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FABRICATION AND IRRADIATION OF SM-2 CORE MATERIALS

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by

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Irradiation and examination have been completed on four capsules of specimens prepared in the SM-2 Core Materials Development Program. Each capsule contained five to eight flat-plate dispersion specimens roll clad with Type 347 stainless steel. Six capsules containing 36 specimens are still under irradiation in the ETR. Variables being studied include use of (1) fuel concentrations of nominal 26 and 40 w/o UO_2 and 34 w/o UN, (2) hydrothermal UO_2, spherical UO_2, and UN fuels, (3) B_4C, NbB_2, and ZrB_2 dispersion poisons and no poison loading, (4) a half-loading of reference ZrB_2 dispersion poison, (5) a boron-10 stainless steel picture frame, and (6) sintering or green pressing alone for core preparation.

The specimens in the first four capsules underwent burnups of 20 to 40 a/o of the uranium-235 at surface temperatures near 700 F. It was found that the burnable poisons, both dispersed in the fuel and alloyed in the cladding, performed satisfactorily. No specimens containing a nominal 40 w/o UO_2 or equivalent UN concentration were included in the first four capsules, but at the 26 w/o loading both the hydrothermal and spherical UO_2 performed well. Encouraging results were obtained for the nonsintered core.

INTRODUCTION

The irradiation program discussed in this report was outlined as an integral part of the SM-2 Core Materials Development Program under sponsorship of Alco Products. Because of a time limitation, it was necessary to fabricate irradiation specimens and initiate the irradiation program before much of the development work had been completed. For this reason, the reference-type irradiation specimens were not fabricated strictly according to final fuel-element specifications; however, the variables incorporated into the program give a well-rounded irradiation evaluation of SM-2 core materials.

The irradiation-program specimen variables of fuel type, fuel loading, poison type, and poison loading are listed in Table 1. Note that an SM-1-type specimen was prepared for comparison. Specimens will be listed throughout this report according to the specimen-type designation in Table 1. As listed in the table, a group of specimens containing suppressors is also included in the program. The Eu_2O_3 used in this suppressor dispersion was also produced by a different technique than is presently being employed.

The program consists of the irradiation of ten capsule loadings of specimens. The first three were noninstrumented capsules for insertion in the core of the MTR, and the last seven capsules contained thermocouples and internal heaters and were inserted in the ETR. The preparation of the specimens for all ten capsules is covered

TABLE 1. SM-2 CORE MATERIALS IRRADIATION SPECIMENS

Type	Fuel Loading, w/o			Type of Boron Compound		Matrix	Remarks
	Hydrate UO_2	Spherical UO_2	UN	Type	Enrichment		
1	24.18	--	--	B_4C	SM-1 loading, natural	Prealloyed	Fabricated by SM-1 Process
2	26	--	--	B_4C	Natural	Ditto	--
3	26	--	--	None	--	Ditto	--
4	--	26	--	None	--	Ditto	--
5	26	--	--	ZrB_2	Natural	Ditto	--
6	--	26	--	ZrB_2	Natural	Ditto	--
7	--	26	--	ZrB_2	Natural	Ditto	One-half boron loading
8	--	26	--	ZrB_2	Natural	Ditto	Contains suppressor
9	26	--	--	ZrB_2	Natural	Ditto	--
10	--	26	--	ZrB_2	Natural	Ditto	--
11	26	--	--	NbB_2	Natural	Ditto	--
12	--	26	--	ZrB_2	Natural	Ditto	Green compact
13	--	26	--	NbB_2	Natural	Ditto	--
14	--	26	--	Boron-10	Enriched	Ditto	Boron-stainless alloy picture frame
15	--	--	34.1	ZrB_2	Natural	Ditto	Increased fuel loading
16	38.4	--	--	ZrB_2	Natural	Ditto	Ditto
17	--	38.4	--	ZrB_2	Natural	Ditto	Ditto

in this report. Six of the seven capsules scheduled for irradiation in the ETR are still in the reactor; therefore, this report is a preliminary evaluation of the SM-2 core materials as concluded from data obtained from the three capsules irradiated in the MTR and one instrumented capsule irradiated in the ETR. The burnup obtained in these specimens ranged from 20 to 45 uranium-235 a/o with a specimen-surface temperature near 700 F.

The four capsules examined to date do not involve specimens with UN fuel or nominal 40 w/o UO_2. A report will be issued containing the data obtained with these variables as well as higher burnup data with all other variables at the completion of the program.

FABRICATION OF IRRADIATION SPECIMENS

Preparation of Core Compacts

Materials

Type 347 stainless steel with a maximum combined cobalt and tantalum content of 0.01 w/o was chosen for the structural material of the fuel element and was therefore used in the irradiation specimens. This material is stabilized with niobium and possesses good strength and corrosion resistance. The wrought metal was used as cladding and a prealloyed minus 325-mesh powder was used as the matrix material in the specimens. The powder was used in the prealloyed condition rather than as a mixture made up of the elemental constituents. This prevented the reaction which occurs between the nickel and boron compound before the elemental powders are placed in solution.

A compatibility study was conducted between Type 347 stainless steel and the boron compounds of CaB_6, CrB, CrB_2, FeB, Fe_2B, MoB, NbB_2, NiB, Ni_2B, SiB_3, TiB_2, ZrB_2, and YB_6. After the results of this study were evaluated ZrB_2 was chosen as the reference burnable poison and NbB_2 as an alternate.

A continued study[1] with ZrB_2 showed that it was desirable to sinter the compound prior to incorporation in the fuel core. It was also found that a vacuum sinter is more reliable and superior to a hydrogen or argon sinter. Although ZrB_2 is thought to be stable in pure hydrogen, pure argon, and in vacuo, it will react with oxygen which is normally present to a limited degree in commercial hydrogen and argon atmospheres.

The reference burnable poison used in the irradiation specimens was ZrB_2 heat treated in a vacuum of 1×10^{-4} mm of mercury for 3 hr at 2300 F and furnace cooled. The mesh size was minus 150 plus 200. Two batches of ZrB_2 powders were presintered and analyzed for boron content. The first contained 17.4 w/o boron and the second 16.6 w/o boron. The quantity of the compound added to the dispersion was in proportion to the boron content.

(1) References at end.

The alternate burnable poison chosen for this irradiation program is NbB_2. This material was used in the particle-size range of minus 150 plus 200 mesh and was analyzed and found to contain 16.6 w/o boron after being heat treated for 3 hr at 2300 F in vacuo.

Reactor-grade B_4C having a particle size of minus 150 plus 200 mesh and a boron content of 77.4 w/o was incorporated into the program as a standard to compare with previous results obtained in the SM-1 fuel elements.

Hydrothermal UO_2 was the reference fuel in the SM-1 fuel core. It was thought, however, that an improved particle distribution and core structure could be obtained with spherical UO_2 particles. Both hydrothermal and spherical UO_2 were used as fuel in the irradiation specimens. The hydrothermal UO_2 contains 88.17 per cent uranium which is 93.15 per cent enriched. The spherical particles have a uranium content of 87.98 per cent which is 93.17 per cent uranium-235.

The alternate fuel chosen for the irradiation program was UN. This uranium compound is a promising high-temperature fuel compatible with the other core constituents and has the advantage of containing a greater weight of uranium per unit volume than UO_2. This material was produced at Battelle as described in BMI-1365 and contains 94.60 w/o uranium of a 93.15 per cent enrichment. This powder was screened and the minus 100 plus 200-mesh fraction dispersed as the fuel phase in the compacts.

The suppressor-section matrix of Type 8 specimens is composed of elemental powders of minus 325-mesh electrolytic iron, electrolytic chromium, and carbonyl nickel. The elemental powders are preferred for the suppressor since they are of higher purity than prealloyed stainless steel powders and the Eu_2O_3 powder chosen for the suppressor and control material has a tendency to react with the impurities found in the prealloyed powders. The Eu_2O_3 powder dispersed in these specimens was prepared by pressing minus 325-mesh, high-purity, Eu_2O_3 powder at 50 tsi and sintering in a hydrogen atmosphere for 10 hr at 2750 F. The sintered compact was then crushed and screened to minus 100 plus 200 mesh.

An alternate method of incorporating a poison into a fuel element is by alloying it with the structural materials. It was decided to investigate this method as a backup effort to the work on the dispersed poison compound. The specimen type designated to investigate this alternate is Type 14 in which the side plates contain boron-10 alloyed in the stainless steel. The alloy was prepared by blending boron-10 and nickel powders, pressing, and sintering for 1/2 hr in a dry-hydrogen atmosphere at 1550 F. The compact was then arc melted to form NiB^{10} which was analyzed and added as the boron-10 source in an induction melt of Type 347 stainless steel. An analysis of the steel after forging and rolling to a thickness desirable for machining to frame thickness showed the boron-10 content to be 0.475 w/o.

Blending

A V-type mixer was used throughout the powder-blending steps. Sufficient powder to compact five specimens was blended at one time by first mixing the matrix phase with the burnable poison, if any, for 1 hr dry. The fuel was then added, and blending was continued for 1 hr. A dispersing-agent binder of 1/2 w/o methyl alcohol-camphor (Ceremul "C" was used for the green-pressed specimens, Type 12) was added to the

blend and the powders were mixed for the final hour. Caution was exerted in the handling and pressing of the blend to prevent any unnecessary disturbance to the blend which might result in a separation of the constituents into their respective density levels.

The suppressor sections in the two Type 8 specimens were blended in a similar manner to the fueled sections. The elemental-matrix powders were blended for 1 hr dry before the 1-hr blending in of the Eu_2O_3. The final hour of blending was done with the addition of 1/2 w/o methyl alcohol-camphor solution.

Pressing and Sintering

All specimens were compacted at 50 tsi in a double-acting die with the exception of Types 3 and 4. These two specimen types contain no boron and were pressed at 35 tsi previous to step sintering in hydrogen for 16 hr at 1600 F followed by 2 hr at 2250 F with an intermediate temperature increase of 170 F per hr. These highly porous cores were sintered over a long period of time to permit the hydrogen to flow through the pores in the cores and obtain an internal cleaning action in the cores. This procedure produces excellent results with cores pressed with elemental powders; however, there was insufficient improvement in the prealloyed-matrix core to warrant the longer, more expensive sintering procedure. Specimens of Type 4A were, therefore, pressed at 50 tsi and straight sintered 2 hr at 2000 F in hydrogen as were Type 1 and Type 2 specimens, which contained B_4C as the dispersed burnable poison.

The specimens containing the alternate burnable poison, NbB_2, were sintered for 2 hr at 2000 F in a vacuum of 1×10^{-4} mm of mercury, whereas, all specimens containing the reference poison, ZrB_2, were sintered for 2 hr at 2200 F in a vacuum of 1×10^{-4} mm of mercury.

All sintered specimens were coined at 50 tsi in the original double-acting die.

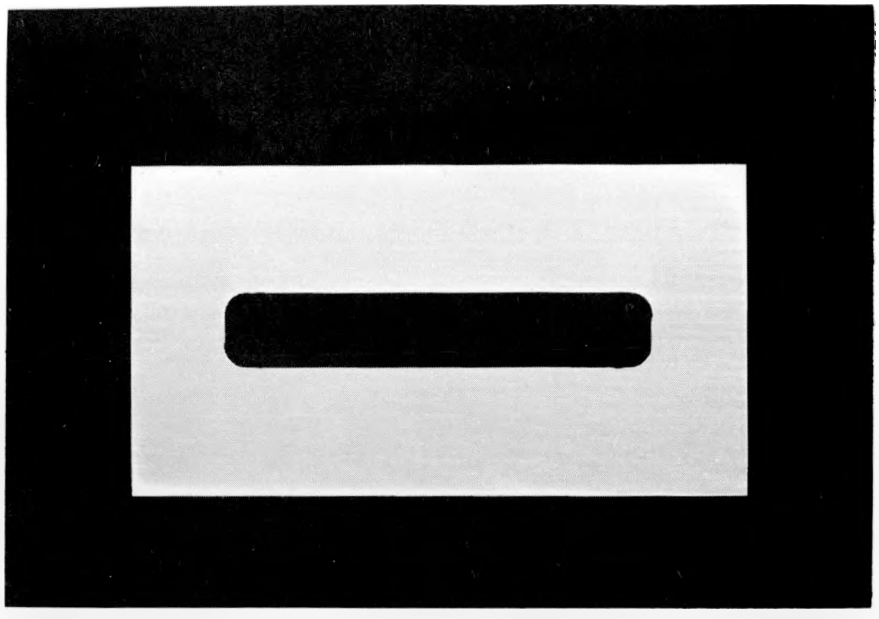
Preparation of Billets

Frames

All specimens were clad in a picture-frame-type billet of commercial Type 347 stainless steel. Five frame designs were used to contain the various types of specimens. Cover plates of the same commercial Type 347 stainless steel were prepared for each frame to give the desired core-cladding ratio of 5-30-5 after rolling. A 0.040-in.-thick cover plate was used on the standard 0.225-in.-thick frames. The frame for the SM-1 type specimen required a cover plate 0.074 in. thick to comply with the rolling schedule specified by ORNL[2] for the SM-1 fuel elements.

Figure 1 shows the frame as prepared for the SM-1 type specimens. This frame was designed to hold five specimens between spacers of Type 347 stainless steel. The over-all dimensions of the frame were 3-5/8 by 2 by 0.415 in. with cavity dimensions of 2-1/2 by 0.460 by 0.415 in.

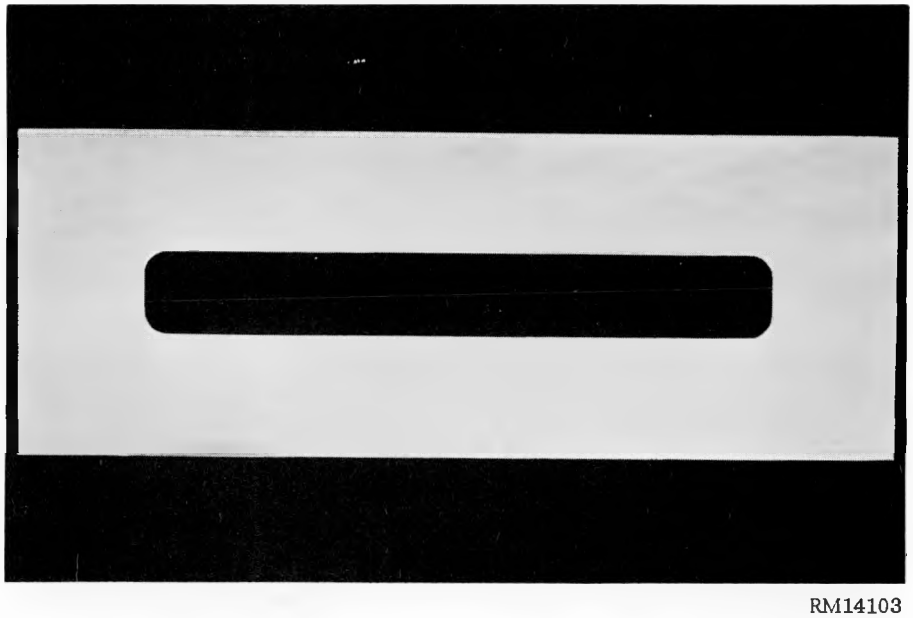
The standard frame used for the major portion of the specimens was of the same design, as shown in Figure 2, but was machined to different dimensions. This frame,



RM14104

FIGURE 1. TYPE 347 STAINLESS STEEL PICTURE FRAME USED IN FABRICATING SM-1 TYPE SPECIMENS

The frame dimensions are 3-5/8 by 2 by 0.415 in. with a cavity 2-1/2 by 0.460 in.



RM14103

FIGURE 2. SM-2 REFERENCE TYPE 347 STAINLESS STEEL PICTURE FRAME USED FOR FABRICATING IRRADIATION SPECIMENS

The frame dimensions are 5.167 by 1.990 by 0.225 in. with a cavity of 3.667 by 0.490 in.

with slight modifications in the cavity dimensions, was used for all specimens except Types 1, 12, and 14. The over-all dimensions were 5.167 by 1.990 by 0.225 in., with a cavity of 3.667 by 0.490. The frame cavity for the specimen types containing the high fuel loading was decreased in width to 0.440 in. to reduce the over-all fuel content yet maintain a higher fuel concentration for evaluation under irradiation conditions. Figure 3 shows a standard frame loaded with specimens and spacers prior to welding.

The specimens to be rolled in the green-pressed condition were inserted into a locked-spacer frame, as shown in Figure 4. This frame measured 4.75 by 2.50 by 0.225 in. and had specimen cavities of 0.168 by 0.490 in. This type of frame was found to be necessary because of the fragility of the green-pressed compacts. The locked spacers give more support and better dimensional tolerances to the less-cohesive-type specimen and eliminate much of the handling and fitting necessary with the movable-spacer frame.

The frame shown in Figure 5 was designed to incorporate the boron-10 stainless steel alloy in the specimen side cladding. The cover plates and end cladding were commercial Type 347 stainless steel

Cleaning, Assembly, and Welding

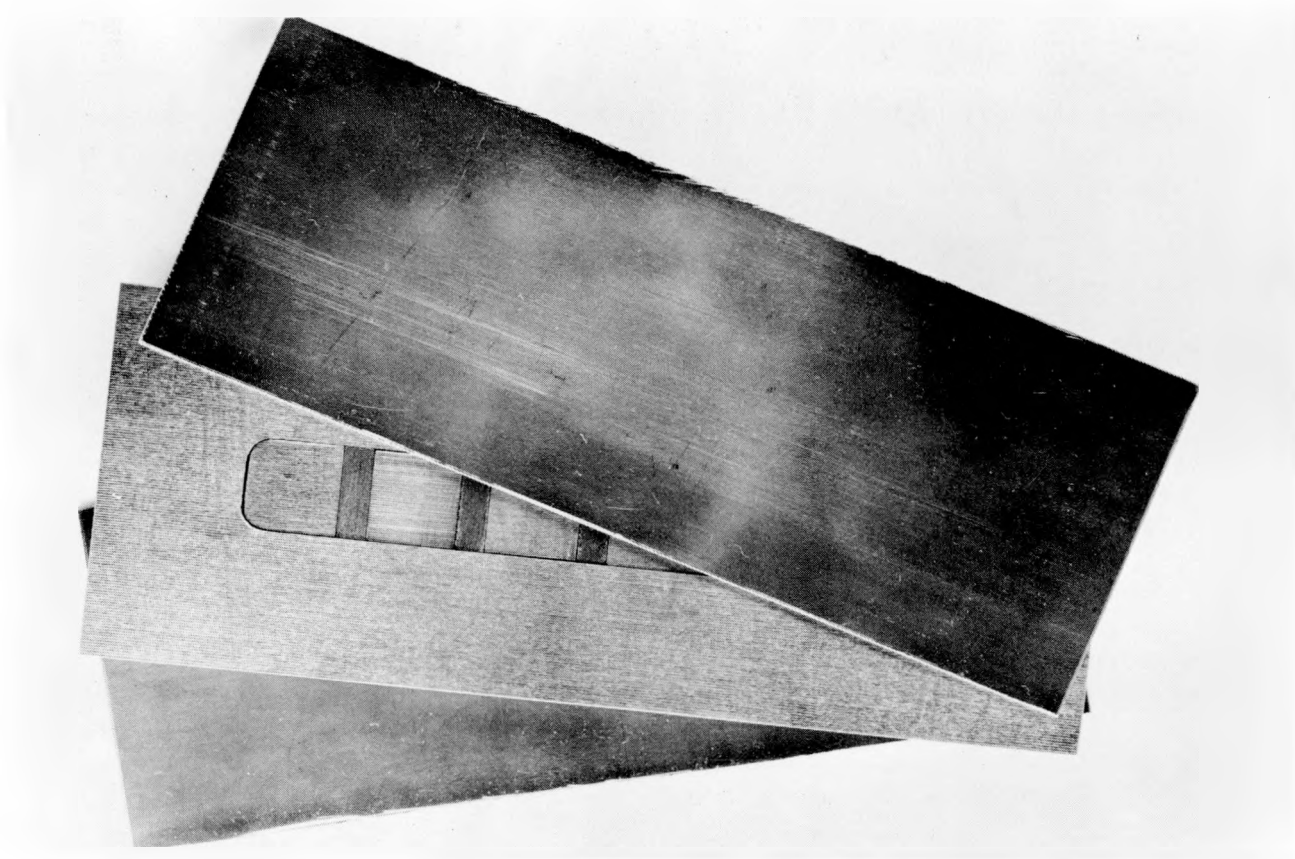
A standard cleaning technique was used for all frames, spacers, and cover plates. This procedure consisted of degreasing all metal surfaces with alcohol and steel wool prior to scrubbing in 180 F Alconox solution, followed by thoroughly rinsing in hot, cold, and hot water baths. The frames and cover plates were immediately dried with a lint-free absorbent as they were taken from the last hot rinse. Clean gloves were worn at all times during the cleaning and assembly operations.

The cores were given a light pass over with 600-grit paper immediately prior to assembly into their respective frames. Figure 3 shows an assembled billet of alternate spacers and cores just before the cover plates were clamped in place and the billet was inserted into an argon-filled dry box for welding. Figure 6 shows the billet after the cover plates and evacuation stem were welded in place. The billet was then leak checked, placed on an evacuation system, and degassed to a vacuum of 1×10^{-3} mm of mercury at 500 C and sealed by forging closed the evacuation tube.

Roll Cladding and Annealing

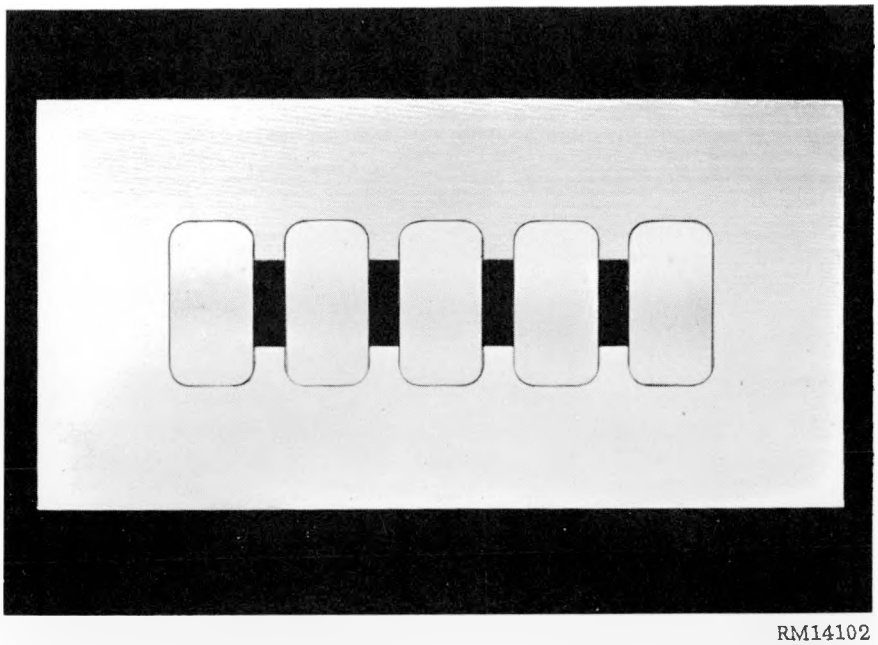
All billets were rolled from a hydrogen-atmosphere muffle following a 45-min preheat at temperature in accordance with the rolling schedule developed during the fabrication study. Each plate was turned and revolved 180 deg around the x-axis and y-axis after each roll pass and given a final anneal of 30 min in the muffle from which it was rolled. The plates containing the alternate dispersion poison, NbB_2, were rolled and annealed at a temperature of 2000 F. All others were hot rolled and annealed at 2200 F.

The rolling reductions for the Type 1 specimens followed the reference SM-1 rolling schedule specified by ORNL.[1] This started with a billet thickness of 0.563



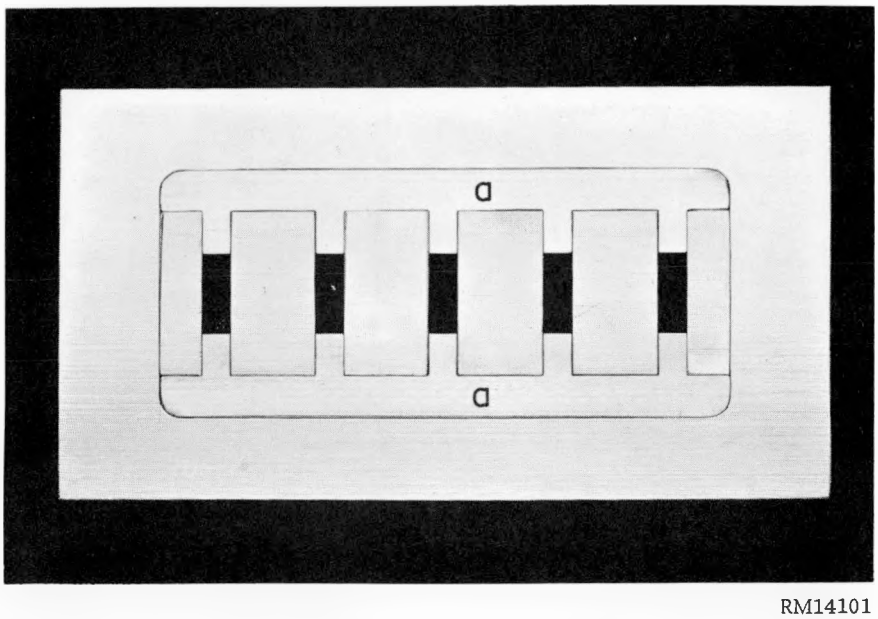
RM12839

FIGURE 3. STANDARD SM-2 TYPE IRRADIATION BILLET LOADED AND PREPARED FOR WELDING



RM14102

FIGURE 4. TYPE 347 STAINLESS STEEL FRAME FOR ROLL CLADDING SM-2 GREEN-PRESSED TYPE 12 IRRADIATION SPECIMENS



RM14101

FIGURE 5. PICTURE FRAME FOR ROLL CLADDING TYPE 14 SM-2 IRRADIATION SPECIMENS

Insert designated "a" is Type 347 stainless steel 0.475 w/o boron-10.

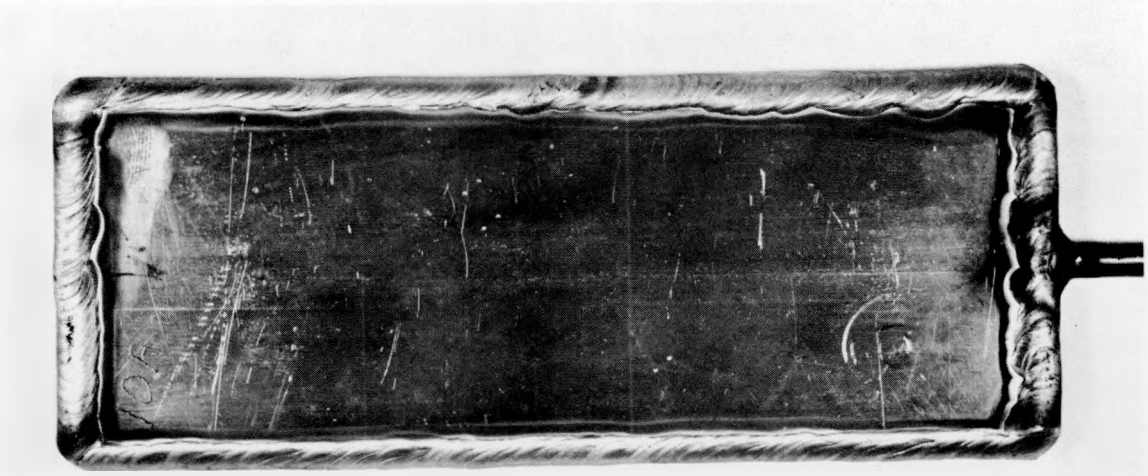


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FIGURE 6. ASSEMBLED AND WELDED SM-2 IRRADIATION-SPECIMEN BILLET

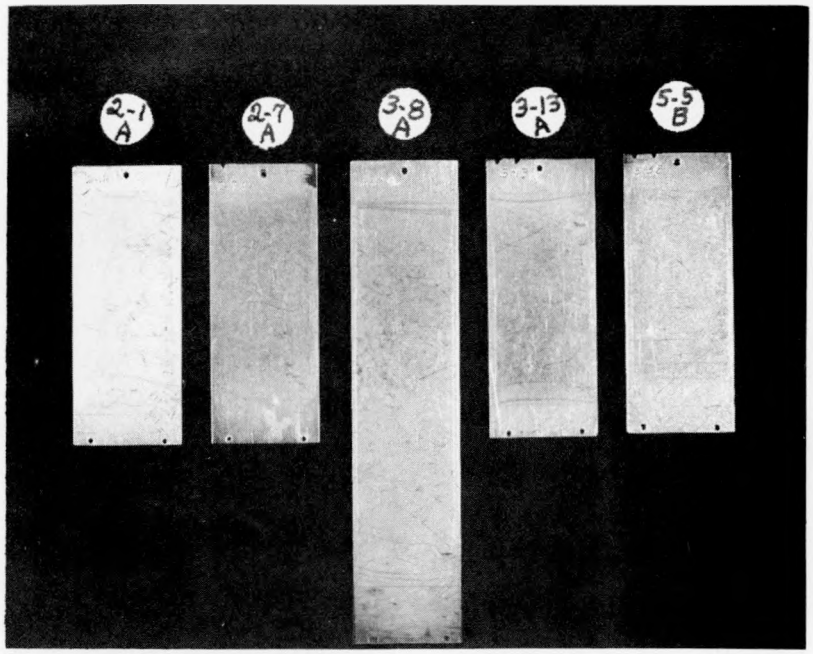

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FIGURE 7. PREIRRADIATION PHOTOGRAPH OF THE SM-2 SPECIMENS IRRADIATED IN CAPSULE BMI-32-9

and was reduced by hot rolling to 0.050 in. by reductions of 10, 10, 20, 40, 40, 40, and 40 per cent. After surface scale was removed by pickling, the plate was cold rolled to 0.040 in. and flat annealed in a dry-hydrogen atmosphere for 1 hr at 2050 F. The plate was protected from the jig surface during the flat annealing by Fiberfax paper.

The SM-2 type specimens were hot rolled from 0.305 to 0.050 with reductions in thickness of 40, 20, 20, 20, 20, and 20 per cent followed by a light reduction pass through the mill to give the desired 0.050 in. The surface scale was removed with a pickle of 88-10-2 parts water, HNO_3 and HF, respectively, and subsequently cold rolled to 0.040 in. to give a total reduction ratio of 7-1/2 in comparison to the SM-1 ratio of 14. The flat anneal on these specimens was also 1 hr at 2050 F in dry hydrogen with the plates separated by Fiberfax paper.

Machining

The five individual specimens were sheared from each plate with 1/2 in. excess stock on each edge. To assure positive identification a Vibratool number was made on the excess stock of each specimen prior to shearing. The specimens were then radiographed, and the film was used as a template in marking the exact location of the core and final specimen dimensions to insure proper alignment of the core in the specimen. The excess stock was sheared to within 1/16 in. of the final dimensions, and the specimens were radiographed. If any adjustments in centering the specimens were necessary, they were made previous to filing the specimens to final size. The final identification number was marked with a Vibratool in the upper left corner of each specimen after the final specimen size was outlined and before the original identifying number was sheared away with the excess stock. The specimen mounting holes were marked and center punched with the use of a template which was prepared with the use of a microscope to insure accurate location of the holes. The two alignment holes were placed at that end of the specimen proximal to the feathered end of the core. Figure 7 shows a photograph and Figure 8 shows the dimensions of the three types of specimens employed.

Inspection and Identification

Each machined specimen was radiographed to verify the proper centering of the cores in the specimen. The two best specimens of the five in each pack as judged from the radiograph and visual inspection were chosen for encapsulation with the third ranking specimen held as a backup and control. The fourth and fifth specimens were used as chemical-analysis specimens for boron content and for metallographic examinations.

The two specimens chosen for encapsulation were end notched for rapid identification in the respective capsule. The notch also insured identification should blistering or other radiation damage obliterate the Vibratool marking. These specimens were leak tested by boiling for three 1/2-hr periods in standard solutions of 50-50 HNO_3-H_2O. The three solutions from each specimen were then analyzed for uranium content in an effort to detect any defects in cladding. The second and third pickles remain constant to the standard solution if no defects are present.

Both sides of the specimens to be encapsulated were photographed at 1X and 4X magnifications as a final record of identification and preirradiation appearance.

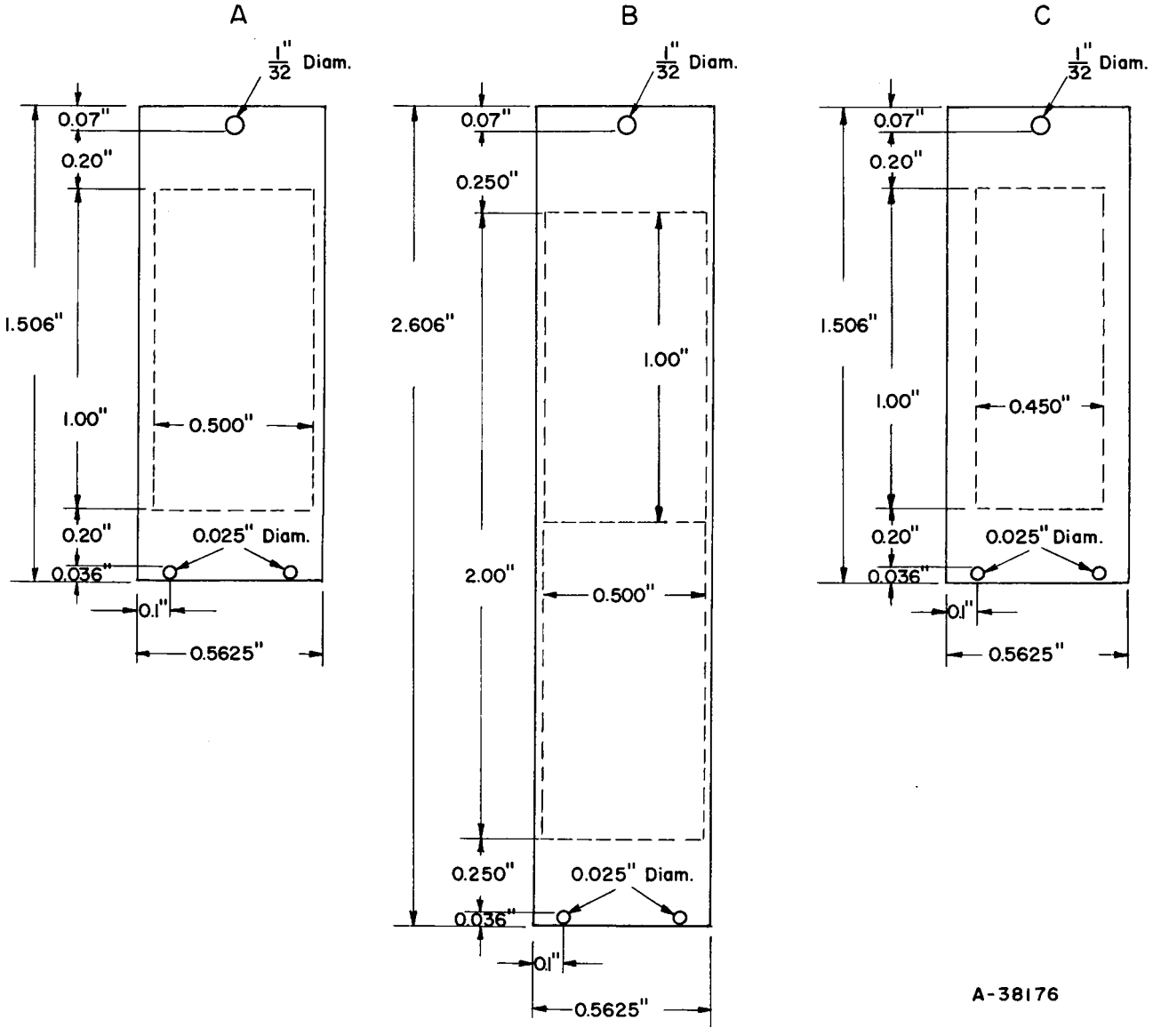


FIGURE 8. DRAWINGS OF SPECIMENS IRRADIATED IN THE SM-2 CORE MATERIALS DEVELOPMENT PROGRAM

A Specimen containing standard fuel loading
B Specimen containing standard fuel loading combined with suppressor section on bottom-Type 8
C Specimen containing increased fuel loading-Types 15, 16, 17.

TABLE 2. FUEL AND POISON CONTENTS OF SM-2 IRRADIATION SPECIMENS

Specimen Identification				Fuel				Poison		Core
Capsule	No.	Type	Notches	Type	Loading, w/o	Weight, g	Uranium-235, g	Type	Boron, w/o	Weight, g
BMI-32-1	2-7V	7	1	Spherical UO_2	26	0.514	0.421	ZrB_2	0.095	1.9780
	1-2-2	2	0	Hydrate UO_2	26.1	0.515	0.426	B_4C	0.192	1.9736
	2-5V	5	2	Hydrate UO_2	26	0.514	0.425	ZrB_2	0.199	1.9788
	2-9	9	3	Hydrate UO_2	26	0.513	0.424	NbB_2	0.209	1.9740
	5-3	3	4	Hydrate UO_2	26	0.514	0.425	None	--	1.9780
	5-4	4	5	Spherical UO_2	26	0.515	0.422	None	--	1.9790
	5-8V	8	(a)	Spherical UO_2	26	0.513	0.421	ZrB_2	0.240	1.9720
BMI-32-2	2-4-14	14	0	Spherical UO_2	25.9	0.515	0.422	B^{10}	(b)	1.9870
	2-2-2	2	1	Hydrate UO_2	26.1	0.516	0.427	B_4C	0.192	1.9764
	3-3	3	2	Hydrate UO_2	26	0.514	0.425	None	--	1.9775
	3-9	9	3	Hydrate UO_2	26	0.478	0.395	NbB_2	0.209	1.8400
	8-13	13	4	Spherical UO_2	26	0.514	0.421	NbB_2	0.202	1.9780
	5-5V	5	5	Hydrate UO_2	26	0.514	0.425	ZrB_2	0.199	1.9777
	5-1	1	6	Hydrate UO_2	24.2	0.476	0.394	B_4C	0.021	1.9670
	4-10V	10	7	Spherical UO_2	26	0.514	0.421	ZrB_2	0.192	1.9780
BMI-32-3	3-13	13	1	Spherical UO_2	26	0.512	0.420	NbB_2	0.204	1.9700
	3-7V	7	2	Spherical UO_2	26	0.514	0.421	ZrB_2	0.095	1.9770
	2-3-14	14	0	Spherical UO_2	25.9	0.515	0.422	B^{10}	(b)	1.9894
	1-1	1	3	Hydrate UO_2	24.2	0.474	0.392	B_4C	0.021	1.9592
	4-4	4	4	Spherical UO_2	26	0.514	0.421	None	--	1.9772
	2-10V	10	5	Spherical UO_2	26	0.514	0.421	ZrB_2	0.192	1.9760
	1-8V	8	(a)	Spherical UO_2	26	0.514	0.421	ZrB_2	0.240	1.9786
BMI-32-4	1-12V	12	0	Spherical UO_2	26	0.595	0.488	ZrB_2	0.209	2.29
	2-3	3	1	Hydrate UO_2	26	0.575	0.475	None	--	1.9790
	4-7A	7	2	Spherical UO_2	26	0.514	0.421	ZrB_2	0.093	1.9770
	3-4	4	3	Spherical UO_2	26	0.516	0.423	None	--	1.9852
	2-6V	6	4	Spherical UO_2	26	0.514	0.421	ZrB_2	0.213	1.9780
	4-11	11	5	Hydrate UO_2	26	0.514	0.425	NbB_2	0.207	1.9780
BMI-32-5	4-5B	5	3	Hydrate UO_2	26	0.510	0.422	ZrB_2	0.197	1.9630
	2-6A	6	5	Spherical UO_2	26	0.514	0.421	ZrB_2	0.200	1.9760
	5-11	11	1	Hydrate UO_2	26	0.514	0.425	NbB_2	0.207	1.9750
	2-13	13	2	Spherical UO_2	26	0.513	0.421	NbB_2	0.203	1.9220
	2-15B	15	4	UN	34.1	0.661	0.582	ZrB_2	0.315	1.9390
	5-17B	17	0	Spherical UO_2	38.4	0.710	0.582	ZrB_2	0.350	1.8476
BMI-32-6	3-1A	1	3	Hydrate UO_2	24.18	0.473	0.391	B_4C	0.048	1.9550
	2-4A	4	2	Spherical UO_2	26	0.514	0.421	None	--	1.9750
	3-9A	9	1	Hydrate UO_2	26	0.505	0.418	NbB_2	0.222	1.9422
	4-13A	13	0	Spherical UO_2	26	0.512	0.420	NbB_2	0.204	1.9698
	3-15B	15	5	UN	34.1	0.662	0.583	ZrB_2	0.315	1.9410
	1-16B	16	4	Hydrate UO_2	38.4	0.703	0.581	ZrB_2	0.430	1.8320
BMI-32-7	4-1A	1	5	Hydrate UO_2	24.18	0.475	0.393	B_4C	0.048	1.9630
	3-4A	4	0	Spherical UO_2	26	0.514	0.421	None	--	1.9760
	3-5C	5	4	Hydrate UO_2	26	0.514	0.425	ZrB_2	0.183	1.9774
	3-6V	6	3	Spherical UO_2	26	0.514	0.421	ZrB_2	0.213	1.9776
	2-16B	16	2	Hydrate UO_2	38.4	0.697	0.576	ZrB_2	0.430	1.8160
	3-17B	17	1	Spherical UO_2	38.4	0.710	0.582	ZrB_2	0.350	1.8500

TABLE 2. (Continued)

Specimen Identification				Fuel				Poison		Core
Capsule	No.	Type	Notches	Type	Loading, w/o	Weight, g	Uranium-235, g	Type	Boron, w/o	Weight, g
BMI-32-8	1-1A	1	1	Hydrate UO_2	24.18	0.476	0.394	B_4C	0.048	1.9678
	1-4A	4	0	Spherical UO_2	26	0.514	0.421	None	--	1.9768
	7-13	13	5	Spherical UO_2	26	0.514	0.421	NbB_2	0.202	1.9770
	2-14A	14	2	Spherical UO_2	26	0.552	0.452	B^{10}	(b)	2.1212
	1-8A	8	(a)	Spherical UO_2	26	0.510	0.418	ZrB_2	0.222	1.9610
BMI-32-9	2-1A	1	0	Hydrate UO_2	24.18	0.476	0.394	B_4C	0.048	1.9668
	5-5B	5	2	Hydrate UO_2	26	0.513	0.424	ZrB_2	0.197	1.9742
	2-7A	7	1	Spherical UO_2	26	0.515	0.422	ZrB_2	0.093	1.9790
	3-13A	13	3	Spherical UO_2	26	0.512	0.420	NbB_2	0.204	1.9691
	3-8A	8	(a)	Spherical UO_2	26	0.513	0.421	ZrB_2	0.222	1.9713
BMI-32-10	4-3	3	3	Hydrate UO_2	26	0.512	0.423	None	--	1.9680
	4-5C	5	1	Hydrate UO_2	26	0.514	0.425	ZrB_2	0.183	1.9772
	1-6A	6	0	Spherical UO_2	26	0.514	0.421	ZrB_2	0.200	1.9780
	2-9A	9	2	Hydrate UO_2	26	0.512	0.423	NbB_2	0.222	1.9704
	1-12C	12	4	Spherical UO_2	26	0.49	0.402	ZrB_2	0.1684	1.9
	3-14A	14	5	Spherical UO_2	26	0.553	0.453	B^{10}	(b)	2.1256

(a) This specimen is easily recognized by its increased length. It was, therefore, not notched.

(b) Boron-10 was alloyed in the stainless steel frame. An analysis of the entire specimen showed a boron content of 0.023 w/o.

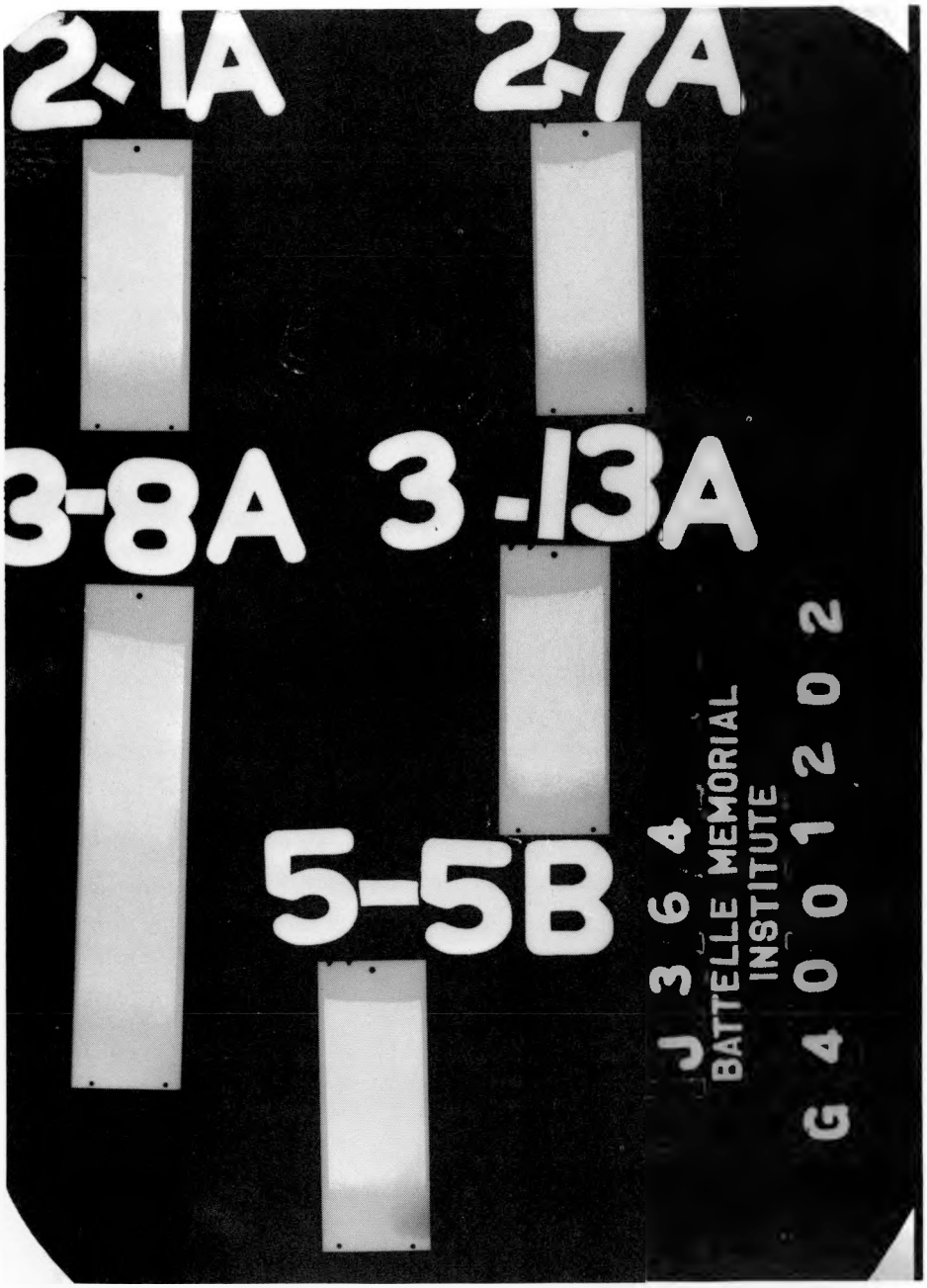


FIGURE 9. PREIRRADIATION RADIOGRAPH OF A CAPSULE LOADING OF SM-2 IRRADIATION SPECIMENS

Specimens were irradiated in Capsule BMI-32-9 in the ETR.

Table 2 lists the fuel and poison content of each specimen and the capsule in which it was irradiated. Figures 7 and 9 are a typical photograph and radiograph, respectively, of a capsule loading of specimens.

CAPSULE DESIGN AND SPECIMEN ENCAPSULATION

The capsules employed in this irradiation were designed to maintain nominal specimen-surface temperatures of 600 F during irradiation in reactor unperturbed thermal-neutron fluxes of 3 to 4 x 10^{14} nv, and gamma-heating rates of 5 w per g. Uninstrumented Capsules BMI-32-1, BMI-32-2, and BMI-32-3 contained seven, eight, and seven plate-type specimens, respectively, immersed in NaK with a split copper-block insert adjacent to, but not touching them. The various high-thermal-conductivity components in the design, i.e., the copper inserts and the nickel water-contacting outer shell, were selected for the purpose of dissipating the large quantities of fission heat generated while maintaining the relatively low specimen-surface temperature. The components of the uninstrumented capsules are shown in Figure 10.

Capsule BMI-32-4, which was irradiated in the ETR, was basically similar to the MTR capsules except that its six specimens were contained in two separate axial compartments. The upper compartment contained four specimens and this zone was instrumented with auxiliary electric heaters and thermocouples; the lower compartment was not instrumented. Component parts of this capsule are shown in Figure 11.

The specimens in all four capsules were suspended on hangers which fitted into grooves between the two copper blocks. The lower edges of the specimens were fitted with small wire rings through the cladding frames to keep them from contacting the copper blocks. A more detailed analysis of the capsule design and irradiations, including results of nuclear mock-up experiments, is reported in a separate topical report.(3)

Table 3 presents a résumé of the in-pile histories of the four capsules involved. The MTR irradiations progressed normally except that during Cycles 129 and 130 several capsules, including the three in the BMI-32 series, were discharged from the reactor while the source of some fission-product activity in the reactor coolant was being pinpointed.

During the irradiation of Capsule BMI-32-4, several thermocouple and heater failures occurred. The temperature data which were procured indicated that the surface temperature of the peak-flux specimen (1-12V) was close to 600 F throughout the exposure. The data are treated in detail in BMI-1516.

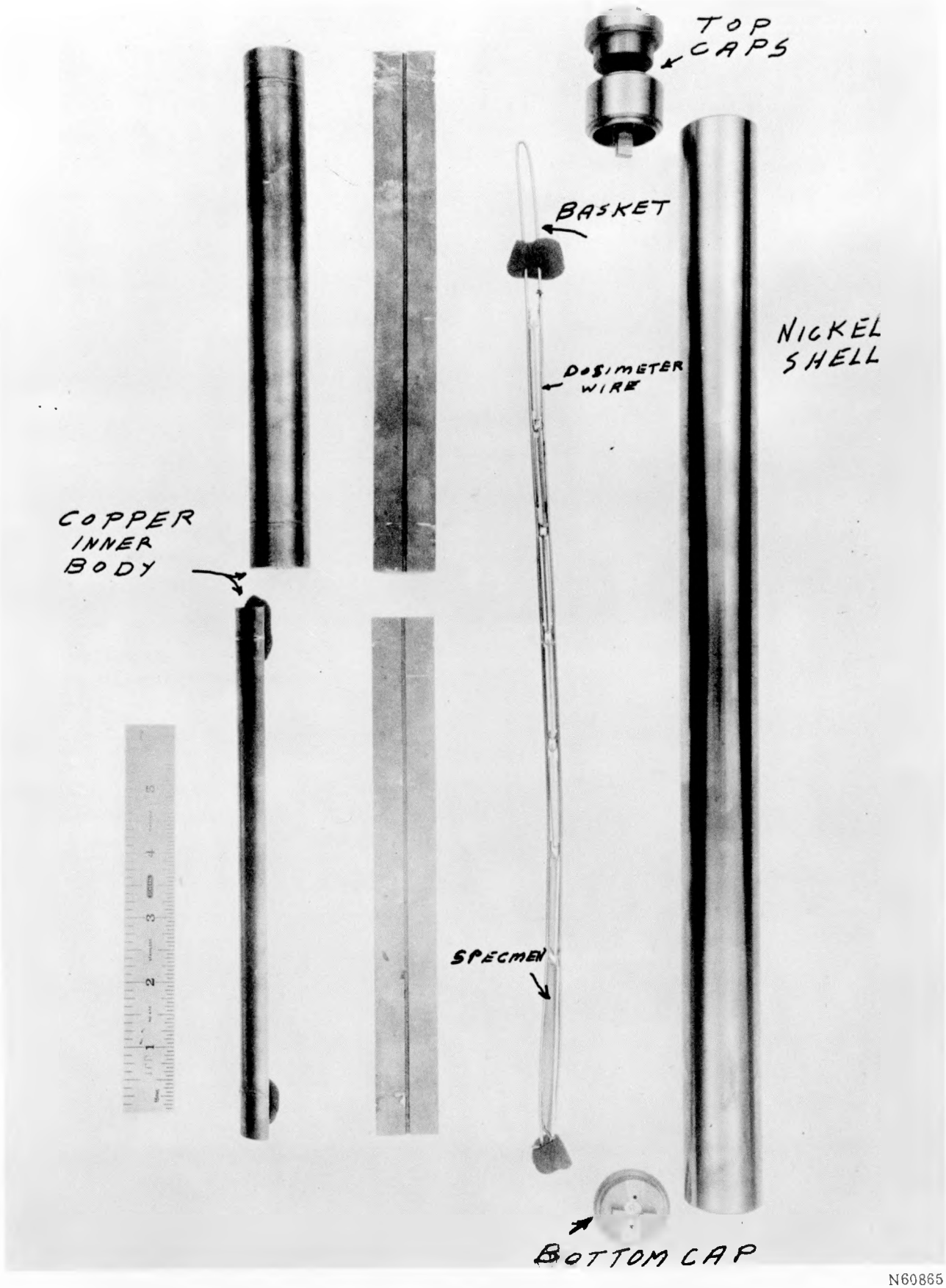


FIGURE 10. COMPONENTS OF ONE OF THE UNINSTRUMENTED CAPSULES IRRADIATED IN THE MTR

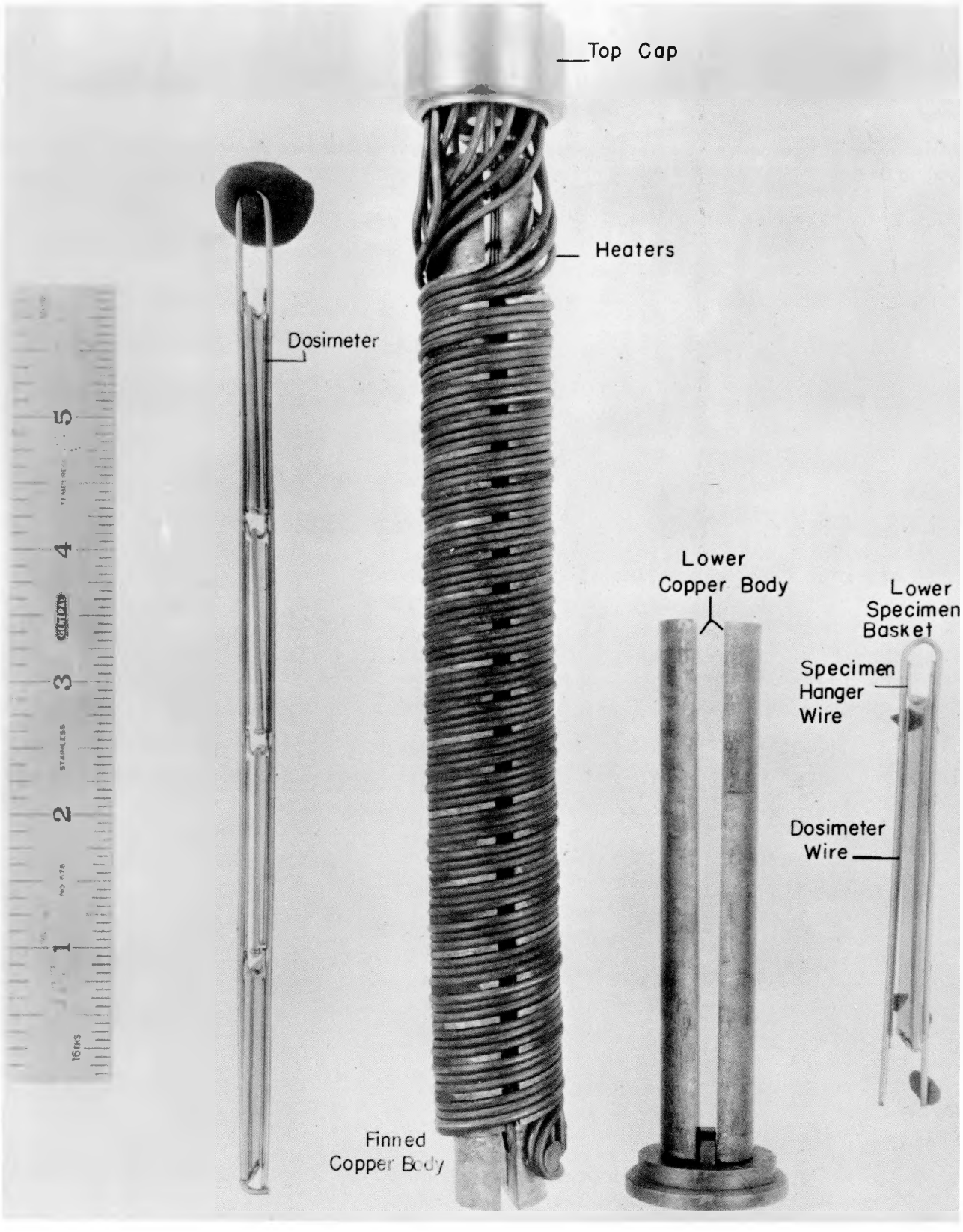


FIGURE 11. COMPONENTS OF INSTRUMENTED CAPSULE IRRADIATED IN THE ETR

Capsule BMI-32-4 is shown here.

TABLE 3. RÉSUMÉ OF IN-PILE HISTORY FOR CAPSULES BMI-32-1, BMI-32-2, BMI-32-3, AND BMI-32-4

Capsule	Irradiation Position	Loading and Discharge Data for Capsules		Peak Reactor-Quoted Unperturbed Flux, 10^{14} nv	Length of Irradiation, days
		Loaded (1959)	Discharged (1960)		
BMI-32-1	MTR-L-53	Cycle 126 (August 10)	Cycle 137 (March 28)	4.2	150
BMI-32-2	MTR-L-56	Cycle 127 (August 31)	Cycle 137 (March 28)	4.6	140
BMI-32-3	MTR-L-58	Cycle 128 (September 21)	Cycle 137 (March 28)	4.1	117
BMI-32-4	ETR-O-6	Cycle 20 (September 8)	Cycle 28 (April 4)	2.8	86

All four capsules were shipped from NRTS in Cask HC-12, arriving at the BMI Hot-Cell Facility on April 18, 1960. The cask was immediately opened and post-irradiation examination of the capsules was initiated in order that results of the test would be rapidly made available. The examination included measurements of the quantity of fission gas released from the specimens, visual inspection of the capsule components and specimens, measurements of specimen density and dimensions, analysis of dosimeter wires and analytical determination of fuel burnup, metallographic examination of selected specimens, and postirradiation heat treatment of selected specimens. Experimental details and the results of each phase of the postirradiation examination are discussed separately below.

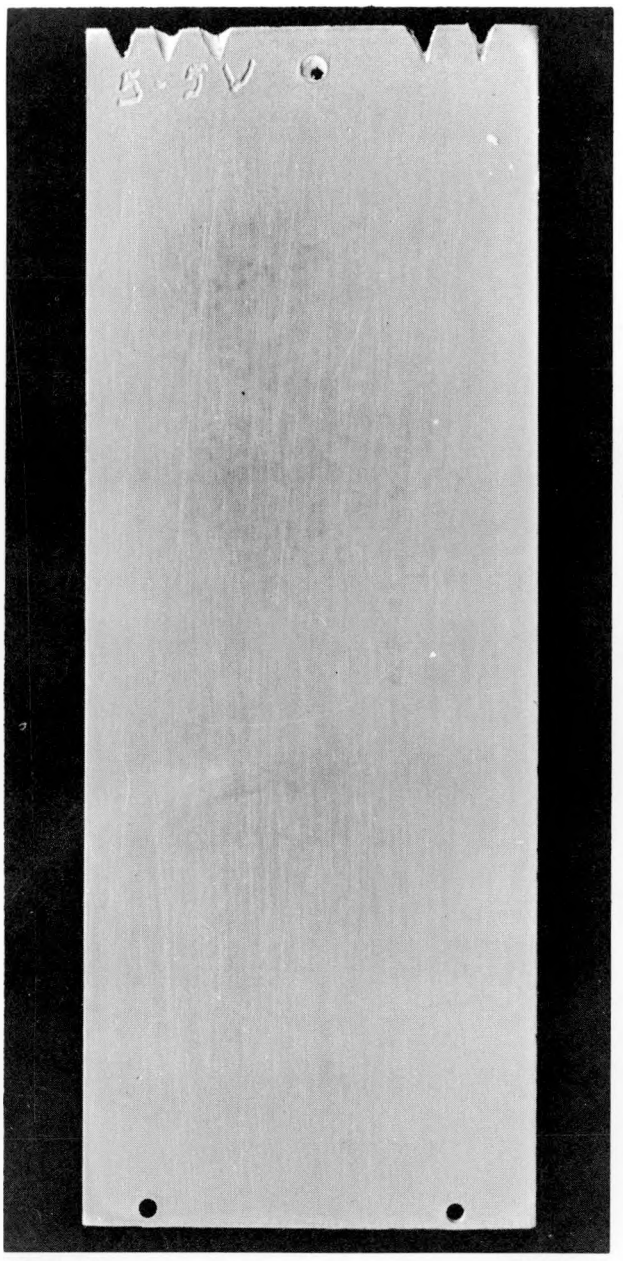
POSTIRRADIATION EXAMINATION OF SPECIMENS

Capsule Opening and Fission-Gas Sampling

It was necessary to grind the bottom surface of the capsules in order to remove dents and insure a good vacuum seal for fission-gas puncturing. Capsule BMI-32-2 and both compartments of BMI-32-4 were punctured and samples of the gas were obtained. When BMI-32-1 and BMI-32-3 were punctured, internal gas pressure forced NaK from the punch hole. Subsequently, it was noticed that these two capsules contained failed specimens and measurable quantities of fission gas.

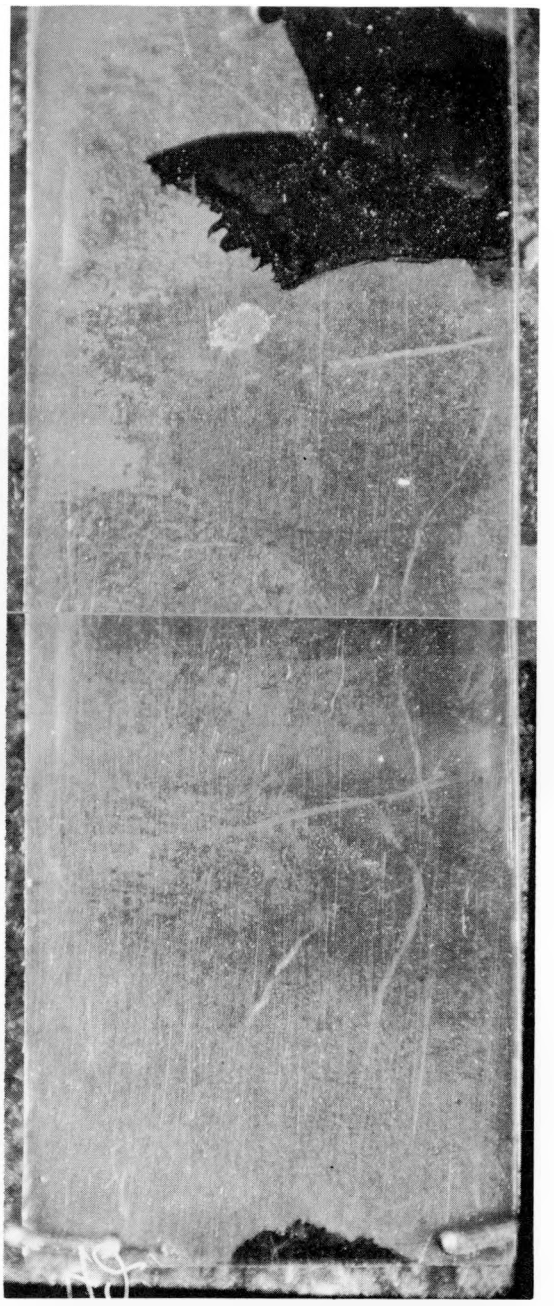
Visual Examination

All specimens were visually examined and photographed at magnifications of 4, 6, or 12X upon removal from the capsule. All specimens appeared in good physical condition, as shown by the typical specimens in Figures 12 and 13, with the exception of one from Capsule BMI-32-1 and two from Capsule BMI-32-3. Because the temperature history of these three specimens is considered unique, they will be discussed in greater detail in a separate section of this report.



About 4X RM12681

a. Before Irradiation



About 4-1/2X HC4903 and HC4904

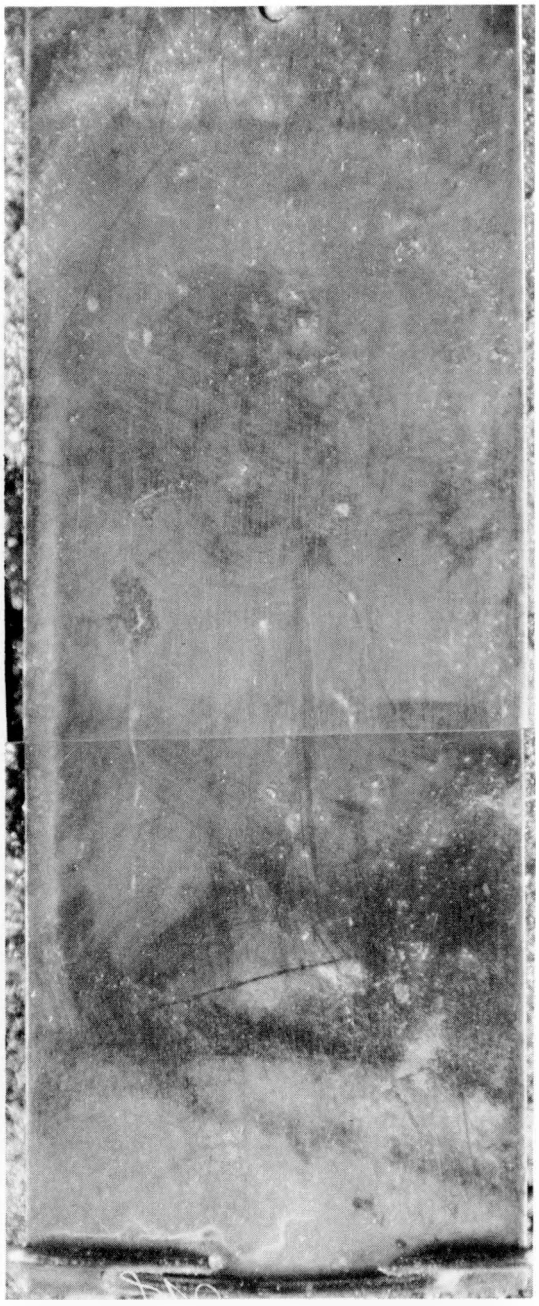
b. After Irradiation

FIGURE 12. EFFECT OF IRRADIATION ON THE APPEARANCE OF SPECIMEN 5-5V FROM CAPSULE BMI-32-2

This specimen was irradiated to about 45 uranium a/o, sustaining a 2.9 per cent density decrease.

4-1/2X HC4899 and HC4900

a. Specimen 4-10V from Capsule BMI-32-2



About 4-1/2X HC4866 and HC4867

b. Specimen 1-12V from Capsule BMI-32-4

FIGURE 13. APPEARANCE OF TWO ADDITIONAL SPECIMENS AFTER IRRADIATION

Both specimens were irradiated to a burnup of approximately 40 a/o uranium and both underwent a density decrease of 2.2 per cent.

Neutron Dosimetry

Neutron dosimeters fabricated from 0.020-in.-diameter nickel-0.6 w/o cobalt wire were irradiated in positions adjacent to the faces of each specimen. The dosimeter wires were removed, identified, and radiochemically analyzed to determine the quantity of cobalt-60 formed during the irradiation. Because of the difficulties encountered in opening some of the capsules, the identifications of some of the dosimeters were lost. These were not analyzed.

The neutron flux derived from the dosimeter-wire analyses was translated to specimen-effective flux by use of the semiempirical method of W. B. Lewis.[4] The uncertainties associated with this approach to specimen attenuation are difficult to appraise, although a common approach is to assign to it a ±15 per cent limit. The dosimetry-derived burnup values (reported in Table 4) were obtained by the equation

$$\beta = \frac{\sigma_f}{\sigma_a}\left(1-e^{-\phi\sigma_a t}\right) 100 \ , \qquad (1)$$

where

σ_f = cross section for fission = 5.90×10^{22} cm^2

σ_a = adsorption cross section = 6.96×10^{22} cm^2

ϕ = neutron flux

t = time of irradiation.

Burnup Determination by Isotopic and Cesium Analyses

Twelve samples were analyzed by radiochemical and mass spectrometric techniques to determine fuel burnup. These analyses were performed by the Phillips Petroleum Company Chemical Processing Plant. The samples were selected from specimens receiving detailed metallographic examination and included a minimum of two specimens from each capsule. For the most part, the samples were transverse cross-sectional slabs; hence, the values obtained were the average of the high-burnup specimen edges as well as the lower burnup central portions of the samples.

Results of the analyses included uranium-234, -235, -236, and -238 contents of both irradiated samples and unirradiated samples included for control purposes. The isotopic burnup values appearing in Table 4 were calculated by the following equation which is based principally on the change in uranium-235 content. The derivation of the equation takes into account changes in uranium-236 concentration but not changes in the concentration of uranium-234 or -238. In the case of highly enriched specimens, changes in the uranium-238 concentration can be ignored:

$$F = \frac{(1+\alpha)(E_o - E_1)}{E_o(1+\alpha-E_1)} \ , \qquad (2)$$

TABLE 4. SUMMARY OF BURNUP DATA FOR SPECIMENS IRRADIATED IN CAPSULES BMI-32-1, BMI-32-2, BMI-32-3, AND BMI-32-4

Specimen	Burnable Poison in Core[a]		Total Uranium-235 Depletion, per cent of Original Uranium-235 Atoms Present			
	Boron, w/o	Compound	Predicted From Nuclear Mock-Up Data and MTR Flux Estimates	Estimated From Capsule Dosimetry	Obtained From Isotopic Analyses[b]	Obtained From Radiochemical Analyses
Capsule BMI-32-1 (MTR Position L-53)						
2-7V (top)	0.095	ZrB_2	65	--	39.1 ± 0.3	37
1-2-2	0.192	B_4C	67	--	--	
2-5V	0.199	ZrB_2	68	--	--	
2-9	0.209	NbB_2	70	--	--	
5-3	None	--	72	--	44.5 ± 0.3	47
5-4	None	--	72	45	44.0 ± 0.3	45
5-8V (bottom)[c]	0.240	ZrB_2	--	37	40.0 ± 0.3	36
Capsule BMI-32-2 (MTR Position L-56)						
2-4-14 (top)	0.022	B^{10} (cladding)	63	44	--	
2-2-2	0.192	B_4C	67	--	45.4 ± 0.3	56
3-3	None	--	69	--	--	
3-9	0.209	NbB_2	72	50	--	
8-13	0.202	NbB_2	74	--	--	
5-5V	0.199	ZrB_2	73	44	44.3 ± 0.3	54
5-1	0.021	B_4C	71	--	--	
4-10V (bottom)	0.192	ZrB_2	68	--	36.4 ± 0.3	40
Capsule BMI-32-3 (MTR Position L-58)						
3-13 (top)	0.204	NbB_2	56	32	35.1 ± 0.3	39
3-7V	0.095	ZrB_2	58	34	--	
2-3-14	0.022	B^{10} (cladding)	61	37	37.4 ± 0.3	42
1-1	0.021	B_4C	62	36	--	
4-4	None	--	61	34	--	
2-10V	0.192	ZrB_2	57	30	35.1 ± 0.3	31
1-8V (bottom)[c]	0.240	ZrB_2	--	27	--	
Capsule BMI-32-4 (ETR Position O-6						
1-12V (top)	0.209	ZrB_2	40	38	39.5 ± 0.3	42
2-3	None	--	40	34*	--	
4-7A	0.093	ZrB_2	39	31*	--	
3-4	None	--	35	28	--	
2-6V	0.213	ZrB_2	27	23	--	
4-11 (bottom)	0.207	NbB_2	22	19	20.7 ± 0.3	24

(a) The Type 347 stainless steel-clad cores contained the nominal 26 w/o UO_2 (highly enriched) dispersed in Type 347 stainless steel.

(b) Limits are expressed with 95 per cent confidence.

(c) These were double-length specimens in which a UO_2-fueled core and a Eu_2O_3 (suppressor material) core were both present. Each core was of standard size and, in the fabrication process, laid end to end and clad. Because of their high poison content, no attempt was made to predict their burnups.

where

F = fractional total burnup of uranium-235

α = capture-to-fission ratio; in the above specimens, this ratio varied between 0.195 and 0.206

E_o = fractional uranium-235 content of unirradiated material

E_1 = fractional uranium-235 content of irradiated material.

It is believed that isotopic burnup analyses are quite reliable, particularly in the case of high-burnup specimens. The uncertainty associated with burnup limits as reported is regarded as less than 1 per cent with a 95 per cent confidence.

The cesium analyses received at Battelle, also shown in Table 4, consisted of the uranium and cesium concentrations for dissolution samples in units of mg per ml and dpm per ml, respectively. The burnup formula applied in this case is as follows:

$$\beta = \frac{(1 + \alpha)}{A_o^{25}} \frac{N_{cs}}{N_{cs} + N_u Y}, \qquad (3)$$

where

β = fractional total burnup of uranium-235

N_{cs} = concentration of cesium-137 in sample solution, atoms per ml

N_u = concentration of total uranium in sample solution, atoms per ml

Y = fission yield for cesium-137, atoms per fission

A_o^{25} = atom fraction of uranium-235 in preirradiation sample.

The fission yield and the half-life of cesium-137 appearing in this equation hold some uncertainty. Values utilized are 0.0615 and 28.6 years, respectively, as recommended by an ASTM committee on fissionable-materials burnup. Probably, the combination of these uncertainties is in the ±15 per cent range.

Measurements of Physical Dimensions and Density

The physical dimensions of each irradiated specimen, with the exception of 1-8V, were obtained with vernier micrometers. Neither density nor dimensional measurements were made on Specimen 1-8V because of its badly fused and ruptured condition. Moreover, the specimen was broken into two sections during handling. The values for the remainder of the specimens are reported in Table 5. They represent 16 to 24 thickness measurements, 8 to 12 width measurements, and 4 length measurements.

The apparent density of each intact specimen was measured by weight difference in carbon tetrachloride at room temperature. These measurements, also reported in Table 5, are accurate to ±0.03 g per cm^3. Considering the burnups to which the specimens were subjected, the changes in density and dimensions of all except blistered specimens were small.

TABLE 5. POSTIRRADIATION DATA FOR CAPSULES BMI-32-1, BMI-32-2, BMI-32-3, AND BMI-32-4

Capsule	Specimen	Fuel		Burnable Poison		Estimated Uranium Burnup[a], a/o		Dimensional Change, per cent			Density Decrease, per cent
		Type of UO_2	Loading, w/o	Type	Boron, w/o	From Capsule Dosimetry	From Isotopic Analysis	Length	Width	Thickness	
BMI-32-1	2-7V	Spherical	26	ZrB_2	0.095	--	39.1	--	Blistered	--	--
	1-2-2	Hydrate	26.1	B_4C	0.192	--	--	0.07	0.09	5.3	1.9
	2-5V	Hydrate	26	ZrB_2	0.199	--	--	0.15	0.07	6.0	2.2
	2-9	Hydrate	26	NbB_2	0.209	--	--	0.79	0.19	2.6	2.7
	5-3	Hydrate	26	None	--	--	44.5	0.11	0.09	4.8	2.5
	5-4	Spherical	26	None	--	45	44.0	0.04	0.09	4.5	2.5
	5-8V[b]	Spherical	26	ZrB_2	0.240	37	40.0	--	--	UO_2-SS 5.5 Eu_2O_3-SS 0.0	1.4
BMI-32-2	2-4-14	Spherical	25.9	B^{10}	(c)	44	--	0.17	0.34	5.0	1.4
	2-2-2	Hydrate	26.1	B_4C	0.192	--	45.4	--	0.12	6.3	2.7
	3-3	Hydrate	26	None	--	--	--	0.17	0.38	5.5	2.9
	3-9	Hydrate	26	NbB_2	0.209	50	--	--	0.35	5.4	2.6
	8-13	Spherical	26	NbB_2	0.202	--	--	0.15	0.17	5.8	3.1
	5-5V	Hydrate	26	ZrB_2	0.199	44	44.3	--	0.14	6.6	2.9
	5-1	Hydrate	24.2	B_4C	0.021	--	--	0.19	--	4.8	2.4
	4-10V	Spherical	26	ZrB_2	0.192	--	36.4	0.09	0.04	4.2	2.2
BMI-32-3	3-13	Spherical	26	NbB_2	0.204	32	35.1	0.26	0.21	4.2	2.0
	3-7V	Spherical	26	ZrB_2	0.095	34	--	--	0.14	4.5	1.8
	2-3-14	Spherical	25.9	B^{10}	(c)	37	37.4	0.36	0.19	3.6	1.7
	1-1	Hydrate	24.2	B_4C	0.021	36	--	0.75	0.86	3.9	1.9
	4-4	Spherical	26	None	--	34	--	0.24	0.09	3.6	1.8
	2-10V	Spherical	26	ZrB_2	0.192	30	35.1		Blistered - fused		--
	1-8V[b]	Spherical	26	ZrB_2	0.240	27	--		Blistered - fused		--
BMI-32-4	1-12V	Spherical	26	ZrB_2	0.209	38	39.5	--	0.12	6.4	2.2
	2-3	Hydrate	26	None	--	34	--	0.09	0.09	2.9	2.1
	3-4	Spherical	26	None	--	31	--	0.05	0.09	3.6	1.6
	2-6	Spherical	26	ZrB_2	0.213	28	--	0.05	0.09	5.5	1.8
	4-7A	Spherical	26	ZrB_2	0.093	23	--	0.12	0.23	1.7	1.4
	4-11	Hydrate	26	NbB_2	0.207	19	20.7	0.86	0.31	2.7	0.9

(a) Burnups estimated from capsule dosimetry and from actual isotopic analysis.

(b) Specimen incorporates stainless-Eu_2O_3 suppressor.

(c) Boron-10 alloyed in stainless steel picture frame. Content 0.023 w/o boron.

Fission-Gas Analysis

Samples of gas collected from the capsules were analyzed both by mass and gamma-ray spectrographic techniques. Neither krypton-85 nor xenon-133 activity was detected in Capsule BMI-32-2, or in either of the two compartments of BMI-32-4. These capsules did not contain ruptured specimens. Quantities as small as 1.9×10^{-2} microcurie of krypton-85 could have been detected by the gamma-ray spectrometer. This quantity would correspond to about 4.8×10^{-10} cm^3 of fission gas at STP.

Approximately 2.3×10^{-3} cm^3 of krypton-85 was measured in Capsule BMI-32-1, and 1.0×10^{-2} cm^3 of krypton-85 was detected in Capsule BMI-32-3. Assuming that the gas was released only from the failed specimens, the above amounts correspond to about 3.9 per cent of the total krypton-85 produced in Specimen 2-7V (Capsule BMI-32-1) and about 9.6 per cent of the total krypton-85 produced in Specimens 1-8V and 2-10V (Capsule BMI-32-3). The gas sample from Capsule BMI-32-3 also exhibited considerable xenon-133 activity. Xenon-133 activity may have been present in the other capsules, but the presence of Bremsstrahlung from a beta emitter interfered with its detection. It was suspected that the long-lived beta emitter, argon-39, produced by an (n,p) reaction in potassium-39, was responsible for the Bremsstrahlung.

Postirradiation Heating Experiment

Since the capsules that were irradiated in the MTR did not contain heaters or thermocouples, there was no definite way of determining the temperatures at which the specimens were operating. In an attempt to gain further insight on their in-pile operation, four specimens were given a postirradiation anneal of 1 week at 700 F, and closely examined for indications of swelling and impending failure. Specimens receiving the heat treatment were 1-2-2 from Capsule BMI-32-1 and Specimens 8-13, 5-5V, and 4-10V from BMI-32-2. The specimens were heated in an evacuated autoclave with allowances for collection of all gases generated during the entire heating period.

Following the 1-week heating experiment, the specimens were visually examined. No change in appearance of any specimens such as the presence of cracks or blisters was observed. Measurements of density and dimensions showed that the specimens had not swelled as a result of the heating experiment. Samples of the gases obtained from the autoclave were analyzed for krypton-85 and xenon-133 activity, the presence of which would indicate a cladding failure. No fission gas was detected in the gas samples; consequently, it was assumed that the cladding integrity of all specimens was maintained during heating.

A metallographic examination was made on specimens receiving the postirradiation heat treatment. Thorough examination of the cladding, the fuel and poison particles and the matrix revealed no microstructural variation from that observed in other irradiated specimens.

Metallographic Examination

Specimens from each of the four capsules were sectioned for a microstructural study of the changes produced by irradiation. Twelve of the total of 28 specimens were examined, including three specimens that sustained gross swelling and rupture. The specimens were mounted in Bakelite, ground on water-lubricated silicon carbide papers, and polished in Syntron vibrators employing Linde A and B abrasive on Microcloth. Some of the specimens were examined in the etched condition, which necessitated their further preparation by swabbing with cotton balls soaked in glyceregia. It was intended that the examination would include specimens employing most of the variables being evaluated.

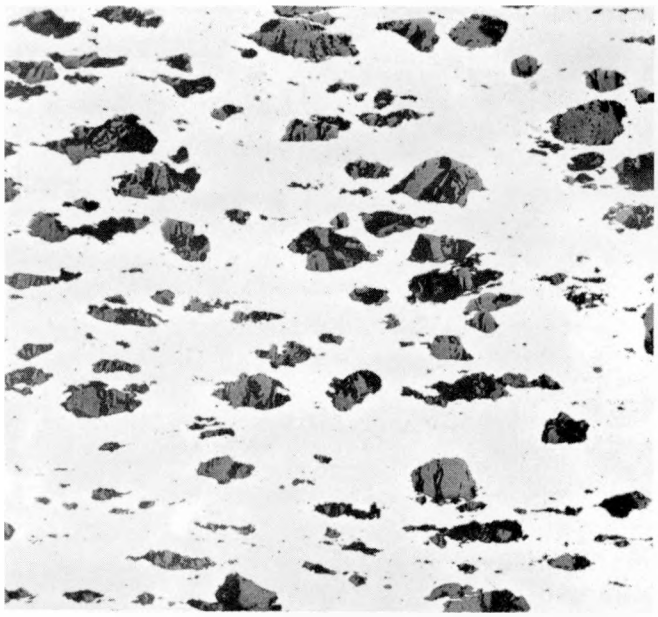
The appearance of unirradiated hydrothermal-process 26 w/o UO_2 in Type 347 stainless steel is shown in Figure 14. The UO_2 particles possess an angular stringered form with a large degree of particle-size variation. The particles of UO_2 contain large and numerous fabrication cracks at right angles to the rolling direction.

The appearance of irradiated hydrothermal-process UO_2 particles dispersed in Type 347 stainless steel is shown in Figure 15. These photomicrographs are of a specimen irradiated to about 45 uranium a/o burnup at a temperature of about 700 F. It was noted that all of the preirradiation fabrication cracks had been healed. Considerable spherical porosity, randomly dispersed in the UO_2 fuel particles, was observed. The previously sharp and irregular UO_2 particle outlines appeared to have become more softened and rounded in contour, with an additional unidentified phase, gray in color, present in many of the particles. This unidentified phase will be referred to throughout the remainder of this report as the gray phase.

Closer observation (see Figure 15a and, also, Figures 17b and 28a) shows that this gray phase found in the metallographic examination occurred chiefly along the outer periphery of UO_2 particles, although it was also observed inside some of the UO_2 particles. The phase has also been observed metallographically in specimens that were irradiated in excess of the 700 F design temperature. In these specimens, to be discussed more fully in later sections, the phase migrated for short distances along grain boundaries of the matrix material. These characteristics indicate the gray phase apparently has a lower melting point and higher fluidity than UO_2; consequently, it is believed that it is an oxide of uranium higher than UO_2, such as U_3O_8 or U_4O_9.

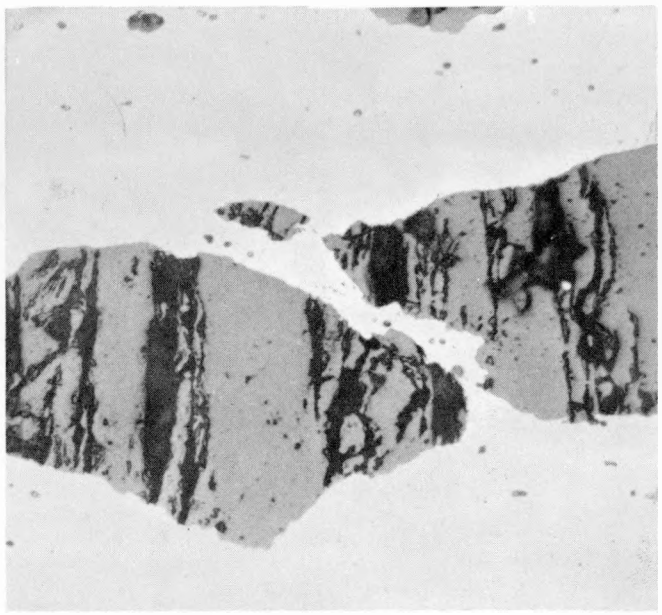
The presence of a fine, white, specular precipitate was also observed in the UO_2 particles. This precipitate, which was particularly evident in the specimens subjected to higher burnup, is believed to contain the oxides of insoluble fission products. Both the gray higher oxide phase and the white specular material were present in UO_2 particles, as shown by Figure 15a. The tendency toward spheroidization of the UO_2 particles may also be observed in this and succeeding photomicrographs.

Upon etching irradiated polished samples in glyceregia, the same general observations were made. In addition, a narrow unetched band (Figure 15b) was observed in the matrix of the Type 347 stainless steel surrounding individual UO_2 particles. The unetched band corresponds to the 6 to 10-mil depth reported for recoil penetration in stainless steel, and is believed to be caused by fission-product recoil from the UO_2 particles.



100X As Polished RM13869

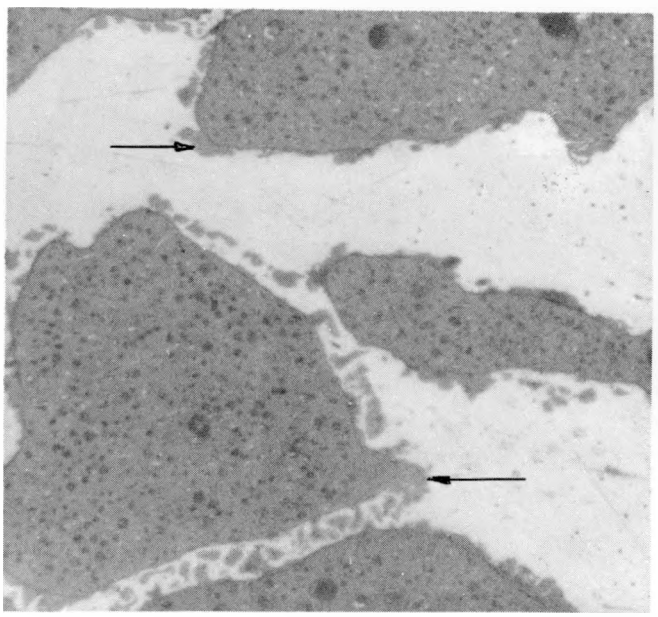
a. Stringered Structure and Large UO_2 Particle-Size Variation



500X As Polished RM12755

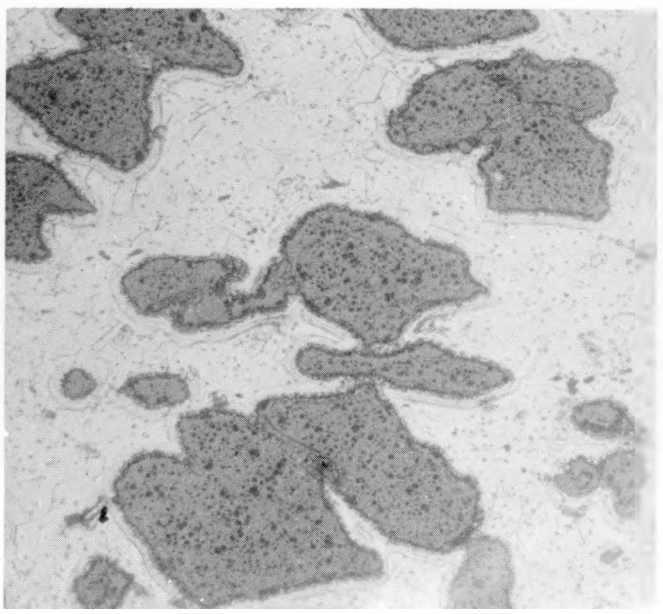
b. Fabrication Cracks in UO_2 Particles

FIGURE 14. APPEARANCE OF TYPICAL UNIRRADIATED HYDROTHERMAL UO_2 (26 w/o UO_2) IN TYPE 347 STAINLESS STEEL MATRIX



500X As Polished HC5167

a. White Precipitates in UO_2 and Gray Phase Surrounding UO_2 Particles



250X Etched HC5129

b. Lightly Etched to Show Recoil-Affected Zone Surrounding UO_2 Particles

FIGURE 15. APPEARANCE OF TYPICAL IRRADIATED HYDROTHERMAL UO_2 IN TYPE 347 STAINLESS STEEL MATRIX

This was Specimen 5-3 irradiated in Capsule BMI-32-1 to a burnup of about 45 a/o uranium. Density change was -2.5 per cent.

The typical appearance of 26 w/o spherical UO_2 particles dispersed in a Type 347 stainless steel matrix is shown in Figure 16. A visual comparison with unirradiated specimen containing hydrothermal UO_2 particles shows that the structure produced with the spherical particles possesses a more uniform appearance and exhibits less stringering of the UO_2 particles.

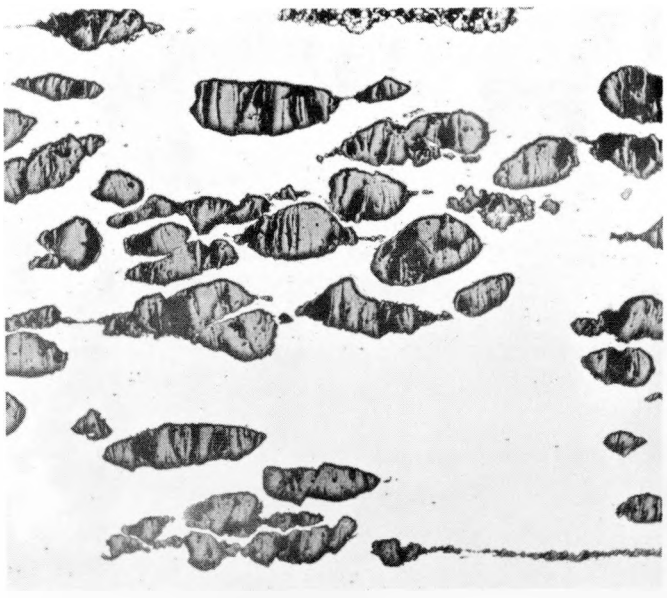
The condition of spherical UO_2 fueled specimen irradiated to a burnup of about 44 uranium a/o is shown in Figure 17. The same phenomena were observed in these specimens as were observed in irradiated specimens prepared with hydrothermal UO_2: the tendency toward spheroidization of the UO_2 particles, the healing of the fabrication cracks in the UO_2, the occurrence of gas-bubble porosity in UO_2 particles, and the presence of both the white specular material and the higher oxide of uranium. Etched specimens showed the narrow recoil-affected zone in the matrix surrounding the UO_2 particles. The recoil zone enclosed both the UO_2 and the higher uranium oxide phases rather than the UO_2 phase alone.

The condition of the cladding, cladding-core interface, and matrix of both types of UO_2-fueled specimens showed no adverse effects as a result of irradiation. No cracks, splits or unbonded areas were observed metallographically in either type of specimen that was irradiated at or near the reference 700 F temperature.

Specimens irradiated in the four capsules contained burnable poisons in the form of B_4C, ZrB_2, NbB_2, and boron-10. The first three poison compounds were dispersed in the matrix of the fuel coupons, and the boron-10 was alloyed into the side frames of the fuel specimens. The poisons were added to specimens containing both hydrothermal and spherical UO_2.

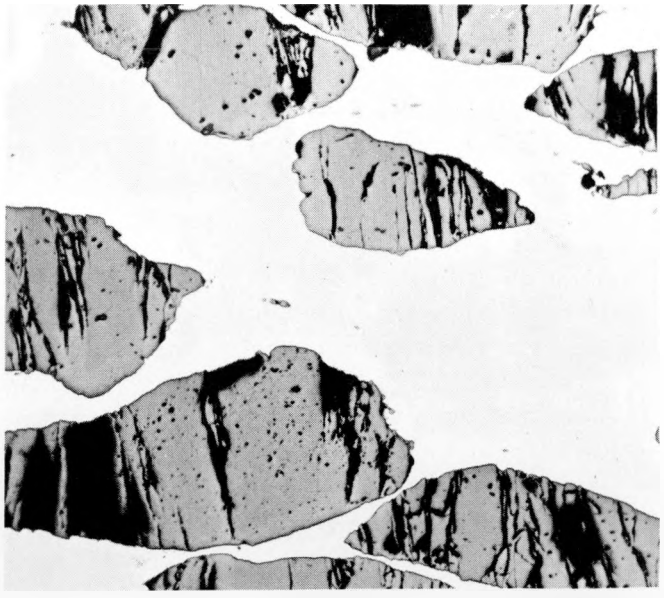
A microstructure of a selected area of an unirradiated control specimen of reference SM-2 fuel (26.0 w/o hydrate UO_2 plus 0.240 w/o boron as ZrB_2 in a Type 347 stainless steel matrix) is shown in Figure 18a. The ZrB_2 particles appear in Figure 18a as sparsely scattered, light-colored particles in which the outer surfaces are ringed by darker colored oxide films. The effects of approximately 40 uranium a/o burnup at temperatures near 700 F are shown in Figures 18b and 18c. In these photomicrographs the reaction zone around the ZrB_2 particles is no greater than found in the preirradiation metallographic examination. This reaction, therefore, took place during fabrication and has no irradiation significance. There is no evidence of porosity in the ZrB_2 particles or surrounding matrix of stainless steel, nor do the particles appear to be fragmented or embrittled. In many cases, fabrication cracks in the ZrB_2 particles appear to have healed in a manner similar to the healing of irradiated UO_2 particles. The condition of the UO_2 particles themselves was similar to that described for irradiated specimens containing no poison. At comparable burnups, over-all specimen swelling did not appear to be greater for specimens containing ZrB_2 than for coupons containing no poison.

An alternate poison considered for use in SM-2 fuel elements is NbB_2. The microstructure of unirradiated control and irradiated specimens containing 26 w/o hydrothermal UO_2 plus 0.207 w/o boron as NbB_2 in Type 347 stainless steel matrix is shown in Figure 19. The NbB_2 is identified as the large whitish particles sparsely and randomly dispersed in the matrix. The NbB_2 particles after a specimen irradiation to about 21 uranium a/o showed no adverse effects of irradiation. The particles did not become porous nor fragment due to the irradiation exposure, and neither evidence of a reaction with the stainless steel matrix nor porosity at the NbB_2-matrix interface was



100X As Polished RM14935

a. Particle Distribution in Matrix

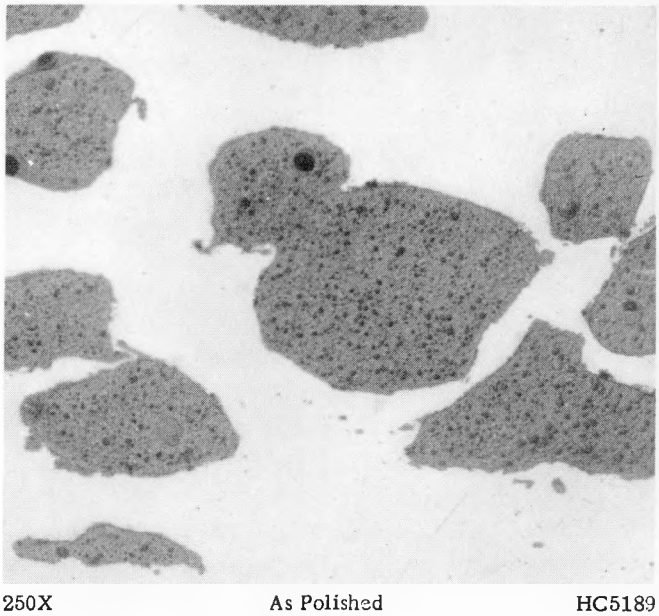


250X As Polished RM14939

b. Detail of UO_2 Particle

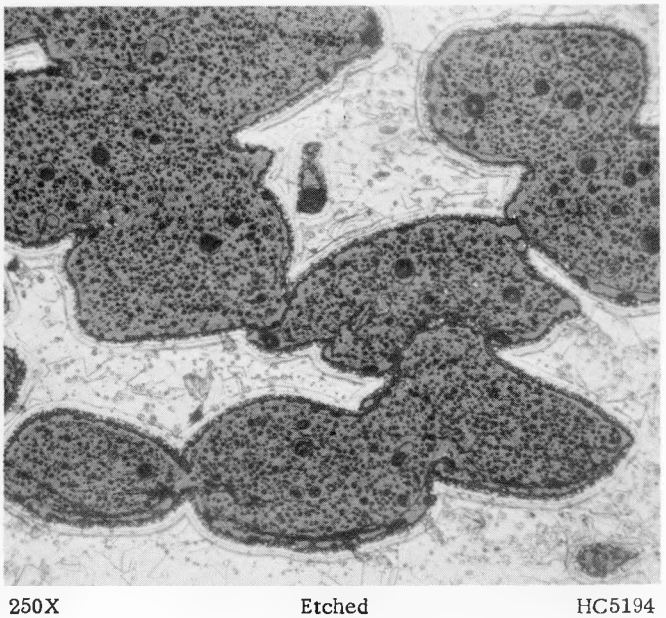
FIGURE 16. APPEARANCE OF TYPICAL UNIRRADIATED SPHERICAL UO_2 IN TYPE 347 STAINLESS STEEL MATRIX

Note large number of fabrication cracks.



250X As Polished HC5189

a. Spheroidization of UO_2 Particle and Absence of Cracks

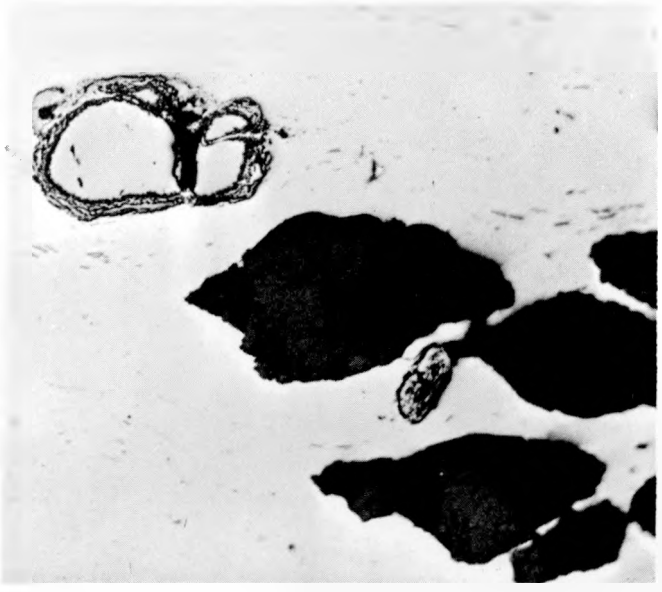


250X Etched HC5194

b. Recoil-Affected Zone Surrounding UO_2 Particles

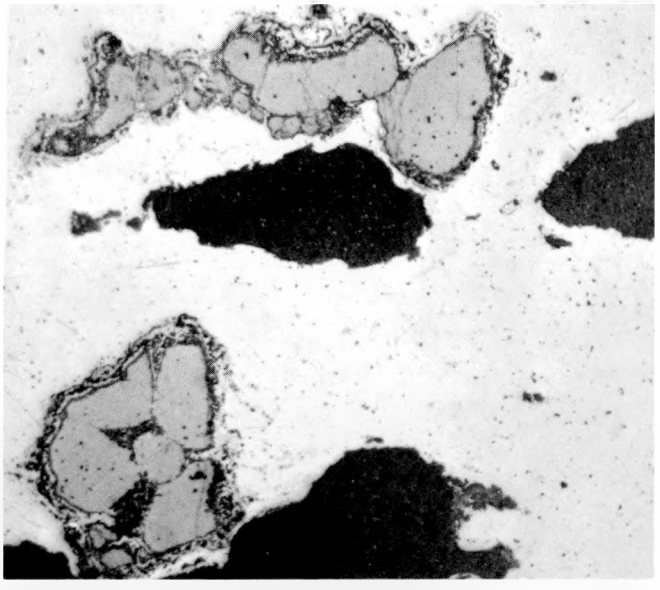
FIGURE 17. APPEARANCE OF TYPICAL IRRADIATED SPHERICAL UO_2 IN TYPE 347 STAINLESS STEEL MATRIX

This was Specimen 5-4 irradiated in Capsule BMI-32-1 to a burnup of 44 a/o uranium. Specimen density change was -2.5 per cent. Note that all fabrication cracks are healed.



250X As Polished RM12881

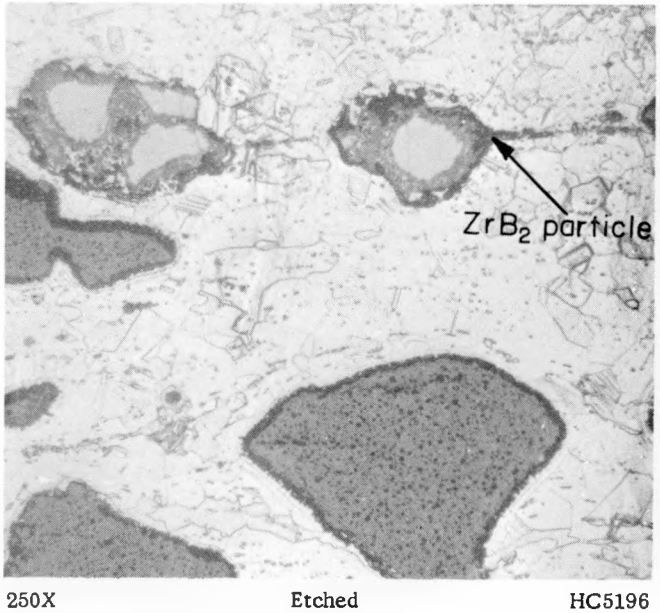
a. Unirradiated Control Specimen



250X As Polished HC5164

b. Irradiated Specimen

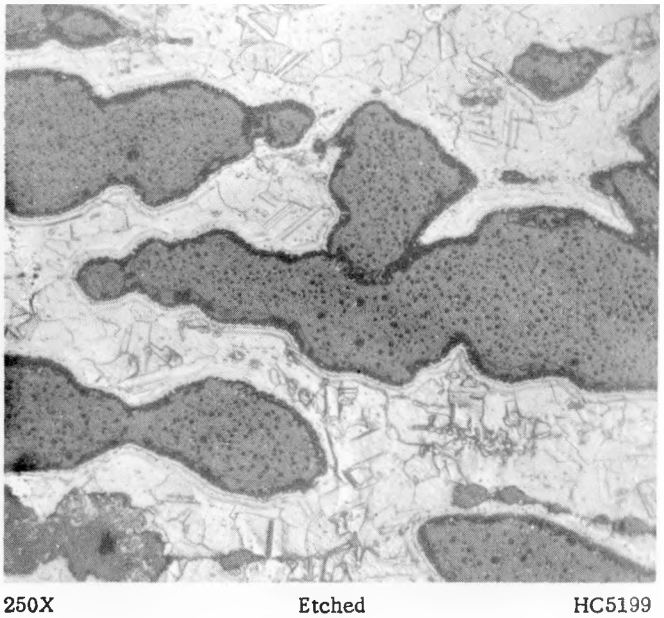
Note excellent condition of ZrB_2 particles, healed UO_2 particles, gas bubbles, and white precipitate in UO_2.



250X Etched HC5196

c. Irradiated Specimen

Cladding-core interface is at side of photomicrograph.



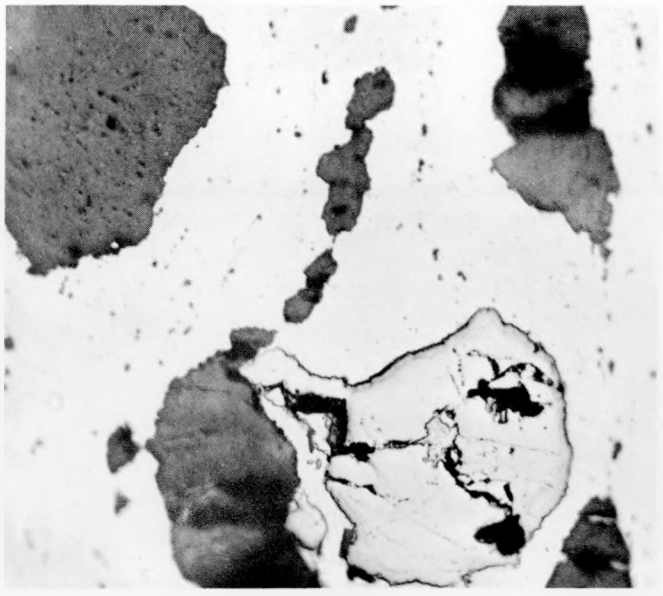
250X Etched HC5199

d. Irradiated Specimen

Heavy etching was used to show recoil-affected zone surrounding UO_2 particles.

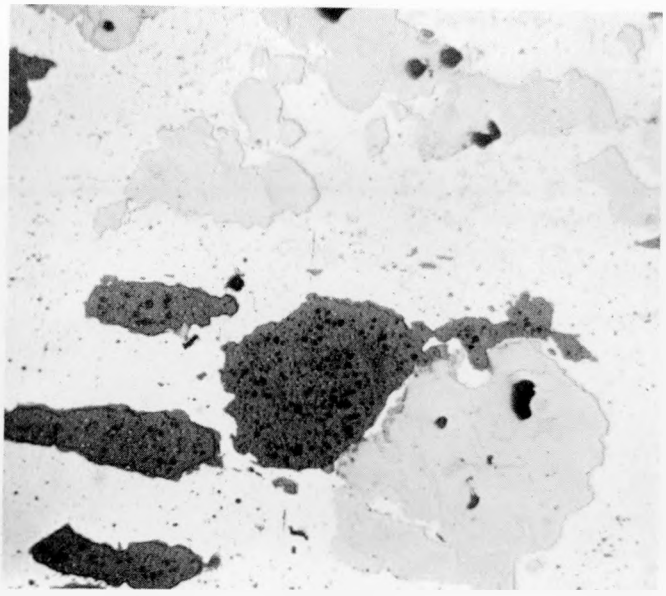
FIGURE 18. UNIRRADIATED CONTROL AND FUELED PORTION OF IRRADIATED SPECIMEN 5-8V

This specimen was composed of 26.0 w/o spherical UO_2 and 0.240 w/o boron as ZrB_2 in Type 347 stainless steel matrix. It achieved a burnup of 40 a/o uranium and underwent a density change of -1.4 per cent. The second half of this specimen was composed of 22.6 w/o Eu_2O_3 in Type 347 stainless steel matrix.



250X As Polished RM13195

a. Unirradiated Control

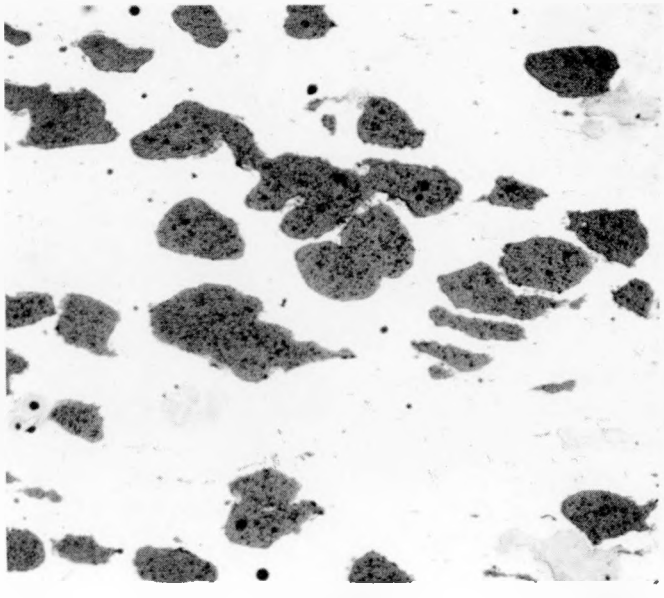


250X As Polished HC5256

b. Condition of the Poison Particles and the UO_2 After Irradiation

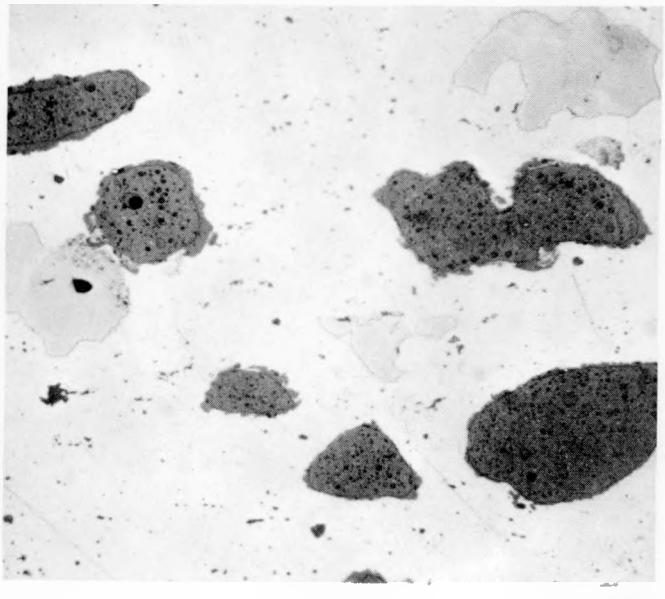
FIGURE 19. CONTROL AND IRRADIATED SPECIMEN 4-11 COMPOSED OF 26.0 w/o UO_2 IN TYPE 347 STAINLESS STEEL MATRIX AND 0.207 w/o BORON AS NbB_2

This specimen was irradiated to about 21 a/o uranium burnup. The density change was -0.9 per cent.



100X As Polished HC5233

a. Over-All View Showing Particle Distribution



250X As Polished HC5237

b. Condition of UO_2 and NbB_2 Particles

FIGURE 20. SPECIMEN 3-13 IRRADIATED TO ABOUT 35 a/o URANIUM BURNUP

This specimen contained 26 w/o spherical UO_2 and 0.204 a/o boron as NbB_2 in Type 347 stainless steel matrix. The density change was -2.0 per cent.

observed. Some of the larger particles contained voids near their centers, but it was noticed that many of the voids were also present in the unirradiated particles. The condition of the UO_2 particles in proximity to the NbB_2 particles was no different than the condition of fuel particles in specimens not containing poison. The same general statements also apply to irradiated specimens fueled with spherical UO_2 and containing NbB_2 as a poison (Figure 20).

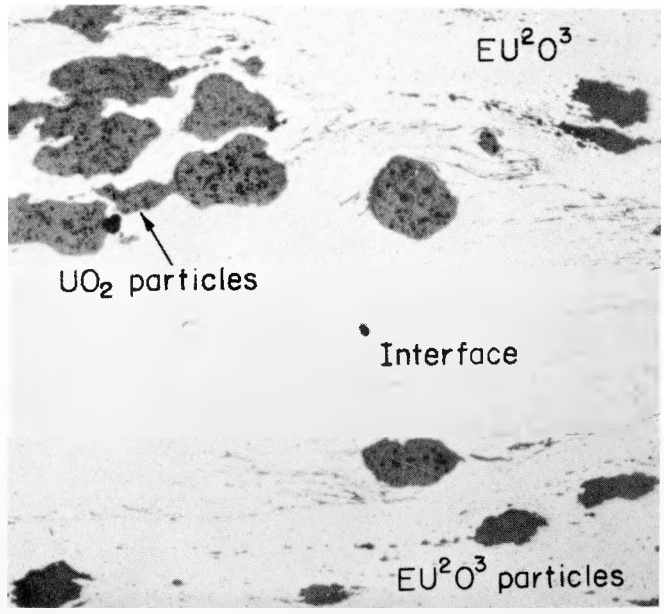
The microstructure of an unirradiated suppressor specimen composed of 22.6 w/o Eu_2O_3 in Type 347 stainless steel is shown in Figure 21a. This material was irradiated as half of a double-length specimen (the other half was reference SM-2 fuel) in Capsules BMI-32-1 and BMI-32-3. Figure 21b shows the irregular nature of the interface between fuel and suppressor sections of Specimen 5-8V after irradiation. The fuel-bearing portion of the specimen is in the upper half of the photomicrograph and shows no unusual effects of irradiation. The irradiated as-polished appearance of the suppressor section is shown in Figure 21c. The particles of Eu_2O_3 appear to possess a finer grain structure and less porosity than in the preirradiated condition. Etching the irradiated specimens with glyceregia caused a vigorous attack on the Eu_2O_3 particles (Figure 21d).

One of the specimens irradiated in Capsule BMI-32-4 was prepared and rolled as a green-pressed fuel compact. This specimen was therefore lower in density than the sintered and coined specimens prepared by the reference fabrication procedure. A representative microstructure of an unirradiated control specimen of reference composition fuel prepared by this technique is shown in Figure 22a. The fuel particles, not being restrained by a high-density matrix, possessed more porosity than observed for samples prepared by routine techniques; otherwise, the microstructure of UO_2 and poison particles was no different from routinely prepared samples. Microstructures of an irradiated unannealed specimen irradiated to about 39.5 uranium a/o burnup are shown in Figures 22b and 22c. The most obvious difference between the microstructure of this and those of other irradiated specimens examined was the greater amount of porosity present in the UO_2 particles which would be anticipated because of the lower preirradiation density. No difference in matrix structure was observed. The density change in this specimen was -2.2 per cent at a burnup of 39.5 uranium a/o, actually less than the change in density for routinely fabricated specimens having comparable burnups.

Two of the specimens that were given a 1-week postirradiation heat treatment at 700 F were sectioned and examined metallographically. Both specimens contained reference SM-2 fuel, differing from one another only in the type of UO_2 contained. Specimen 5-5V (Figure 23) contained hydrate UO_2, and Specimen 4-10V (Figure 24) contained spherical UO_2. The examination revealed no unusual variation in microstructure over that observed in specimens described and pictured previously. Since no apparent microstructural changes were produced by the heating experiment, it is assumed that most of the other specimens were also irradiated at or near this temperature.

100X As Polished RM12879

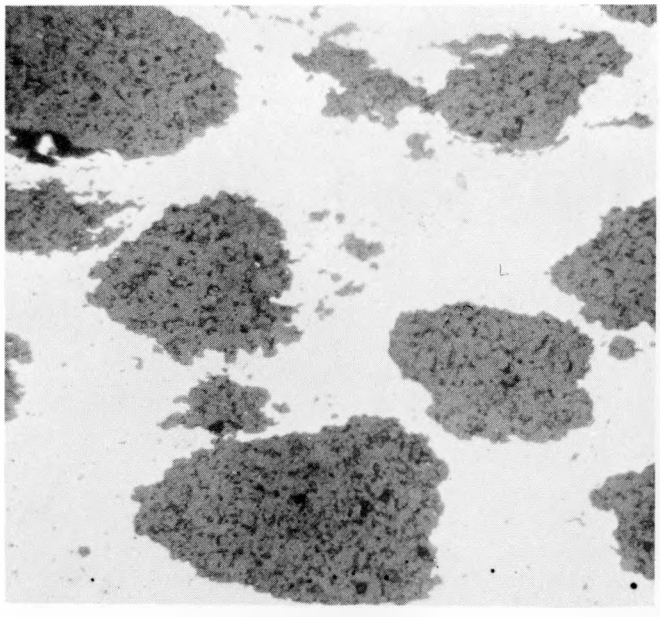
a. Unirradiated Control Specimen



100X As Polished HC5162

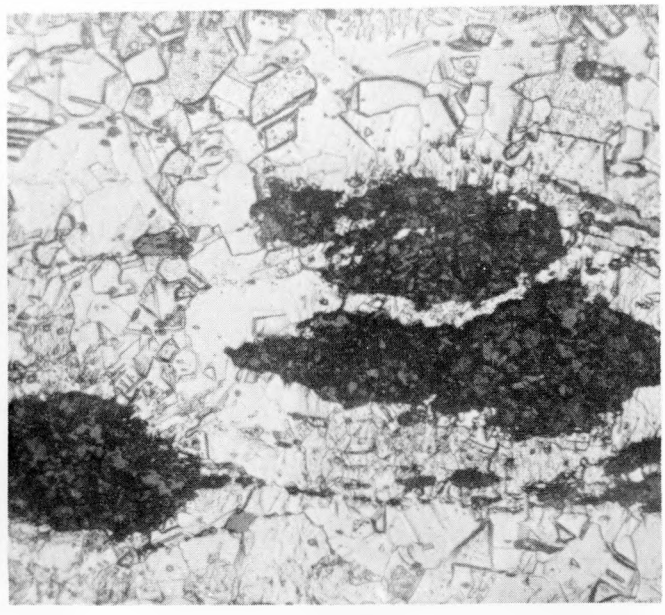
b. Irradiated Specimen

Note the irregular interface produced between the suppressor and fueled sections.



250X As Polished HC5221

c. Irradiated Specimen



250X Etched HC5197

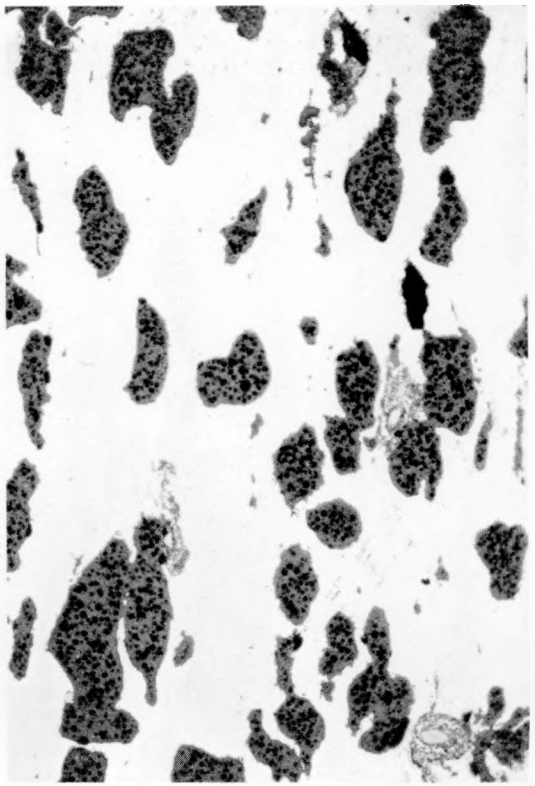
d. Irradiated Specimen

FIGURE 21. SUPPRESSOR SECTION OF 22.6 w/o Eu_2O_3 IN TYPE 347 STAINLESS STEEL MATRIX

The fuel-bearing portion of the irradiated specimen received a burnup of 40.0 a/o uranium in Capsule BMI-32-1.

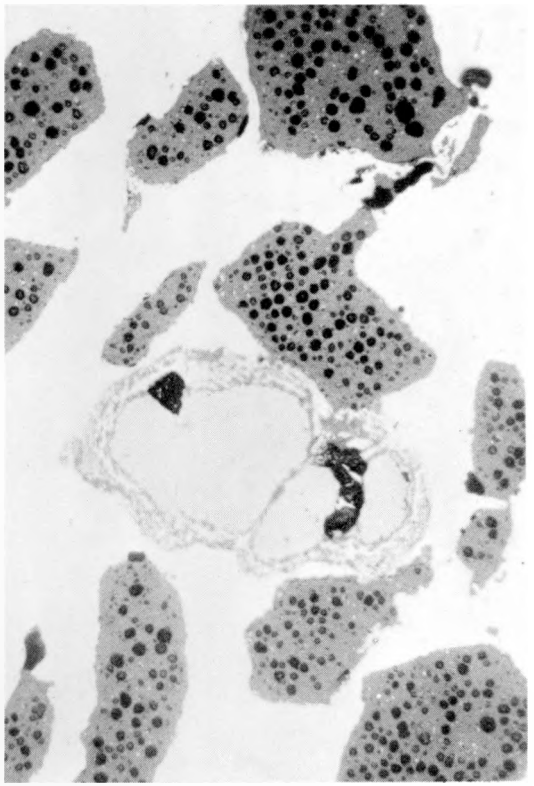
250X As Polished RM12858

a. Unirradiated Control



100X As Polished HC5247

b. Particle Distribution in Irradiated Specimen

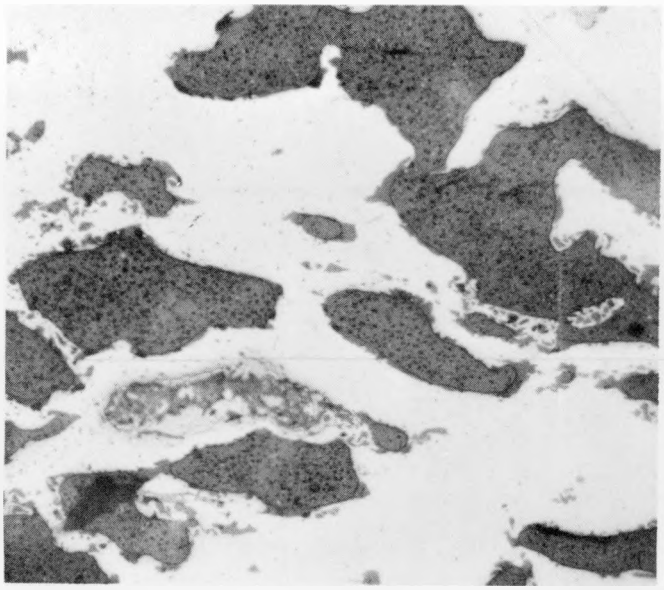


250X As Polished HC5242

c. Voids and White Precipitate in UO_2 Particles in Irradiated Specimen

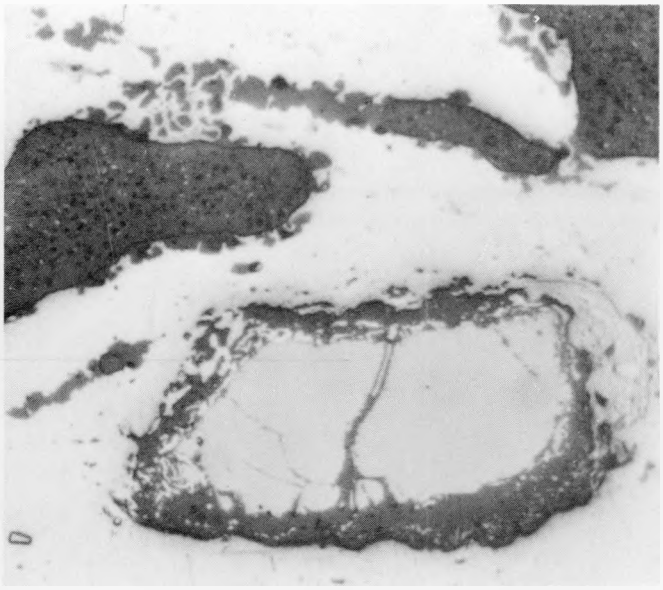
FIGURE 22. EFFECT OF IRRADIATION ON SPECIMEN OF 26.0 w/o SPHERICAL UO_2 PLUS 0.209 w/o BORON AS ZrB_2 IN PREALLOYED TYPE 347 STAINLESS STEEL POWDER

The specimen shown here, 1-12V, was rolled as green pressed. Burnup was 39.5 a/o of the uranium at about 700 F. The density decrease after irradiation was 2.2 per cent.



250X As Polished HC5371

a. Particle Distribution

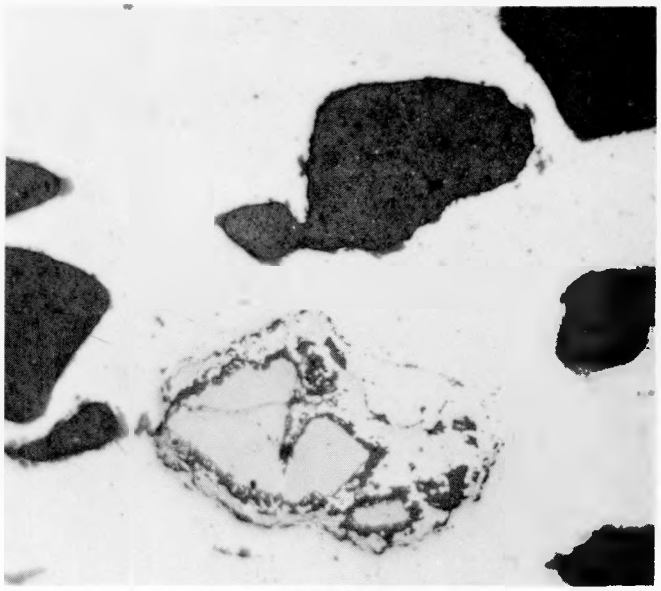


500X As Polished HC5374

b. Detail of UO_2 and ZrB_2 Particles

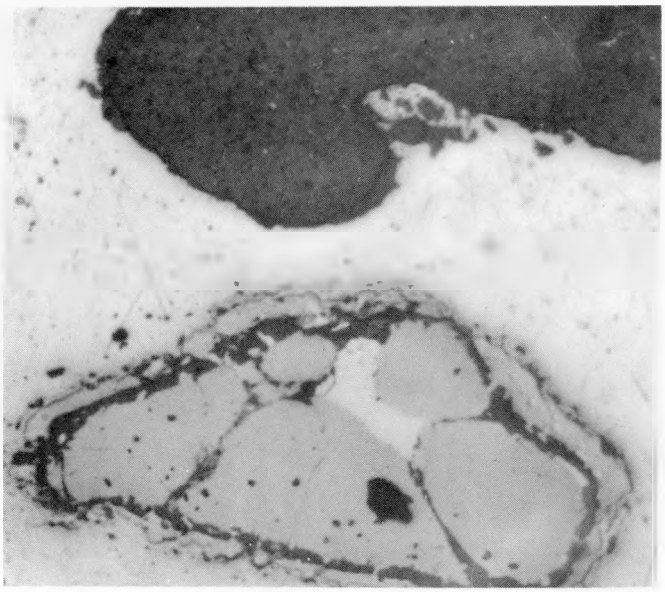
FIGURE 23. POSTIRRADIATION STRUCTURES OF SPECIMEN 5-5V, CONTAINING 26.0 w/o HYDROTHERMAL UO_2 AND 0.199 w/o BORON AS ZrB_2 IN TYPE 347 STAINLESS STEEL MATRIX

This specimen was irradiated to 44 a/o uranium burnup and given a postirradiation heat treatment of 1 week at 700 F. Postirradiation density change was -2.9 per cent. No measurable density change was observed after the 1-week heating experiment.



250X As Polished HC5375

a. Particle Distribution



500X As Polished HC5378

b. Detail UO_2 and ZrB_2 Particles

FIGURE 24. POSTIRRADIATION STRUCTURES OF SPECIMEN 4-10V CONTAINING 26.0 w/o SPHERICAL UO_2 AND 0.192 w/o BORON AS ZrB_2 IN TYPE 347 STAINLESS STEEL MATRIX

This specimen was irradiated to 36 a/o uranium burnup and given a 1-week heat treatment at 700 F. Postirradiation density change was -2.2 per cent. No measurable change in density as a result of heating was found. Note gray phase occurring in stainless matrix around UO_2 particles.

Examination of Failed Specimens

The failed specimens were carefully examined to determine the cause of failure. It was evident from the examination that the specimens were heated to excessive temperature, in some areas above the melting point of the stainless steel. Since all of the failed specimens were situated at end positions within the capsules, it is theorized that at some point during the irradiation the specimens were bare of NaK used as the heat-transfer medium.

Although these specimens failed by inadvertent overheating, well beyond the specified test temperature, an extensive examination was performed to obtain insight into the mechanism of failure in fuels of this type.

The three failed specimens were 2-7V from Capsule BMI-32-1 and Specimens 1-8V and 2-10V from Capsule BMI-32-3. The failures were of the type that could have been produced if the specimens were not completely submerged in NaK. The two failed specimens in Capsule BMI-32-3 were positioned at the bottom of the capsule and, conceivably, could have been exposed if the capsule were inadvertently irradiated in an inverted position.

Specimen 2-7V contained two small blisters near the upper half of the fuel-bearing area on one side and a slightly larger blister in the same area on the opposite side. The appearance of the blisters present on the surface of Specimen 2-7V is shown by photomacrographs in Figure 25.

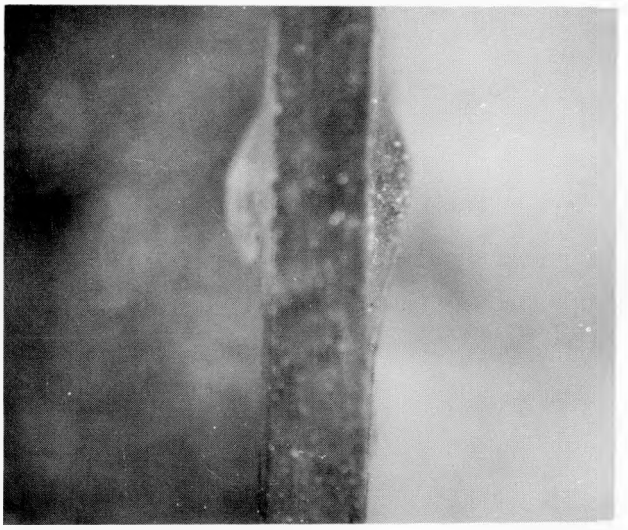
The two failed specimens in Capsule BMI-32-3 (Specimens 1-8V and 2-10V) showed considerably more damage and evidence of overheating. Specimen 1-8V, which was a double-length specimen containing a suppressor section of 22.6 w/o europium oxide (Eu_2O_3) in Type 347 stainless steel and a reference SM-2 fuel specimen, contained a badly fused or melted zone at the junction of the fuel and suppressor. Blistered and melted areas were observed elsewhere in the fueled half of the specimen, although no damage was observed in the Eu_2O_3 portion of the specimen. The specimen, which is shown in Figure 26, broke in two during handling shortly after the photomacrograph was taken.

Specimen 2-10V, which was positioned directly above 1-8V, was also very badly damaged during in-pile exposure. As shown in Figure 27, the specimen contains many large blisters and cracks in the fueled portion along with an area that appears to have been molten at some time during irradiation.

Some of the microstructures observed during examination of the overheated specimens are shown in Figures 28 and 29. Figure 28 shows the microstructure of two selected areas in Specimen 2-10V. This specimen received a burnup of 35 uranium a/o at a temperature sufficiently high to produce localized melting. Figure 28a shows how the gray uranium-bearing phase has moved by grain-boundary migration from the periphery of the UO_2 particle to coalesce at the intersection of grain boundaries in the stainless steel matrix. Because of its obviously greater fluidity, this phase is thought to be a higher uranium oxide such as U_3O_8 or U_4O_9. It is also possible that the phase may be a complex metal oxide of cerium, chromium, lanthanum, or yttrium of the M_2O_3 type. Figure 28b shows numerous large holes in the UO_2 particles and associated phases. The large gas bubbles were produced after the matrix had ruptured and, consequently, offered less restraint to the growth of the UO_2 particles.

4X HC4883

a. Front View of Two Small Blisters

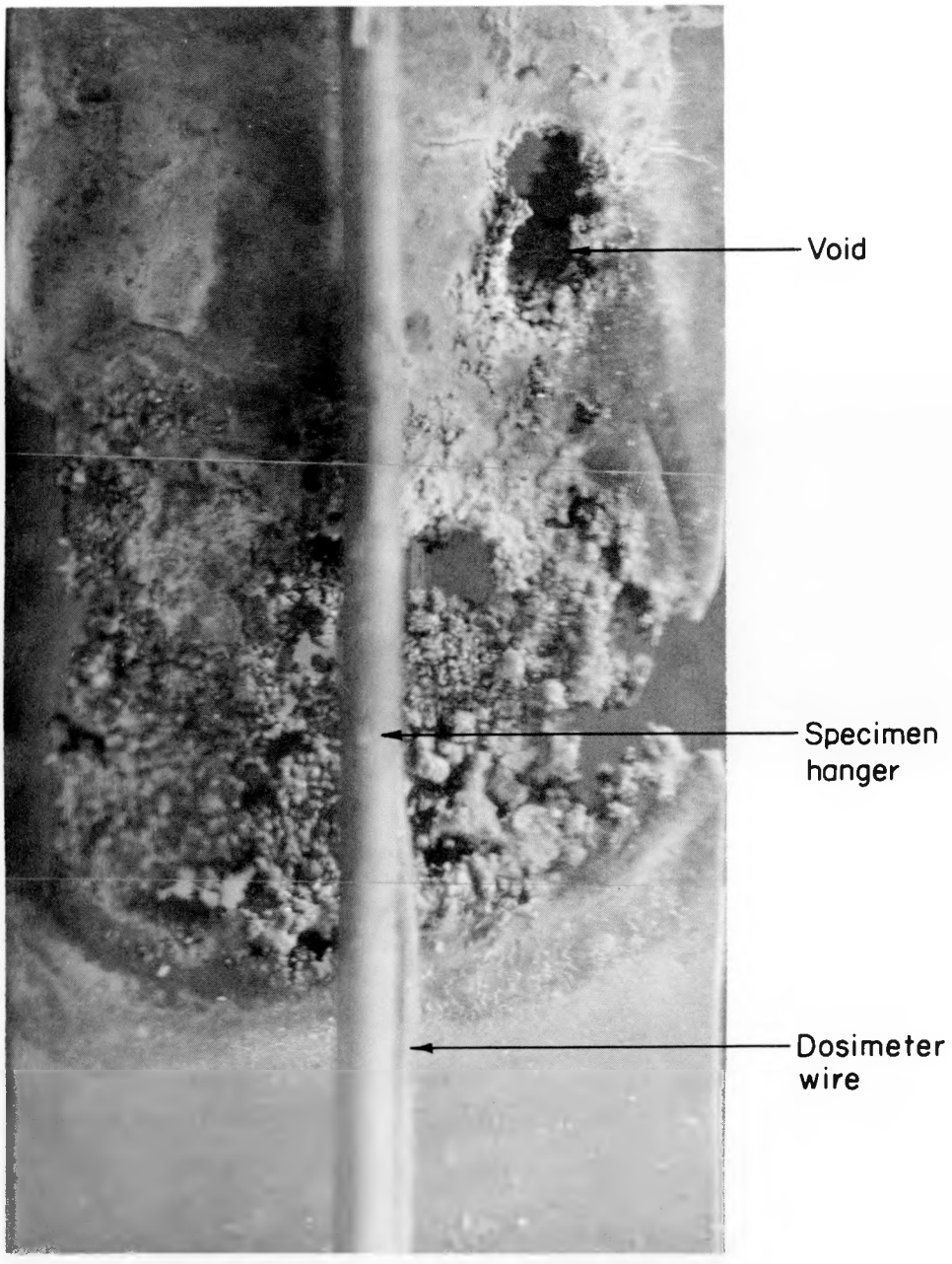


12X HC4885

b. Side View Showing Blisters Near Top of Specimen

FIGURE 25. FRONT AND SIDE VIEWS OF SPECIMEN 2-7V IRRADIATED TO ABOUT 39.0 a/o URANIUM BURNUP

Specimen contains 26.0 w/o spherical UO_2 and 0.095 w/o boron as ZrB_2 in a matrix of Type 347 stainless steel.



6X HC4829, HC4828, and HC4827

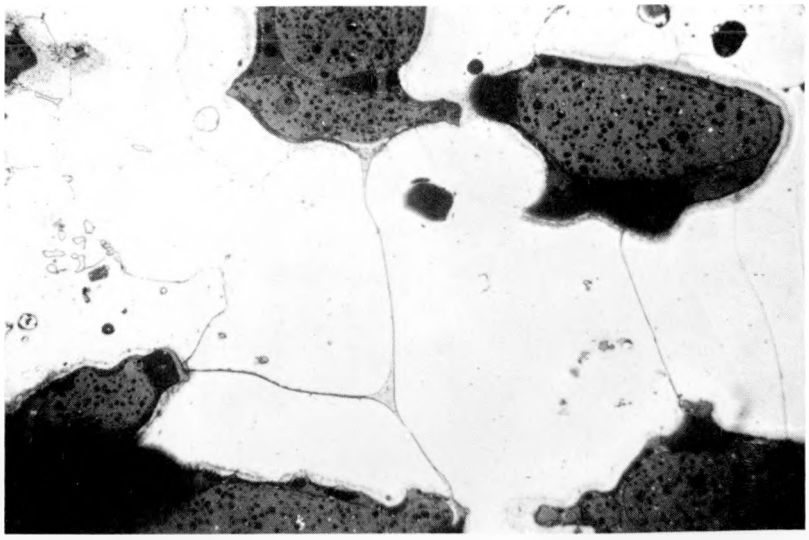
FIGURE 26. SPECIMEN 1-8V FROM CAPSULE BMI-32-3

This was a double-size specimen. One-half its length was a suppressor section containing 22.6 w/o Eu_2O_3 in Type 347 stainless steel. The other half was reference SM-2 fuel. The junction of fuel and suppressor section is shown here.

6X HC4831, HC4832, HC4833, and HC4834

FIGURE 27. SPECIMEN 2-10V FROM CAPSULE BMI-32-3

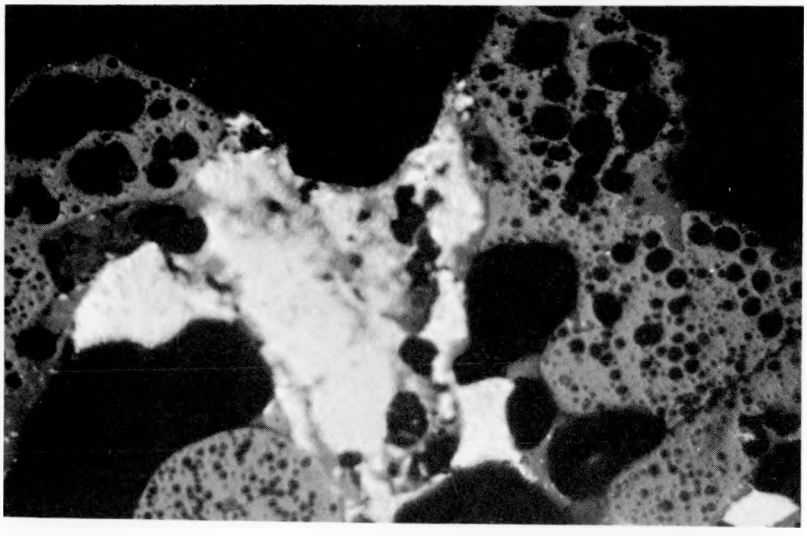
This specimen and its adjacent specimen (1-8V) were badly blistered and showed evidence of high-temperature operation.



250X Etched HC5292

a. Area Revealing Extreme Fluidity of Uranium-Bearing Phase

This phase (gray) migrated along the Type 347 stainless steel grain boundaries to coalesce in the matrix, as can be seen at bottom center.



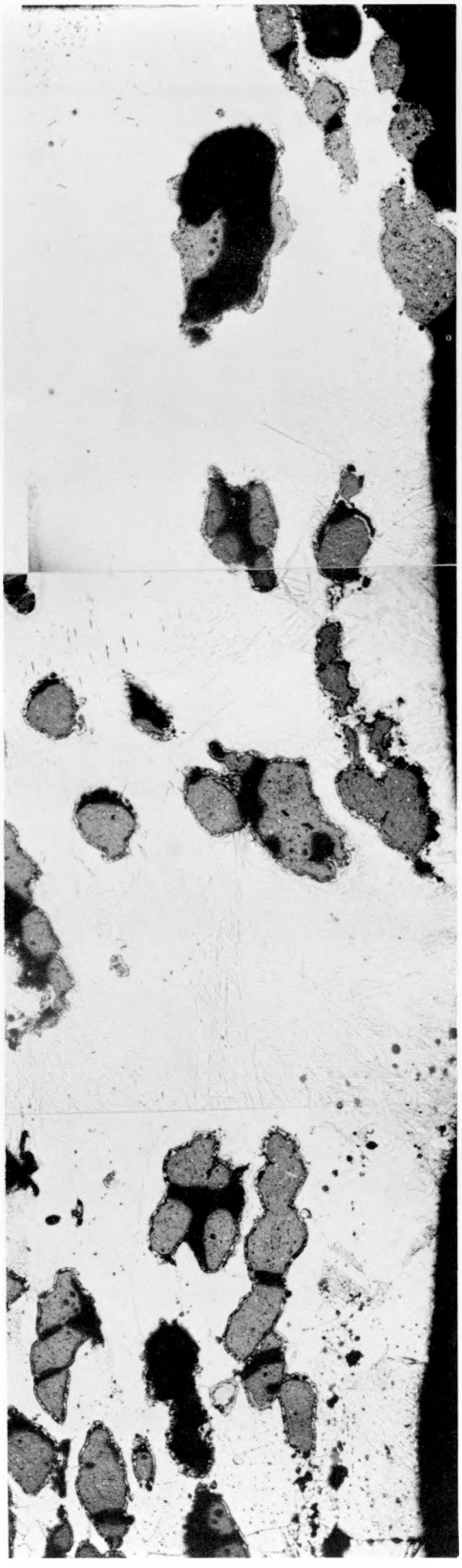
250X As Polished HC5310

b. Large Fission-Gas Bubbles in UO_2 Particles

Note that the bubbles are largest in the UO_2 particles that are not confined in the matrix.

FIGURE 28. SELECTED AREAS FROM SPECIMEN 2-10V

This specimen was irradiated to a uranium burnup of 35 a/o at a temperature sufficiently high to cause localized melting.



75X HC5276, HC5277, and HC5281

FIGURE 29. SEQUENCE OF POSTIRRADIATION PHOTOMICROGRAPHS SHOWING THE MOVEMENT OF PARTICLES OF UO_2 THROUGH MOLTEN STAINLESS STEEL MATRIX AND CLADDING

At the bottom of the photomicrographs, the UO_2 particles are contained within the matrix. In the center of the traverse, the matrix and cladding became molten and particles of UO_2 have migrated through the matrix nearly to the edge of the cladding. Near the top of the traverse, the UO_2 particles have completed the migration to the edge of the cladding. This sequence indicates that the stainless steel was molten in localized areas at some time during irradiation, allowing the UO_2 particles to move freely through the stainless steel. Specimen 1-8V is shown.

Figure 29 is a photomicrograph showing the presence of UO_2 fuel particles on the surface of Specimen 1-8V. Near the bottom of the photomicrograph, the UO_2 particles are contained within the matrix. Near the center of the depicted area, there is evidence that the Type 347 stainless steel matrix became molten, allowing the UO_2 particles to reposition themselves. At the top of the photomicrograph, particles of UO_2 have emerged from the matrix and cladding and are embedded in stainless steel at the surface of the specimen. The gray phase generally associated with UO_2 particles is absent in the displaced UO_2 particles, although the recoil-affected zone which occurs in conjunction with UO_2 particles is present. The size of the white particles in the UO_2 suggests they have coalesced to form fewer but larger masses.

DISCUSSION AND CONCLUSIONS

In general, irradiation of the initial four capsules of alternate and reference SM-1 and SM-2 fuels was successful. The burnup range of 20 to 45 uranium a/o achieved, however, was less than the 35 to 70 uranium a/o initially anticipated. Twenty-five of the total of 28 specimens experienced maximum density changes of -3.1 per cent or less. The general condition of the specimens indicated that the fuel material should be capable of withstanding higher burnups without serious damage. During the in-pile exposure, the specimens were subjected to heat fluxes on the order of 10^6 Btu/(hr)(in.), which is considerably greater than fluxes to which the actual SM-2 fuel element will be subjected. Fission gas was retained within the cladding of all specimens except those exhibiting failure due to localized overheating. Samples of gas obtained on selected specimens during postirradiation annealing at 700 F contained no xenon or krypton activity, indicating that cladding integrity was maintained at this operating temperature.

Fission gas was collected from both capsules that held ruptured specimens. The greatest amount of fission gas was obtained from Capsule BMI-32-3, in which two specimens, 1-8V and 2-10V, failed.

The causes for failure of the three specimens are not readily apparent. Obviously, the specimens were heated considerably in excess of the intended 700 F specimen-surface temperatures. Since all specimens contained approximately the same UO_2 loading, and all were positioned in approximately the same thermal-neutron flux, it must be assumed that the transfer of fission heat was impaired in the subject cases. This could have occurred if the specimens were not completely immersed in NaK coolant. It is possible that Capsule BMI-32-3, which contained as its two bottom specimens, 2-10V and 1-8V, was inadvertently loaded and irradiated upside down. This may have occurred when the capsule was replaced in the reactor after examination for leakage.

The metallographic examination of the sound specimens revealed the following:

(1) All UO_2 particles showed evidence of sintering. All preirradiation fabrication cracks were healed, and much evidence was observed to indicate the UO_2 particles tend to become spheroidal. Spherical porosity was observed in the UO_2 particles, the amount and size apparently determined by the amount of burnup and size of UO_2 particle and temperature.

(2) A considerable amount of a uranium-bearing phase which is gray in color was observed within some of the UO_2 particles and around the periphery of most of the particles. This gray phase apparently possesses a lower melting point and greater fluidity than UO_2. Although no attempts were made to identify this phase, it is believed that it may be a higher uranium oxide such as U_3O_8 or U_4O_9.

(3) A white specular precipitate is present in the UO_2 particles. The precipitate is believed to be an insoluble fission-product oxide. Both the white precipitate and the gray uranium-bearing phase have been previously observed and reported. Neither of the phases appears to contribute adversely to the irradiation behavior of the specimens.

(4) The ZrB_2 and NbB_2 burnable poisons appear largely unaffected by irradiation. In many cases, fabrication cracks in the NbB_2 particles appeared to have been healed during irradiation. There was no evidence of porosity, cracking, or swelling in the matrix surrounding the particles.

(5) In the etched condition, the stainless steel matrix immediately surrounding the UO_2 particles remained unetched. The width of the unetched zones corresponded to the 6 to 10-μ recoil range reported for stainless steel.

(6) No microcracks were observed in the matrix of the unfailed specimens, and the cladding was intact in all specimens examined.

Aside from the three specimens that were obviously overheated, the performance of all specimens in the four capsules was satisfactory for the burnup obtained. On the basis of the results of the various postirradiation examinations, the following general observations were made:

(1) There is no apparent difference in the irradiation behavior of specimens made up with either hydrate or commercial spherical UO_2.

(2) The presence of the 22.6 w/o Eu_2O_3-Type 347 stainless steel suppressor does not noticeably affect the behavior of the adjacent fuel core.

(3) A specimen prepared by direct rolling of a green fuel compact, of lower density, performed satisfactorily. However, there was considerably more porosity in the UO_2 particles of this specimen than in specimens with comparable burnups prepared by standard procedures.

(4) A specimen containing 0.023 w/o boron-10 incorporated in the stainless steel picture frame performed satisfactorily.

(5) No adverse effects were noted for specimens incorporating relatively large loadings of burnable poisons in the form of ZrB_2, NbB_2, and B_4C. No cracking or porosity was observed in the surrounding stainless steel matrix, and there was considerable evidence to indicate that

the fabrication cracks in the poison particles healed in a manner similar to UO_2 fuel particles during irradiation.

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