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Argonne National Laboratory

THE EBR-II MATERIALS-SURVEILLANCE PROGRAM:

V. Results of SURV-5

by

W. E. Ruther, J. D. Staffon,
B. G. Carlson, and T. R. Allen

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W. E. Ruther,* J. D. Staffon,** B. G. Carlson,*** and T. R. Allen

Engineering Division
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**Engineering Division
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THE EBR-II MATERIALS-SURVEILLANCE PROGRAM:

V. Results of SURV-5

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ABSTRACT

In March of 1965, a set of surveillance (SURV) samples was placed in the EBR-II reactor to determine the effect of irradiation, thermal aging, and sodium corrosion on reactor materials. Eight subassemblies were placed into row 12 positions of EBR-II to determine the effect of irradiation at 370°C. Two subassemblies were placed into the primary sodium basket to determine the effect of thermal aging at 370°C. One half of all samples were exposed to primary system sodium while one half were sealed in capsules with a helium atmosphere. Fifteen different structural materials were tested in the SURV program. In this work, the properties of these materials irradiated at 370°C to a total fluence of 3.2×10^{22} n/cm^2 were determined. These materials are the fifth set of irradiated subassemblies to be examined as part of the SURV program (SURV-5). The properties analyzed were weight, density, microstructure, hardness, tensile and yield strength, and fracture resistance.

Of all the alloys examined in SURV-5, only Berylco-25 showed any significant weight loss. Stainless steel (both 304 and 347) had the largest density decrease, although the density decrease from irradiation for all alloys was less than 0.4 percent. The microstructure of both Berylco-25 and the aluminum-bronze alloy was altered significantly. Iron- and nickel-base alloys showed little change in microstructure. Austenitic steels (304 and 347) harden with irradiation. The hardness of Inconel X750 did not change significantly with irradiation. The ultimate tensile strength of Inconel X750, 304 stainless steel, 420 stainless steel and welded 304 changed little due to a fluence increase from 2.2×10^{22} n/cm^2 (the maximum fluence of the SURV-4 samples) to 3.2×10^{22} n/cm^2.

I. INTRODUCTION

In March of 1965, a set of surveillance (SURV) samples was placed in the EBR-II reactor to determine the effects of irradiation, thermal aging, and sodium corrosion on reactor materials. Initially, eight subassemblies (SURV 1-5 and SURV 8-10) containing 15 alloys used in the primary system of EBR-II and also containing shield graphite canned in Type 304 stainless steel were placed in the EBR-II blanket (Row 12) at 370°C. Two other subassemblies (SURV 6-7) were placed in the primary sodium tank storage basket at 370°C to separate thermal and radiation effects. Half of the alloy specimens were exposed directly to reactor sodium, while the remainder were sealed in helium. The alloys included Ampco Grade 18 aluminum bronze; Stellite 6B; Inconel X-750; T-1 tool steel; Berylco-25 (beryllium-copper); Types 304, 347, 416, and 420 stainless steel; Type 17-4 PH stainless steel; and tantalum.

This report presents the results of examination of the test specimens from SURV-5. The SURV-5 materials were irradiated at 370°C to a total peak fluence of 3.2×10^{22} n/cm^2. In addition to the measurements on irradiated samples, control samples were also measured to provide baseline data. The program and experimental methods were described previously [1] as were the results of SURV-1, -2, –3, -4, and -6 [1-4]. In all important aspects, the loading diagrams for these subassemblies are the same as reported [1] for SURV-1.

Although this work was accomplished over 20 years ago, the results were never published in a single document. This report is being published now to ensure that the valuable data from the SURV experiments is widely disseminated and to fully document the early SURV work prior to analysis of the samples from the last four SURV subassemblies (SURV 7-10).

II. DOSIMETRY AND EXPOSURE

SURV-5 was removed from the reactor on December 8, 1974. It had been in the reactor in position 12D-7 at approximately 370°C for 3335 days during which time the reactor logged 84,670 MW days of operation.

TABLE I. Summary of SURV Exposures

SURV	MWd	Total Fluence, n/cm^2
1	11541	9.0×10^{20}
2	26274	3.0×10^{21}
3	41111	1.2×10^{22}
4	64439	2.2×10^{22}
5	84670	3.2×10^{22}

The maximum neutron fluence for SURV-5 was estimated to be about 3.2×10^{22} n/cm^2 (E>0.1 MeV). A summary of exposures for SURV 1 through 5 is given in Table I.

III. RESULTS OF POST-IRRADIATION EXAMINATIONS

A. Weight Change

To determine the corrosion resistance of core structural materials in the primary system sodium, weight loss was measured. Table II shows the weight changes for specimens exposed to the sodium coolant. Before being weighed, the specimens were ultrasonically cleaned in distilled water and rinsed in a 50-50 mixture of methanol and benzene. Intense radioactivity of the tantalum samples precluded their measurement in the hot-cell facility. As with the earlier SURV experiments, only the Berylco-25 specimens underwent significant weight loss. Because of the serious corrosion indicated by the weight loss, Berylco-25 and unclad tantalum components are no longer used in the reactor, although there are still specimens exposed to the coolant in the remaining SURV subassemblies.

B. Density Change

Density was measured to determine the propensity of core materials to swell. Table III shows the density changes for the specimens held in the helium capsules. The platinum-iridium standard used to determine the initial densities of the samples was used to determine their densities after irradiation. Intense radioactivity of the tantalum samples precluded their measurement in the hot-cell facility. All of the alloys decreased in density by less than 0.4 percent.

TABLE II. Variation in Weight of SURV-5 Corrosion Specimens[a]

Material	Weight Change, g
Aluminum Bronze Ampco-18	-0.0014 to -0.0036
Stellite 6B	-0.0001 to -0.0021
Stainless Steel Type 420	0 to -0.0003
Inconel 750X	+0.0004 to 0.0009
Tool Steel T-1	+0.0001 to -0.0008
Stainless Steel Type 347	-0.0001 to -0.0003
Copper-Beryllium Berylco-25	-0.935 to -3.022
Stainless Steel Type 304 + boron	-0.0003 to -0.0009
Stainless Steel Type 17-4 PH	-0.0001 to 0.0005
Stainless Steel Type 304	+0.0002 to -0.0011

[a]Rod specimens with an area of 9.69 cm^2 exposed to EBR-II primary sodium.

TABLE III. Density of SURV-5 Specimens

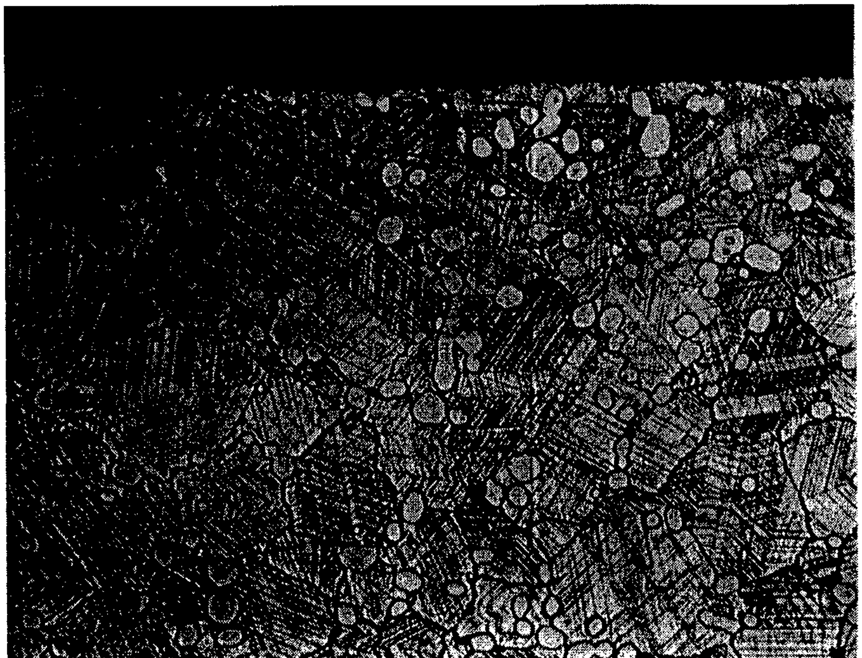
Material	Pre-irradiation[a] Density g/cm^2	SURV-5 Density g/cm^2	Change	% Change
Aluminum Bronze Ampco-25	7.477 + 0.015 - 0.010	7.474	-0.003	-0.04
Stellite 6B	8.376 + 0.003 - 0.011	8.374	-0.002	-0.02
Inconel 750X	8.267 + 0.007 - 0.013	8.259	-0.008	-0.10
Stainless Steel Type 420	7.698 + 0.005 - 0.003	7.692	-0.006	-0.08
Tool Steel, T-1	8.656 + 0.003 - 0.002	8.630	-0.026	-0.30
Stainless Steel Type 347	7.908 + 0.003 - 0.002	7.883	-0.025	-0.32
Stainless Steel Type 416	7.638 + 0.004 - 0.002	7.629	-0.009	-0.12
Copper-Beryllium Berylco-25	8.348 + 0.007 - 0.002	8.341	-0.007	-0.08
Stainless Steel Type 304 + Boron	7.760 + 0.003 - 0.006	7.737	-0.023	-0.30
Stainless Steel 17-4 PH	7.750 + 0.003 - 0.004	7.737	-0.013	-0.17
Stainless Steel Type 304	7.935 + 0.003 - 0.002	7.905	-0.030	-0.38

[a] Initial density (average of 10 specimens).

C. Metallography

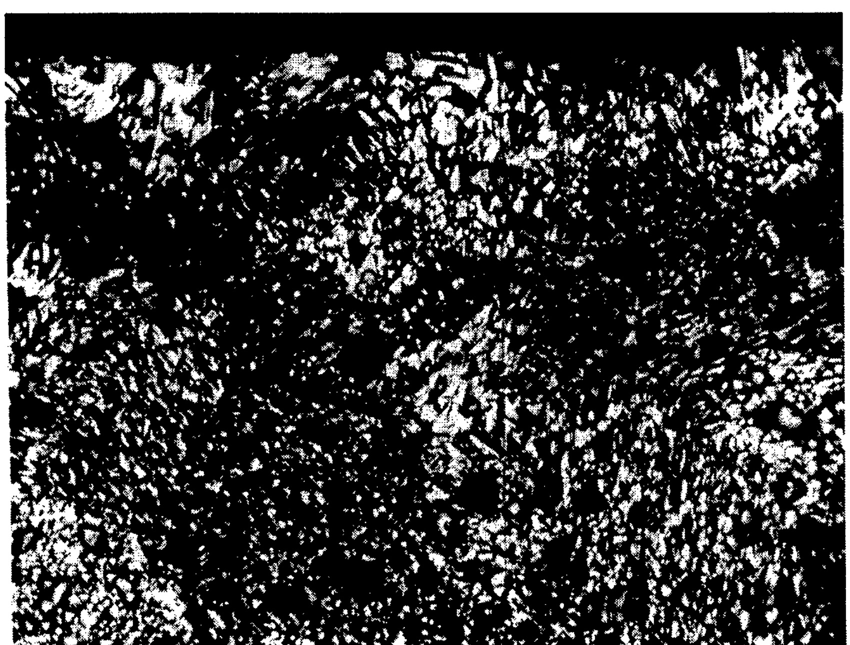
To determine the details of how irradiation and corrosion effect core structural materials, microstructure was examined. The microstructure of irradiated samples is compared to control samples for each alloy in Figs. 1 through 11. Very little change in microstructure was noted in the iron- and nickel-base materials, except for some sensitization observed in the Type 347 stainless steel. The same condition was seen in the SURV-4 Type 347 stainless steel sample, but considered anomalous, possibly caused by improper heat treatment. However, the unirradiated and thermally aged samples from the same lots do not reveal carbide precipitation, and perhaps this sensitization is real and caused by the irradiation environment.

Microstructure of the Berylco 25 is altered and shows heavy surface attack from sodium. The aluminum bronze material has changed appearance due to what appears to be agglomeration of one of the phases. The appearance of Stellite 6B was also radically changed compared with the control sample. Reaction of the Stellite to its etchant was much faster, suggesting a chemical change in the alloy. In the 304 stainless steel, a small amount of carbide is now present on some of the grain boundaries, but it is quite localized.



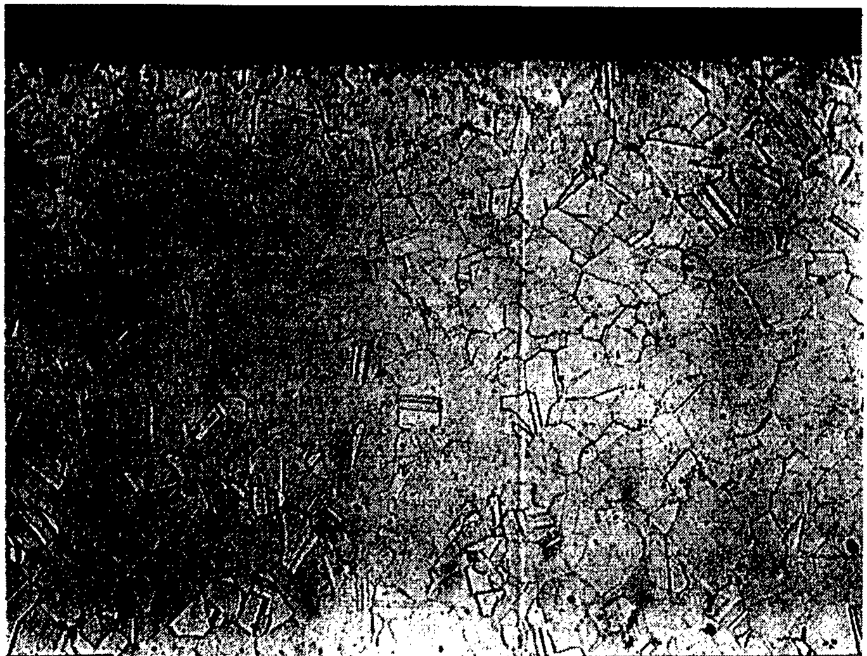
500 X

Control Sample Stellite 6B
($HNO_3 - HCl - H_2O$ etchant)



500 X

Figure 1. SURV-5 Stellite 6B. Structure is completely altered.
($HNO_3 - HCl - H_2O$ etchant)



500 X

Control Sample Inconel X750
(HNO_3 + Acetic + H_2O etchant)

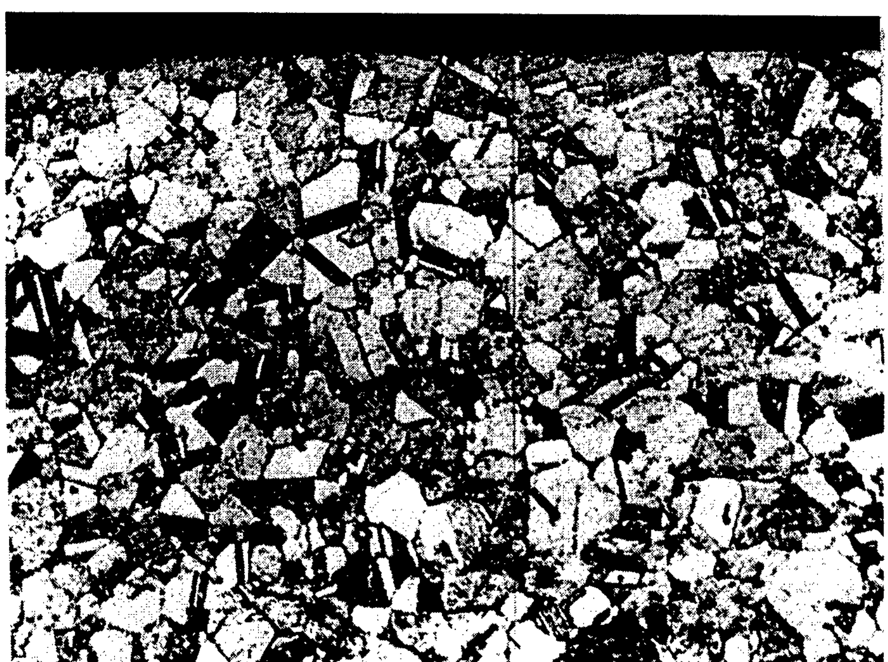


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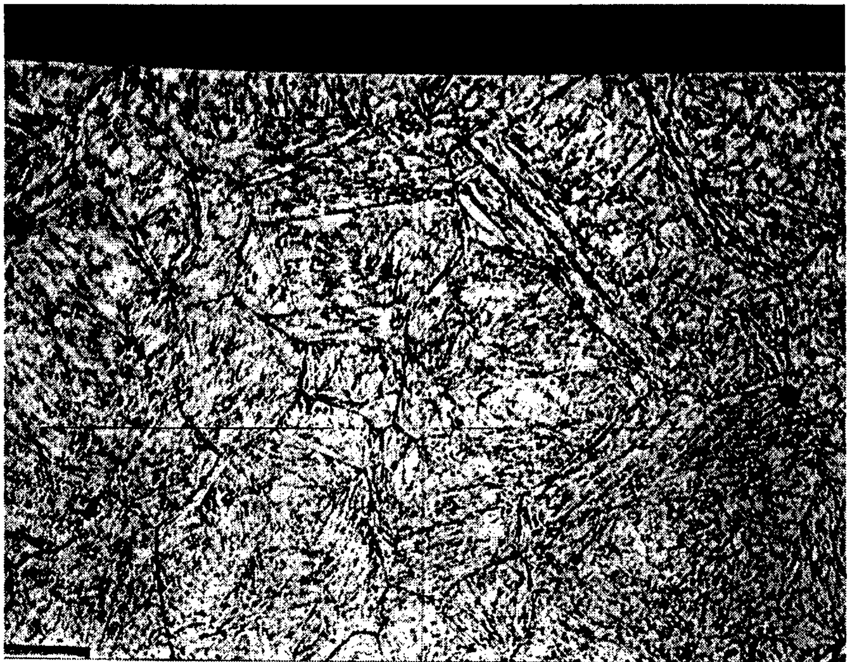
Figure 2. SURV-5 Inconel X750. No significant change in structure is apparent.
(HNO_3 + Acetic + H_2O etchant)



500 X

Control Sample Tyoe 420 Stainless Steel
(HNO_3 – CH_3OOH etchant)

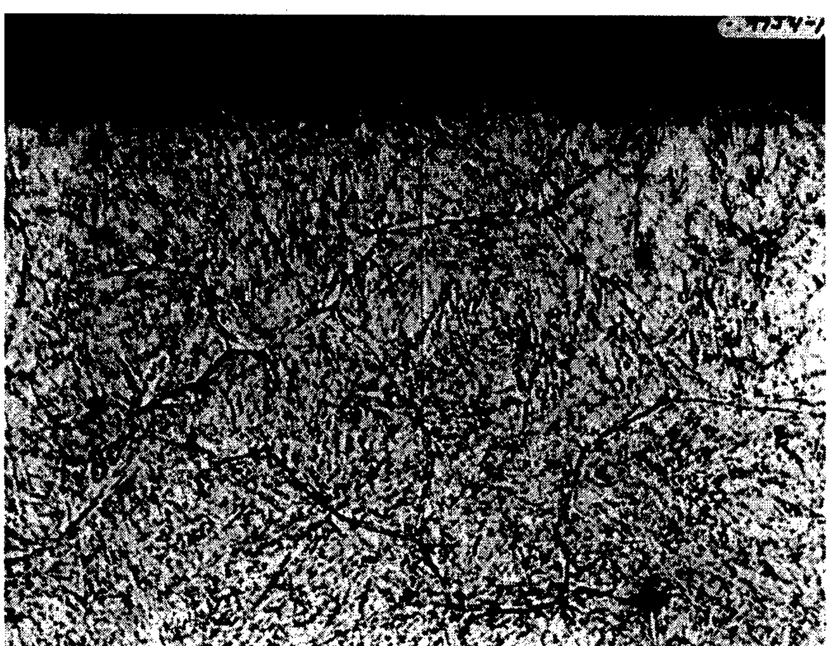


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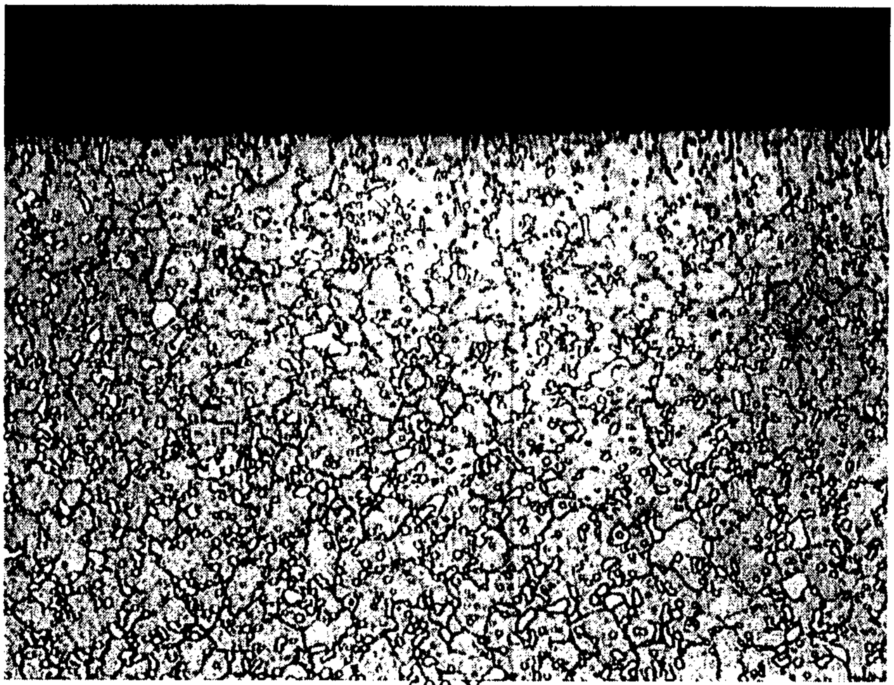
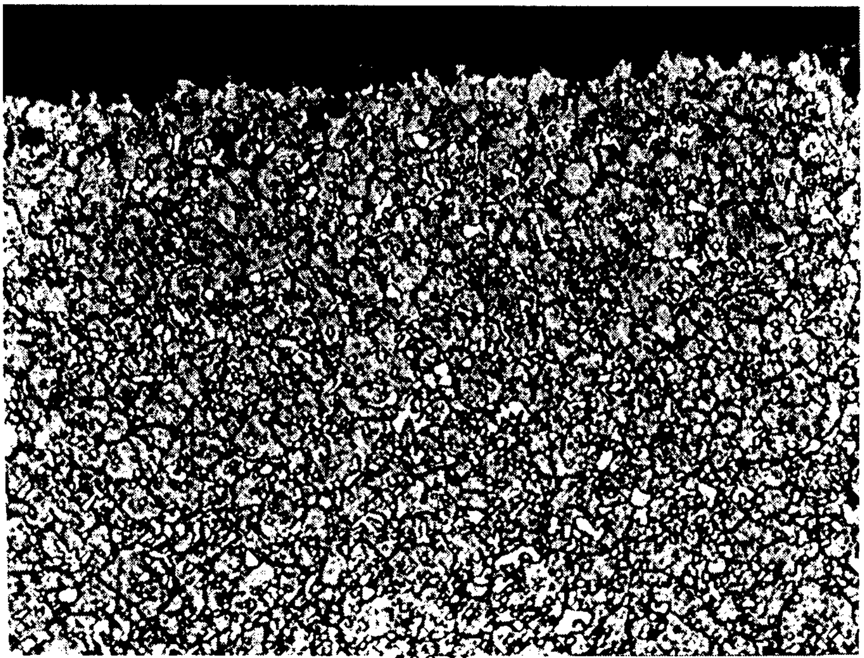
Figure 3. SURV-5 Type 420 Stainless Steel. No change in structure.
(HNO_3 – CH_3OOH etchant)



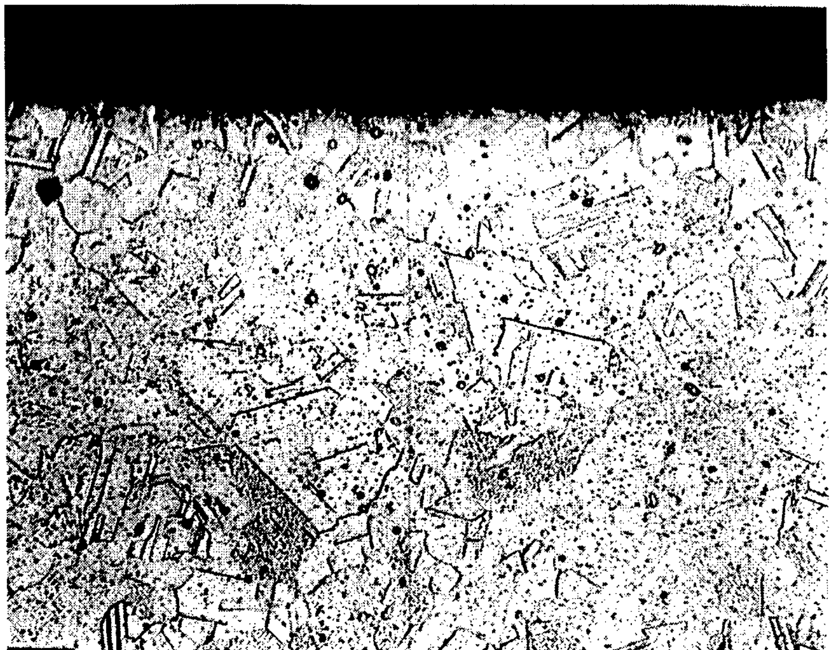
500 X

Control Sample Type T-1 Tool Steel
($HNO_3 + H_2O$ etchant)



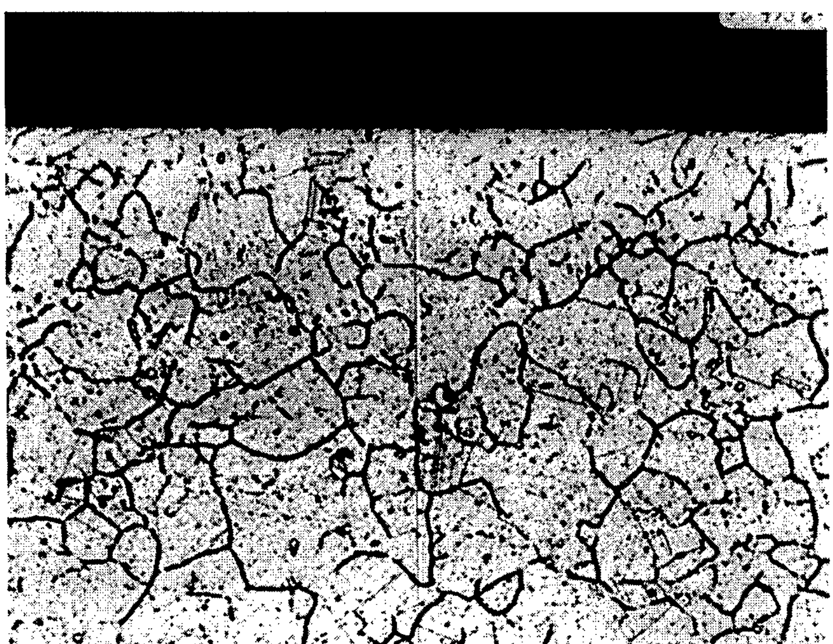
500 X

Figure 4. SURV-5 Type T-1 Tool Steel. No change due to irradiation.
($HNO_3 + H_2O$ etchant)



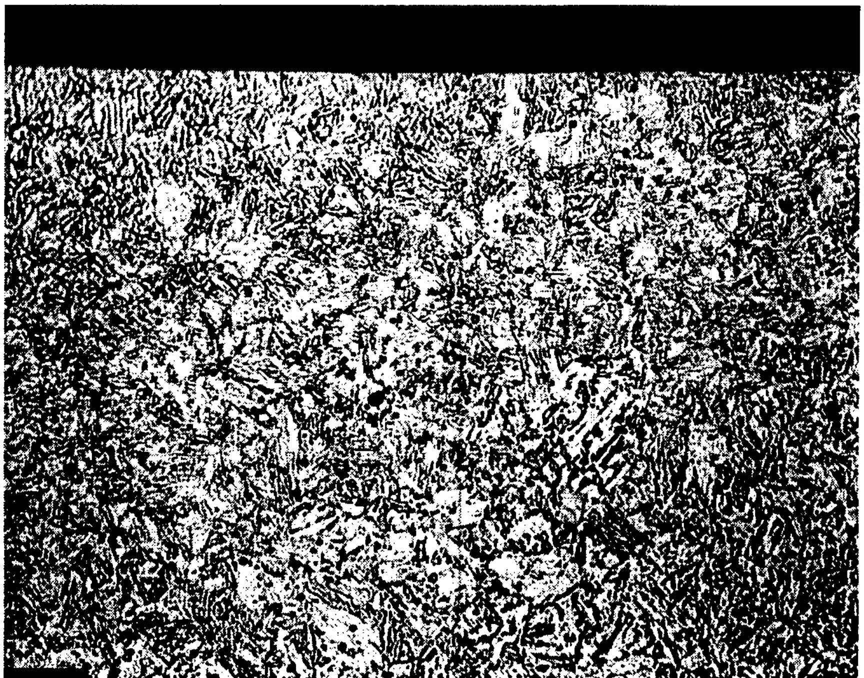
500 X

Control Sample Type 347 Stainless Steel
(Oxalic etchant)



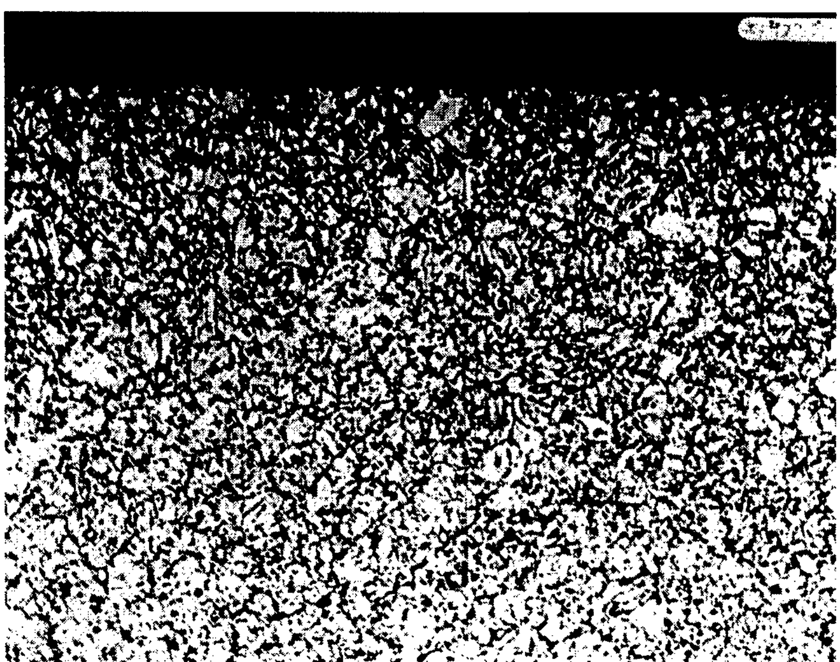
500 X

Figure 5. SURV-5 Type 347. Unexpected sensitization is seen.
(Oxalic etchant)



500 X

Control Sample Type 416 Stainless Steel
($FeCl_3 + HCl + H_2O$ etchant)

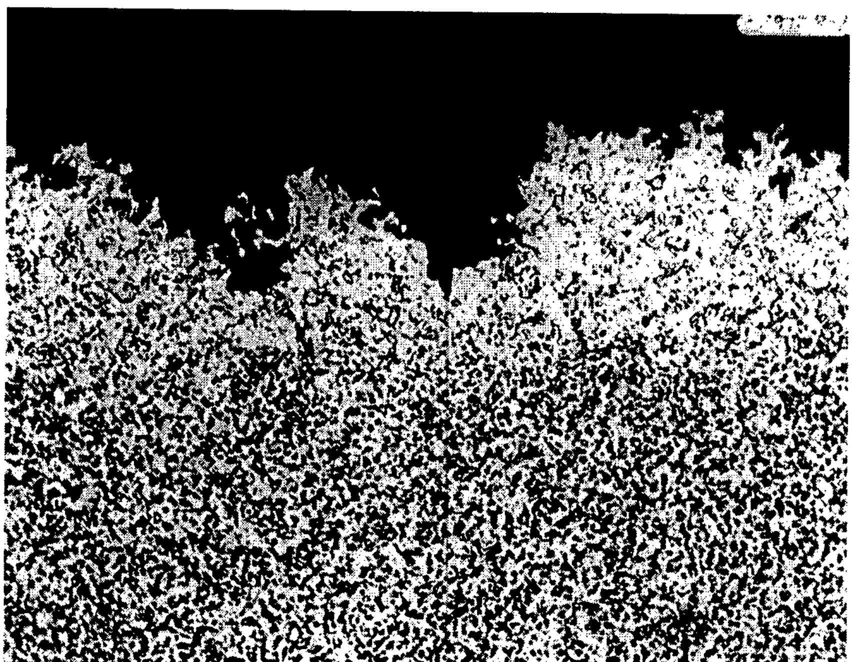


500 X

Figure 6. SURV-5 Type 416 Stainless Steel. The darker martensite in the irradiated sample is probably a tempering effect from thermal aging.
($FeCl_3 + HCl + H_2O$ etchant)

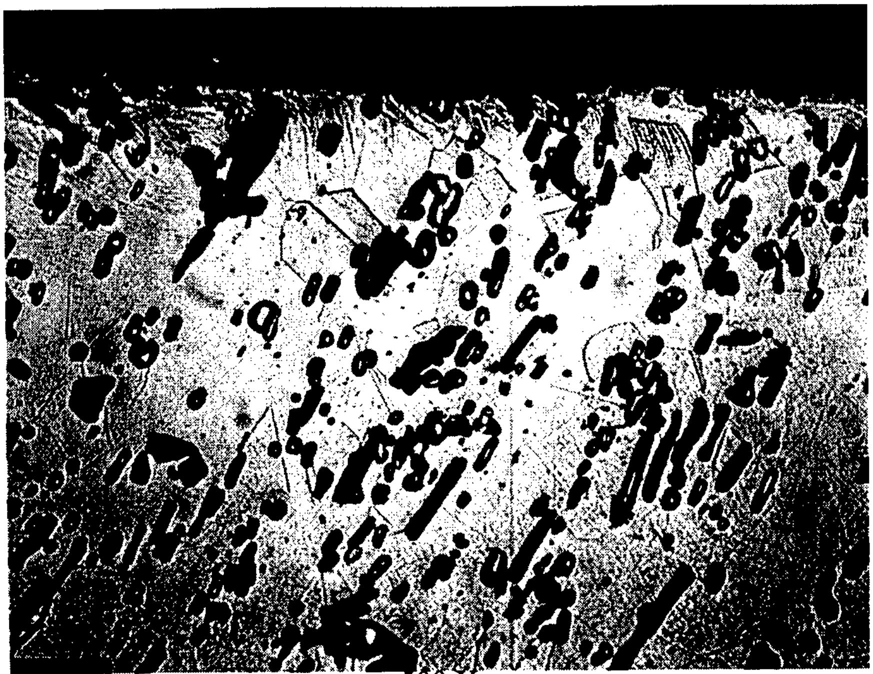
500 X

Control Sample Berylco-25
($NH_4OH - H_2O$ etchant)



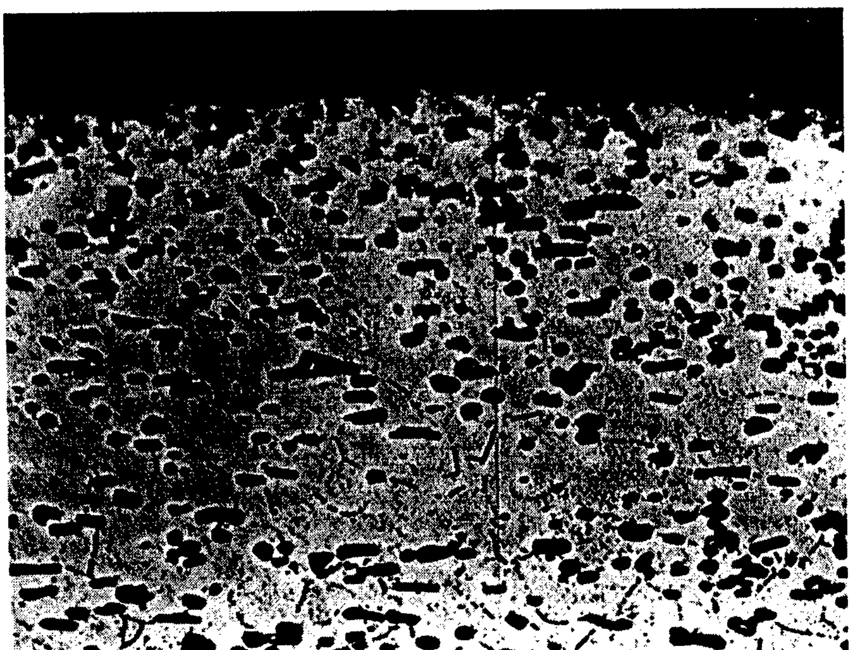
500 X

Figure 7. SURV-5 Berylco-25. Massive attack has corroded the outer diameter of the SURV-5 Berylco material. A complete alteration of the microstructure has also taken place.
($NH_4OH - H_2O$ etchant)



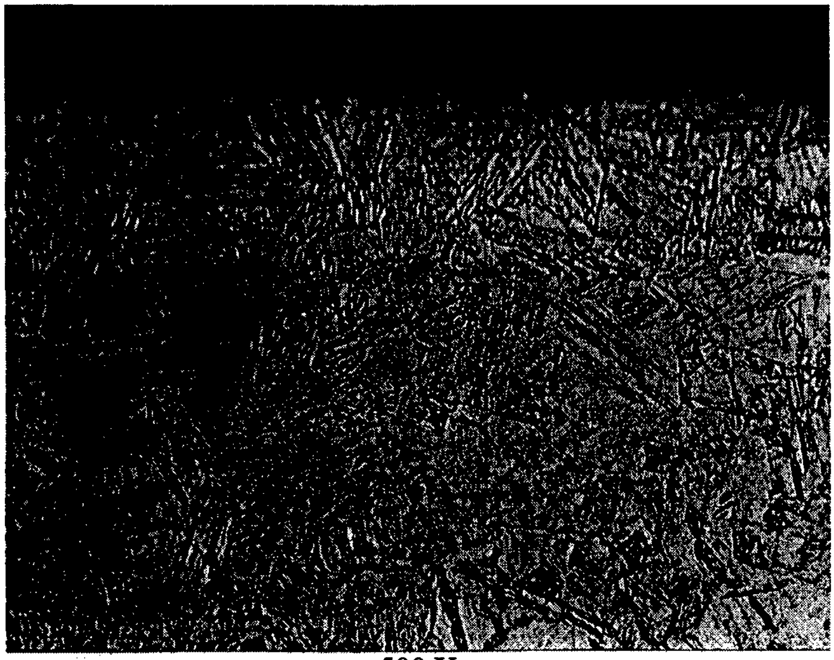
500 X

Control Sample Type 304 +B
(Oxalic etchant)



500 X

Figure 8. SURV-5 Type 304+B. No apparent change due to irradiation.
(Oxalic etchant)



500 X

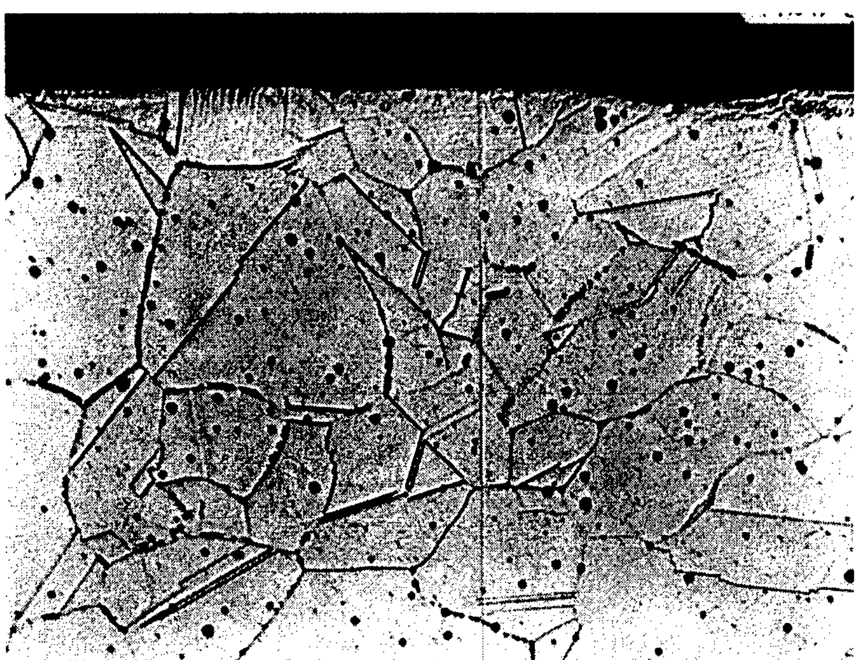
Control Sample Type 17-4 PH
($FeCl_3$ + HCl etchant)

500 X

Figure 9. SURV-5 Type 17-4 PH.
($FeCl_3$ + HCl etchant)

500 X

Control Sample Type 304 stainless steel
(Oxalic etchant)

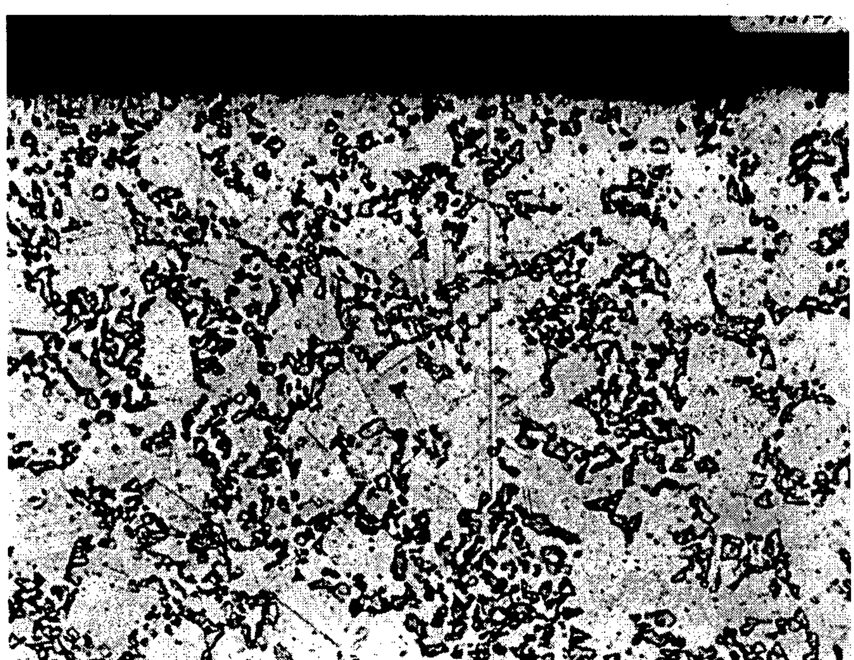


500 X

Figure 10. SURV-5 Type 304. A small amount of carbide precipitation has taken place at some of the grain boundaries in the irradiated sample.
(Oxalic etchant)

500 X

Control Sample Aluminum Bronze
($NH_4OH - H_2O_2$ etchant)



500 X

Figure 11. Aluminum Bronze, SURV-5
($NH_4OH - H_2O_2$ etchant)

D. Hardness

Hardness was measured to estimate changes in strength. Microhardness was measured on a remote Tukon tester using a 500 gram load. Only the maximum fluence samples which had been exposed to sodium were analyzed. A control group of unirradiated or as-received SURV-type samples were prepared with these SURV-5 materials for comparing hardness.

Some inconsistencies were found between the starting or as-received hardness data found in EBR-II Technical Memorandum #48 and the data reported in ANL-7624 (SURV-I Results) [1]. To reconcile these differences, all of the unirradiated control samples were hardness tested. The retesting indicated a higher starting hardness for the 420 steel and the Berylco 25. A substantially higher hardness (378 vs. 298 DPH) was found in the control 17-4 PH material, which is closer to the specified hardness requested at the initiation of the SURV experiment. All of the other materials were essentially as reported in ANL-7624. Hardness of both the unirradiated and SURV-5 materials are shown in Table IV. Additionally, data from SURV-6 (thermally aged for 2994 days at 370°C) is included to show how the hardness changes due to time at temperature only.

A graphic summary of hardness change during irradiation for a portion of the samples is shown in Fig. 12. This summary was compiled from the previous SURV reports and includes SURV 1-5 data. Viewed graphically, the hardenable stainless steels 416, 420, and 17-4 PH show very similar changes in hardness through their reactor life. As a group, they tend to either gain or lose hardness together. Inconel X750 maintained the most consistent hardness throughout the life of the experiment increasing only 20 DPH in almost ten years.

TABLE IV. SURV-5 Hardness (DPH) Compared with As-Fabricated and SURV-6* Reactor Basket Samples

Sample Number	Material	As Fabricated	SURV-5	SURV-6
VA 2	Al Bronze	185	202	222
VB 2	Stellite 6B	382	132	385
VC 2	Inconel X750	372	394	362
VD 1	420 SS	385	365	380
VE 1	T 1 Steel	685	658	685
VF 1	347 SS	155	277	149
VG 3	416 SS	295	310	310
VH 3	Berylco 25	378	144	148
VI 3	304 + B	215	278	227
VJ 3	17-4 PH	378	415	454
VK 2	304	155	288	154

*SURV-6 was stored in the primary sodium tank storage basket for 2994 days.

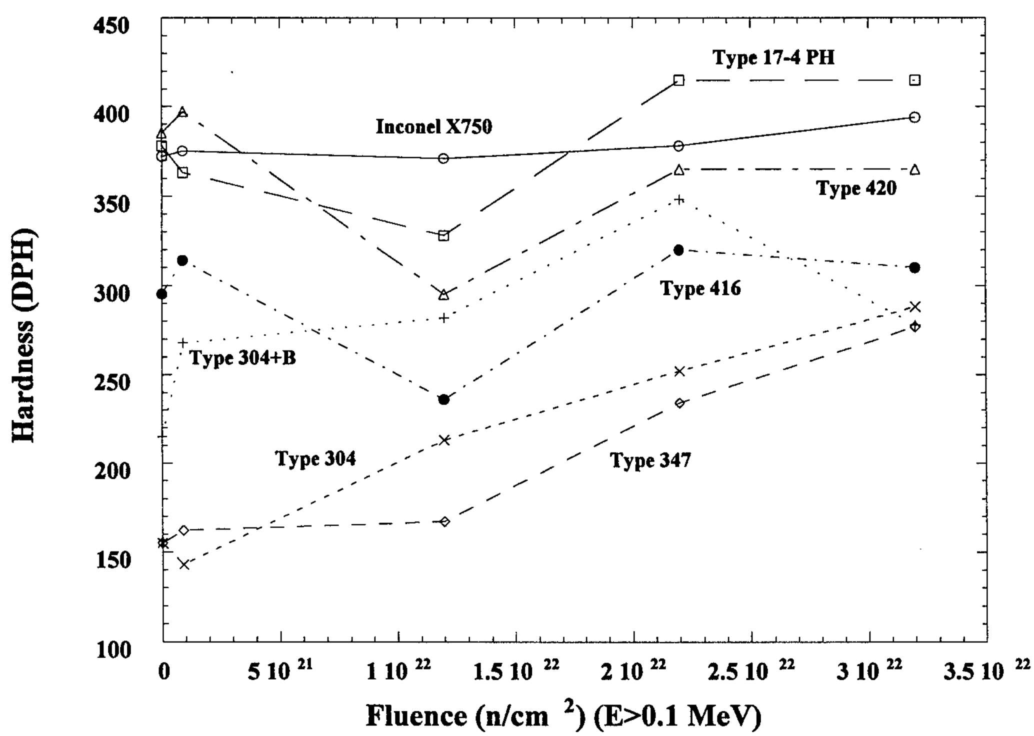


Figure 12. Hardness as a function of fluence for Fe-Cr-Ni SURV materials

E. Strength and Ductility

Tensile tests were performed to determine changes in strength and ductility. The results of tensile tests performed on specimens from SURV-5 are given in Table V. A comparison of the results for SURV-5 specimens with those for SURV-4 [4] (the last set of irradiated samples analyzed) indicates minimal changes in ultimate tensile strength (UTS) for Inconel 750X and Type 420, wrought Type 304, and welded Type 304 stainless steel. Aluminum bronze and EBR-II cover-plate material experienced small increases in UTS and yield strength (YS). The most important observation to the operation of EBR-II is that the EBR-II cover-plate material (made of 304 stainless steel) has retained considerable ductility.

Three archive specimens (with no reactor exposure) of the EBR-II cover-plate material were also tested, at 370°C, for comparison with the irradiated material. The results are given in Table VI. The specimens had been stored in room-temperature air.

F. Bend Tests of Welded Type 304 Stainless Steel

To determine the strengthening effect of fluence on 304 stainless steel, bend test samples were tested at ambient temperature supported on round pins on 2-in. centers, with the load applied perpendicularly to the center of the 3.75 x 0.424-in. face by a third round pin. No sample fractured during the test, and visual examination revealed no cracks or other abnormalities. The results are given in Table VII. No effect of reactor environment is evident.

IV. DISCUSSION

This report is the fifth in the series to provide measurements on the effect of radiation on the properties of EBR-II structural materials. Of all the alloys examined, only Berylco-25 showed any significant weight loss. Stainless steel (both 304 and 347) had the largest density decrease, although the density decrease from irradiation for all alloys was less than 0.4%. The microstructure of both Berylco-25 and the aluminum bronze alloy was altered significantly. Iron- and nickel-base alloys showed little change in microstructure. Austenitic steels (304 and 347) harden with irradiation. Inconel X750 maintains a fairly uniform hardness. The ultimate tensile strength of Inconel X750, 304 stainless steel, 420 stainless steel and welded 304 changed little due to a fluence increase from $2.2\text{x}10^{22}$ n/cm^2 to $3.2\text{x}10^{22}$ n/cm^2.

TABLE V. Results of Tensile Tests of Specimens from SURV-5

Material	Specimen No.	Environment	Test Temp °C	UPS ksi	YS ksi	Elongation %	Reduction in Area
Aluminum Bronze	1	Sodium	370	52.1	46.6	2 [a]	14
	3			51.0	44.5	3 [a]	5
	5	Helium	370	49.8	46.9	13 [b]	7
	7			56.5	50.7	10 [b]	2
	2	Sodium	21	92.5	55.2	15 [b]	12
	4			93.4	54.0	23 [b]	12
	6	Helium	21	97.5	78.8	13 [b]	12
	8			91.2	55.2	13 [b]	10
Inconel X750	1	Sodium	370	165.0	137.0	14	19
	3			161.0	127.0	36	23
	5	Helium	370	167.0	135.0	18	28
	7			159.0	115.0	13 [a]	23
	2	Sodium	21	187.0	160.0	10	10
	4			173.0	133.0	12 [a]	12
	6	Helium	21	187.0	160.0	15	26
	8			174.0	136.0	18	19
Type 420	1	Sodium	370	148.0	131.0	4 [a]	44
	3			151.0	135.0	3 [a]	42
	5	Helium	370	141.0	120.0	13	44
	7			159.0	141.0	15	40
	2	Sodium	21	184.0	162.0	16	44
	4			135.0	113.0	9 [a]	53
	6	Helium	21	174.0	153.0	13	46
	8			141.0	119.0	16	47
Type 304, Wrought	1	Sodium	370	105.0	96.2	16	56
	3			89.6	74.3	10 [a]	44
	5	Helium	370	115.0	108.0	17	49
	7			88.7	72.0	23	58
	2	Sodium	21	117.0	91.2	57	77
	4			121.0	102.0	51	74
	6	Helium	21	117.0	92.7	56	72
	8			124.0	106.0	52	75

TABLE V. Results of Tensile Tests of Specimens from SURV-5
(Continued)

Material	Specimen No.	Environment	Test Temp °C	UPS ksi	YS ksi	Elongation %	Reduction in Area
Type 304, Welded	1	Sodium	370	98.6	92.3	6 [b]	28
	3			74.8	53.7	16 [b]	28
	5	Helium	370	99.0	94.7	13	40
	7			182.0	122.0	11 [b]	19
	2	Sodium	21	109.0	87.6	25 [b]	51
	4			114.0	98.1	23 [b]	38
	6	Helium	21	115.0	95.6	24 [b]	44
	8			115.0	97.5	28 [b]	42
Type 304 EBR-II Cover-plate	1	Sodium	370	81.7	68.7	28	63
	3			87.2	80.3	14 [b]	58
	5	Helium	370	81.4	70.9	27	67
	7			87.2	79.6	25	54
	2	Sodium	21	109.6	85.3	81	75
	4			98.4	76.8	73	80
	6	Helium	21	106.9	85.5	74	75
	8			104.2	75.2	82	76

Conversions: 70 F = 21 C; 700 F = 371 C; 1 ksi = 6.895 MPa.
[a] Elongation inaccurate: sample broke outside gauge marks.
[b] Elongation suspect: sample broke at gauge mark.

TABLE VI. Results of Tensile Tests of Archive Specimens of EBR-II Cover-plate Material.

Specimen	UPS ksi	YS ksi	Elongation %	Reduction in Area
1	63.3	19.4	46	65
2	62.7	20.5	31	44
3	65.7	24.6	42	64

Conversion factor: 1 ksi = 6.895 MPa.

TABLE VII. Results of Bend Test of Welded Type 304 Stainless Steel Bars from SURV-5.

Specimen No.	Reactor Environment	Maximum Force kg
1	Sodium	185
2	Sodium	156
3	Sodium	184
4	Sodium	178
5	Helium	185
6	Helium	165
7	Helium	181
8	Helium	180

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