounted using Hysol 6040 compound.

Thin samples cut from the fuel plates in element ET-51 warped and cracked, suggesting a relieving of locked-in stresses and indicating that after 0.39 a/o burnup the fuel cores were in a brittle, highly stressed condition. Figures 10, 11 and 12 indicate the nature and extent of the cracking observed on $\frac{1}{16}$ -inch thick samples cut from the plates. It is significant to note that the cladding was still quite ductile.

25447

1X

Figure 10. Warped and Cracked $\frac{1}{16}$ -inch Thick Specimen. Typical Condition of Thin Samples Cut from Fuel Plates in Element ET-51.

25732

4X

Figure 11. Cracks in $\frac{1}{16}$ -in Thick Specimen. Typical Condition of Thin Samples Cut from Fuel Plates in Element ET-51

25733

4X

Figure 12. Cracks in $\frac{1}{16}$ -inch Thick Specimen. Typical Condition of Thin Samples Cut from Fuel Plates in Element ET-51.

The metallographic examination of 40 samples cut from various locations throughout fuel element ET-51 revealed cracks in fuel core, with the most extensive cracks occurring at areas adjacent to the side-plate spot welds. The specimen shown in Figure 13 is typical of the samples examined. An examination of the specimen revealed that some of the cracks appeared to have originated at a surface cut by electrical discharge machining. In comparison, the metallographic examination of samples taken from plate 606 of fuel element T-23 (0.11 a/o burnup) revealed cracks only at areas adjacent to the spot welds. There is no doubt that samples from ET-51 were cracked during specimen preparation. However, it cannot be determined from the results whether microcracks did or did not exist in both fuel elements prior to the cutting operation. In no instance in either element did the cracks propagate into the cladding. They stopped at the core-to-clad interface or at the edge of the diffusion zone, as shown in Figure 14.

26554

4X

Figure 13. Cracks in a Typical Metallographic Specimen from Element ET-51

Figure 14. Propagation of Crack through Diffusion Zone at Core-clad Interface

As originally fabricated, finished fuel plates were heated to 825°C, isothermally quenched and held at 640°C for 23 hours, followed by air cooling.⁽⁵⁾ This heat treatment imparted dimensional stability to the fuel. Following the heat treatment, the fuel plates were assembled into a fixture with Zircaloy-2 side plates and spot welded into a completed fuel element. From the postirradiation metallographic examination, it appears that during the welding operation the areas adjacent to the spot welds received a heat treatment in which a small portion of the fuel at the edge of the core was transformed to gamma and then quenched. This quenched structure can be expected to have an irradiation growth rate, G_i , of about 50 times that of the isothermally transformed material.⁽³⁾

This difference in growth rates combined with the higher burnups at the edge of the plates is believed to account for the more extensive cracking found at the spot-weld areas. The results indicate that the core is in a brittle, highly stressed, crack-sensitive condition.

On all the specimens examined the bonds between the core and the clad at areas removed from cracks were in excellent condition.

Typical core structures at various burnup levels are shown in Figure 15. An electrolytic etch containing 5% citric acid and 0.5% nitric acid was used to develop the structure.

As can be seen in Figure 15, the microstructure deteriorates as the burnup increases; after approximately 0.30 a/o burnup, there is little evidence of the original structure present. The original structure is believed to be one of alpha and metastable gamma.⁽¹⁰⁾ The material which appears in the photomicrographs as white particles is thought to be inclusions.

(a)

25269

0.0 a/o

(b)

24909

0.01 a/o

(c)

24911

0.03 a/o

(d)

24883

0.09 a/o

Figure 15. Structure of U-5 w/o Zr-1.5 w/o Nb Core Alloy as a Function of Burnup. All Photos are Bright Field at 250X Mag.

(e)

26553

0.17 a/o

(f)

26552

0.26 a/o

(g)

26551

0.30 a/o

(h)

26556

0.32 a/o

Figure 15 (cont'd.)

Microhardness Measurements

Microhardness measurements were made on the irradiated U-5 w/o Zr-1.5 w/o Nb fuel alloy and on the Zircaloy-2 cladding. A Tukon hardness tester was used with a Knoop indenter and a 500-gram load. The values are tabulated in Tables IV and V, in which are listed the ranges of the readings observed in a given area as a function of burnup or integrated neutron flux above 1 Mev. Figures 16 and 17 are plots of the mean values obtained on the core and cladding at the center of the plates. The curves indicate an increase in hardness with increasing burnup or neutron flux over the range covered.

TABLE IV

Microhardness Measurements on
U-5 w/o Zr-1.5 w/o Nb Alloy

Burnup, a/o	Knoop Hardness Number	
	Center of Plate	Transformed Zone
0.0	334	463-486
0.009	300-374	533-563
0.017	363-401	557-588
0.024	349-392	528-610
0.088	348-390	511-548
0.11	414-455	506-538
0.17	515-534	-
0.26	534-577	-
0.29	534-549	-
0.32	549-618	-
0.33	-	657-668
0.38	-	653-657

TABLE V

Microhardness Measurements
on Zircaloy Cladding

nvt fast ^(a) $\times 10^{-20}$	Knoop Hardness Number	
	Center of Plate	Edge of Plate
0	187-188	176-182
0.3	177-196	193-201
0.9	164-193	193-227
1.7	191-197	208-235
2.2	190-203	214-232
8.4	218-224	-
8.7	183-222	210-246
10	185-208	237-258
13	209-223	-
14	211-232	-
16	252-271	269-283

(a) Calculated integrated neutron flux above 1 Mev.

The measurements on the U-5 w/o Zr-1.5 w/o Nb core material were made at two general locations: near the center of the fuel plates, and adjacent to the side-plate spot welds. Prior to irradiation, the core material adjacent to the spot welds had a hardness 125 to 150 KHN points higher than the remainder of the core. This difference was due to the heat treatment given the core at these areas during the final assembly welding. After an irradiation to burnups on the order of 0.3 to 0.4 a/o, the hardness increased approximately 250 KHN points in the core not affected by the welding operation and 200 KHN points in the areas adjacent to the spot

welds. The Knoop hardness values after irradiation to 0.3 to 0.4 a/o burn-up correspond approximately to Rockwell C hardness values in the low to middle 50's, respectively. Thus the irradiated fuel alloy is quite hard.

Figure 16. Mean Microhardness Readings versus Burnup for U-5 w/o Zr-1.5 w/o Nb Core Alloy

Figure 17. Mean Microhardness Readings versus Integrated Fast Flux for Zircaloy-2 Cladding

Measurements were made on the Zircaloy-2 cladding at both the center and edge of the plates. The values are tabulated in Table V as a function of calculated integrated neutron flux above 1 Mev. In general, the hardness increased as the exposure increased, and after a calculated integrated exposure of about 1.6×10^{21} nvt fast was 80 to 100 KHN points harder than the unirradiated material. For a given level of exposure the edges of the plates were slightly harder than the other areas. This is probably due to lower temperatures at these areas under reactor operating conditions, and the tendency to form hydrides as is evidenced by the results of the hydrogen analyses. For a calculated exposure of 1.6×10^{21} nvt fast the Knoop values correspond approximately to Rockwell C hardness values in the low to middle 20's.

Corrosion Tests

Samples of irradiated fuel core were removed from the plates for corrosion tests in water at 260°C. As noted earlier, the fuel alloy in EBWR was given a heat treatment for maximum dimensional stability rather than for improved corrosion resistance. Over the burnup range of 0.005 to 0.09 a/o, the results of the tests indicate a marked improvement in the

corrosion resistance of the fuel with irradiation.⁽¹¹⁾ The data are tabulated in Table VI.

TABLE VI

Corrosion of Irradiated
U-5 w/o Zr-1.5 w/o Nb
Alloy in Water at 260 to
270°C as a Function of
Burnup⁽¹¹⁾

Burnup, a/o	Corrosion Rate, mg/(cm ²)(day)
0.000	9470
0.005	2500
0.009	2780
0.017	2310
0.024	1880
0.088	1890
0.11	5160

The tabulated rates are based on original exposed area. True rates would therefore be higher, but the quoted rates are valid for purposes of comparison. At present there is no positive explanation for this improvement in corrosion resistance. Thermal spikes associated with the fission product atoms may influence in part the improvement in corrosion rates. The effect may be one of changing the structure from the dimensionally stable, isothermally transformed structure to the corrosion-resistant quenched gamma, with the effect of additional burnup being similar to overaging, which would lower the improved corrosion resistance. In support of this hypothesis, the hardness

values obtained on the irradiated core are comparable to those obtained on unirradiated material by quenching from the gamma phase.

Hydrogen Analyses

Samples were removed from both fuel elements for analyses of their hydrogen contents. Samples were taken from the edges and center of the plates along the plate length. The specimens from the edges of the plates were solid Zircaloy-2, while those from the center contained 0.170 inch of fuel core clad on two sides with 0.020 inch of Zircaloy-2. The results are listed in Tables VII and VIII.

TABLE VII

Hydrogen Analyses on Plate 606 from Fuel Element T-23

Distance from Bottom of Plate, in.	Sample No.	Hydrogen Content, ppm,		Distance from Bottom of Plate, in.	Sample No.	Hydrogen Content, ppm,	
		Edge of Plate	Center of Plate			Edge of Plate	Center of Plate
1 $\frac{1}{2}$	1, 3, 2	31, 94	76 ^(a)	26 $\frac{3}{8}$	25, 27, 26	49, 105	16
5	4, 6, 5	92, 89	11	29 $\frac{3}{8}$	28, 30, 29	53, 100	14
8	7, 9, 8	65, 73	12	32 $\frac{3}{8}$	31, 33, 32	47, 70	16
11	10, 12, 11	58, 65	14	38 $\frac{3}{8}$	34, 36, 35	63, 61	13
14	13, 15, 14	53, 63	15	41 $\frac{5}{8}$	37, 39, 38	58, 74	12
17	16, 18, 17	50, 48	14	44 $\frac{3}{8}$	40, 42, 41	73, 55	11
20	19, 21, 20	88, 49	14	47 $\frac{5}{8}$	43, 45, 44	60, 56	16
23	22, 24, 23	55, 85	21	52 $\frac{1}{2}$	46, 48, 47	65, 72	48 ^(a)

(a) This sample was solid Zircaloy-2.

TABLE VIII

Hydrogen Analyses on Specimens from Fuel Element ET-51

Fuel Element	Distance from Bottom of Plate, in.	Sample No.	Hydrogen Content, ppm	
			Edge of Plate	Center of Plate
E828	6	1, 3, 2	34, 33	14
E828	12	4, 6, 5	26, 69	30
E828	21	7, 9, 8	33, 33	18
E828	30	10, 12, 11	38, 44	26
E828	36	13, 15, 14	37, 31	15
E828	42	16, 18, 17	44, 31	13
E828	48	19, 21, 20	40, 43	11
E821	9	22, 24, 23	55, 43	29
E821	21	25, 27, 26	39, 44	41
E821	39	28, 30, 29	59, 41	12
E803	11	31, 33, 32	73, 51	137(a)
E803	22	34, 36, 35	76, 54	22
E803	43	37, 39, 38	49, 46	12
E829	13	40, 42, 41	68, 52	20
E829	25	43, 45, 44	58, 72	21
E829	49	46, 48, 47	54, 44	15
E813	11	52, 54, 53	52, 34	18
E813	22	49, 51, 50	47, 37	--
E825	7	55, 57, 56	41, 50	33
E825	22	58, 60, 59	47, 38	19
E825	38	61, 63, 62	46, 46	14

(a) This value is questionable and was not used in computing the average value or standard deviation.

Results obtained on plate 606 from fuel element T-23 indicate a higher hydrogen concentration at the edge than at the center of the plate. The values ranged from 31 to 105 ppm at the edge of the plate with an average value of 66 ppm and a standard deviation of ± 17 . In comparison, the values at the center of the plate ranged from 11 to 21 ppm with an average value of 14 ppm and a standard deviation of ± 3 .

Results obtained on the plates from fuel element ET-51 also indicate higher hydrogen concentrations at the edges than at the centers of the plates. The values ranged from 26 to 76 ppm at the edges of the plates

with an average value of 46 ppm and a standard deviation of ± 10 ppm. In comparison, the values at the centers of the plates ranged from 11 to 41 ppm with an average value of 20 ppm and a standard deviation of ± 7 ppm.

Analysis for the hydrogen content of a similar unirradiated plate indicated 30 to 40 ppm at the edge and 11 ppm at the center. Therefore, the data indicate that some hydrogen was picked up by the fuel plates under reactor operating conditions. The plate temperature at the edges is estimated to be 250°C. Based on the work of A. Sawatsky,⁽¹²⁾ the solubility of hydrogen in Zircaloy-2 at 250°C is about 60 ppm. Because the results for each specimen represent a lumped value, it is possible that the hydrogen concentration was significantly higher at the surface of the samples analyzed than in the interior. On the basis of these lumped values it is hypothesized that a small quantity of zirconium hydride may have formed at isolated locations along some plates, although this could not be verified metallographically. It is interesting to note that, although fuel element ET-51 was in the core $2\frac{1}{3}$ times as long as fuel element T-23, their hydrogen contents were of the same order of magnitude, indicating that over the period of time covered, hydrogen pick-up was not a linear function of time.

Postirradiation Annealing

Samples ranging in length from one inch to six inches and having burnups ranging from 0.01 to 0.36 a/o were cut from the plates for post-irradiation annealing studies. These specimens were clad on four sides with Zircaloy-2. An area of fuel 0.170 inch by 3.31 inches was exposed at each end of the sample. The studies were conducted in electrically heated pots containing a heat-treating salt with the following composition: 39% K_2CO_3 , 33% Na_2CO_3 , 28% Li_2CO_3 .

The samples were inserted into the molten salt after it had been stabilized at a desired temperature. Following removal of the samples from the molten bath, they were allowed to cool to room temperature and then placed in a dilute HNO_3 -HF acid bath to remove any adhering salt. The samples were then washed and rinsed in water and alcohol, and allowed to dry, after which their dimensions were measured and densities were determined by immersion in water to which a wetting agent had been added. It was evident that as the test progressed the salt was reacting slowly with the exposed portion of the fuel core; surface roughening of the fuel was more severe the higher the temperature and the longer the time of exposure. The results of the tests are tabulated in Tables IX to XII. The samples weighing about 1000 grams were six inches in length while those weighing 30 to 40 grams were about one-quarter inch in length. The values obtained on samples having approximately 0.35 a/o burnup are plotted in Figure 18. As can be seen in Figure 18, for temperatures of 500°C and 550°C bulk volume increases of 1 to 2% and 5 to 10%, respectively, were noted after 500 hours at temperature. At 600°C a volume increase of 17% was obtained during the first 45 hours of annealing.

TABLE IX

Effect of 450°C Anneal upon Sample of Irradiated EBWR Fuel Plate

Sample	Burnup, a/o	Test Temp, °C	Time at Temp, hr	Cumulative Time at Temp, hr	Weight, g	Volume, cc	Density, g/cc	Total ΔV , % ^(a)
828-2	0.35	-	0	0	1035.35	76.80	13.48	-0.2
		450-454	18	18	1033.87	76.54	13.51	

(a) Calculated from the expression

$$\frac{\Delta V}{V_i} = \frac{\rho_i}{\rho_f} - 1$$

where

 ΔV = change in volume, or $V_f - V_i$ V_f = final volume V_i = initial volume ρ_i = initial density ρ_f = final density

TABLE X

Effect of 500°C Anneal upon Samples of Irradiated EBWR Fuel Plates

Sample	Burnup, a/o	Test Temp, °C	Time at Temp, hr	Cumulative Time at Temp, hr	Weight, g	Volume, cc	Density, g/cc	Total ΔV , % ^(a)	Sample Thickness, in.
HT-3	0.35	-	-	-	1103.83	83.13	13.28	-	0.224
		496-513	24	24	1099.14	83.35	13.19	0.7	-
		491-503	24	48	1098.54	83.40	13.17	0.8	-
		493-502	65	113	1097.63	83.30	13.18	0.8	-
		491-515	108	221	1096.16	83.42	13.14	1.1	-
		496-499	94	315	1095.34	83.41	13.13	1.1	-
		496-505	162	477	1094.13	82.97	13.19	0.7	0.230
HT-4	0.34	-	-	-	1084.37	81.64	13.28	-	-
		496-507	22	22	1080.03	81.17	13.31	-0.2	-
		499-510	95	117	1079.05	81.63	13.22	0.5	-
		490-505	65	182	1077.99	82.23	13.11	1.3	-

(a) See footnote a, Table IX.

TABLE XI

Effect of 550°C Anneal upon Samples of Irradiated EBWR Fuel Plates

Sample	Burnup, a/o	Test Temp, °C	Time at Temp, hr	Cumulative Time at Temp, hr	Weight, g	Volume, cc	Density, g/cc	Total ΔV , % ^(a)	Sample Thickness, in.
828-2	0.35	-	-	-	1033.87	76.54	13.51		0.218
		544-554	20	20	1034.40	78.68	13.15	2.7	-
		550-552	65	85	1032.02	79.30	13.01	3.8	-
		557-560	70	155	1028.76	80.02	12.86	5.1	-
		543-560	76	231	1025.50	77.97	13.15	2.7	-
		546-550	112	343	1022.42	80.02	12.78	5.7	-
		546-553	159	502	1015.57	80.31	12.65	6.8	0.235
825-V	0.28	-	-	-	40.848	2.950	13.85		-
		550-552	65	65	39.589	2.983	13.27	4.4	-
T23-4	0.11	-	-	-	42.686	3.089	13.82		0.218
		550-552	65	65	42.604	3.085	13.81	0.1	-
		557-560	70	135	42.132	3.166	13.31	3.8	0.225
		543-560	76	211	41.210	3.026	13.62	1.5	0.225
		546-550	112	323	40.650	3.030	13.42	3.0	0.225
		546-553	159	482	39.088	3.079	12.70	8.8	0.213
T23-6	0.09	-	-	-	34.172	2.463	13.87		0.216
		550-552	65	65	34.137	2.470	13.82	0.4	-
		557-560	70	135	33.831	2.460	13.75	0.9	0.217
		543-560	76	211	33.430	2.452	13.63	1.8	0.222
		546-550	112	323	32.750	2.416	13.56	2.3	0.226
		546-553	159	482	32.284	2.662	12.13	14.3	0.236
T23-10	0.06	-	-	-	44.924	3.229	13.91		0.215
		550-552	65	65	44.959	3.239	13.88	0.2	-
		557-560	70	135	44.862	3.235	13.87	0.3	0.215
		543-560	76	211	44.633	3.223	13.85	0.4	0.217
		546-550	112	323	44.413	3.216	13.81	0.7	0.219
		546-553	159	482	43.847	3.215	13.64	2.0	0.223
T23-14	0.03	-	-	-	39.962	2.868	13.93		0.214
		550-552	65	65	39.837	2.868	13.89	0.3	-
		557-560	70	135	39.666	2.860	13.87	0.4	0.212
		543-560	76	211	39.401	2.841	13.87	0.4	0.214
		546-550	112	323	39.072	2.832	13.80	0.9	0.215
		546-553	159	482	38.678	2.845	13.60	2.4	0.216
T23-18	0.01	-	-	-	42.130	3.056	13.79		0.212
		550-552	65	65	41.924	3.045	13.77	0.1	-
		557-560	70	135	41.632	3.026	13.76	0.2	0.206
		543-560	76	211	41.045	3.000	13.68	0.8	0.206
		546-550	112	323	40.760	3.009	13.55	1.8	0.208
		546-553	159	482	40.601	3.106	13.07	5.5	0.209

(a) See footnote a, Table IX.

TABLE XII

Effect of 600°C Anneal upon Samples of Irradiated EBWR Fuel Plates

Samples	Burnup, a/o	Test Temp, °C	Time at Temp, hr	Cumulative Time at Temp, hr	Weight, g	Volume, cc	Density g/cc	Total ΔV , % ^(a)	Sample Thickness, in.
HT-9	0.35	--	--	--	1105.23	82.61	13.38		0.219
		582-596	23	23	1103.97	92.13	11.98	11.7	--
		590-596	22	45	1096.48	96.19	11.40	17.4	0.250
821-1	0.36	--	--	--	50.400	3.670	13.73		0.226
		582-596	23	23	48.360	3.838	12.60	9.0	0.222
		590-596	23	45	Specimens disintegrated during test				--
T23-3	0.11	--	--	--	44.065	3.203	13.76		0.218
		582-596	23	23	43.231	3.147	13.74	0.1	0.224
		590-596	22	45	42.394	3.219	13.17	4.5	0.212
T23-5	0.09	--	--	--	44.091	3.185	13.84		0.216
		582-596	23	23	43.527	3.153	13.80	0.3	0.225
		590-596	22	45	42.752	3.140	13.62	1.6	0.221
T23-9	0.06	--	--	--	44.697	3.215	13.90		0.215
		582-596	23	23	44.666	3.215	13.89	0.1	0.217
		590-596	22	45	44.374	3.199	13.87	0.2	0.218
T23-13	0.03	--	--	--	36.532	2.623	13.93		0.214
		582-596	23	23	36.505	2.636	13.85	0.6	0.217
		590-596	22	45	36.318	2.622	13.85	0.6	0.216
T23-17	0.01	--	--	--	42.359	3.063	13.83		0.212
		582-596	23	23	42.245	3.098	13.64	1.4	0.222
		590-596	22	45	41.985	3.041	13.81	0.1	0.209

(a) See footnote a, Table IX.

Figure 18. Percent Increase in Volume of Sections of U-5 w/o Zr-1.5 w/o Nb Alloy Fuel Plate Versus Annealing Time for Various Temperatures

There was considerable scatter in the values for samples with burnups of 0.1 a/o and less, but in general, the volume increases were not as severe for a given temperature and time of heating during the initial periods of heating as those samples having approximately 0.35 a/o burnup. However, after 500 hours at 550°C, samples with burnups of 0.1 a/o and less exhibited volume increases of the same order of magnitude as those having 0.35 a/o burnup.

CONCLUSIONS

1. EBWR fuel elements are in generally satisfactory condition after operation at central fuel temperatures up to at least 400°C and burnup of 0.4 a/o.
2. Volume increases of 6 to 7% ΔV per a/o burnup due to the burnup of uranium can be expected on EBWR fuel plates operated at fuel temperatures up to 400°C and 0.4 a/o burnup.
3. The hardness of the EBWR fuel core alloy has increased with increasing burnup over the range up to 0.4 a/o. For burnups of 0.3 a/o to 0.4 a/o, Rockwell C values in the low to middle 50's respectively, are to be expected.
4. Zircaloy-2 possesses considerable ductility after an integrated exposure of 1.6×10^{21} nvt fast at temperatures of 260 to 325°C.
5. For burnups from 0.005 to 0.11 a/o the corrosion resistance of the irradiated EBWR fuel alloy is superior to that of the unirradiated material.
6. Hydrogen is picked up by the EBWR fuel plates under reactor operating conditions with the probable formation of isolated areas of small amounts of zirconium hydride. The plates operate in demineralized water with a pH of 7 and a surface temperature of 250 to 325°C.
7. Annealing studies on sections of fuel plate at 500 and 550°C indicated bulk volume increases of 1 to 2% and 5 to 10%, respectively, after 500 hours.
8. Annealing studies at 600°C on sections of fuel plate with 0.35 a/o burnup indicated a bulk volume increase of 17% after 45 hours.

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