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EXPERIMENTS TO DETERMINE THE RADIATION STABILITY OF UN DISPERSIONS IN STAINLESS STEEL

by

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A controlled radiation experiment was performed to determine the stability of fuel containing 28 w/o UN dispersed in and clad with Type 318 stainless steel as compared with fuel containing 30 w/o UO_2 dispersed in the same material. The specimens were prepared by hot rolling the fuel coupons in Type 318 stainless steel using the picture-frame technique for initial bonding and reduction. Final dimensions were obtained by cold rolling.

A special radiation capsule was designed which contained heat control and enough thermocouples to insure a good continuous-temperature history throughout the test. This capsule and the method by which the specimens were loaded are discussed in some detail.

Because of the capsule instrumentation, the known MTR position into which the capsule was placed, and the dosimeters placed in the capsule it was possible to obtain a complete flux and temperature history of the capsule during the irradiation. When it was estimated that the specimen burnup was about 7.2 a/o of uranium-235 the capsule was removed from the reactor and returned to the Battelle Hot-Cell Facility.

The postirradiation examination which consisted of fission-gas analysis, density and dimensional determinations, radiochemical and isotropic burnup analysis, and metallography is discussed completely in the report. The results of the various phases of the experiment are discussed and conclusions are drawn on the basis of an integrated evaluation.

These results indicate that the UN dispersions withstood irradiation at temperatures of 1500 to 2000 F and at burnups of 3.5 to 5.0 a/o of the uranium-235 at least as well as the UO_2 dispersions. These conclusions indicate the potential of UN as a high-temperature fuel, however, it is also obvious that many additional radiation experiments are required.

INTRODUCTION

The dispersion of fertile materials such as UO_2 in high-strength matrices has proved to be a reliable method of producing reactor fuels with good radiation stability at relatively high operating temperatures. One major difficulty with dispersion fuels has been the relatively low uranium density which can be obtained. This problem cannot be solved simply by incorporating more fuel in the matrix because the mechanical properties of the fuel are severely affected at high fuel loadings. This in turn limits the maximum burnup to which the fuel may be used at a given temperature. It therefore follows that materials with high uranium densities would serve best in dispersion-type fuels.

Research conducted at Battelle[1] in 1957 indicated that a dispersion of UN in stainless steel might possess considerable potential as a high-temperature fuel-element material. The density of uranium in UN is about 13.5 g per cm^3 as compared with a uranium density of 9.6 g per cm^3 in UO_2. Thus UN would require about 30 per cent less volume in a dispersion fuel than an equivalent loading of UO_2. This suggested not only the possibility of improving the mechanical properties of the dispersion, but, as indicated by study, reduced the tendency for particle fragmentation during fabrication as compared with UO_2. Because of these favorable properties Aerojet-General Nucleonics (AGN) considered UN as a potential GCRE fuel. The irradiation of the UO_2 and UN dispersion specimens was therefore planned to provide a direct comparison of the radiation stability of the two materials under similar conditions of temperature and burnup. Changing emphasis in the GCRE program resulted in postponement of the examination of the irradiated specimens. At this point, ORNL requested that Battelle open the capsule and examine the four irradiated specimens.

A program for the examination of the specimens from Capsule BMI-28-1 was planned in conjunction with ORNL personnel. The results of the examinations are discussed in this report.

EXPERIMENTAL PROCEDURES AND RESULTS

The specimens and irradiation capsule used in this experiment were fabricated at Battelle. The irradiation was conducted in the MTR. Upon completion of the irradiation, the capsule was returned to Battelle where it was examined in the Hot-Cell Facility. Details of the specimen-fabrication techniques, capsule design, irradiation, and post-irradiation examination are discussed separately below.

Fabrication of Specimens

The UN used in the fabrication of the dispersion specimens was prepared by heating 93.16 per cent enriched uranium metal in pure nitrogen at 850 C to form U_2N_3 and then decomposing the U_2N_3 to form a stable UN compound by heating to 1300 C in a vacuum. Particles of UN (mesh size minus 150 to plus 270) were then blended with the components of the Type 318 stainless steel (mesh size minus 325 to plus 400). Four coupons of 28 w/o UN-stainless steel was then pressed at 50 tsi. No sintering or coining of these coupons was required. The four coupons were machined to proper size and fitted into spaces in a Type 318 stainless steel frame for roll bonding. All coupons were cleaned mechanically while frame components were cleaned and degreased with acetone before assembly. After assembly, the cover plates were welded to the frame under an inert atmosphere. The compact was then evacuated to a pressure of less then 10^{-3} mm of mercury at 600 C and sealed. The compact was heated in a hydrogen atmosphere and rolled at 2100 F with an initial 30 per cent reduction in thickness followed by 20 per cent reductions per pass to within 15 per cent of the desired thickness. The hot-rolled compact was pickled until clean and annealed in a dry-hydrogen-atmosphere furnace at 2300 F for 2 hr. The annealed compact was then cold rolled with thickness reductions of 2 to 3 per cent per pass until a thickness of approximately 0.045 in. was obtained. The compact was flat

(1) References at end of text.

annealed in a jig in a hydrogen furnace for 1 hr at 2050 F. The four fueled sections of the compact were then separated and radiographed to mark the exact location of the fueled cores. Using the radiographs as templates, the exact specimen sizes were marked and the specimens sheared and filed to size, 1.4687 in. long by 0.6875 in. wide. Radiographs of the finished specimens were obtained to check the exact position of the core. Each specimen was tested for flaws in the cladding by immersion for three 1/2-hr periods in boiling 50 per cent HNO_3 solutions. Each solution was analyzed for uranium content. This test indicated that all four specimens were free of cladding flaws Each specimen was then identified by a number vibratooled in the upper left corner and also by notches filed in the edges.

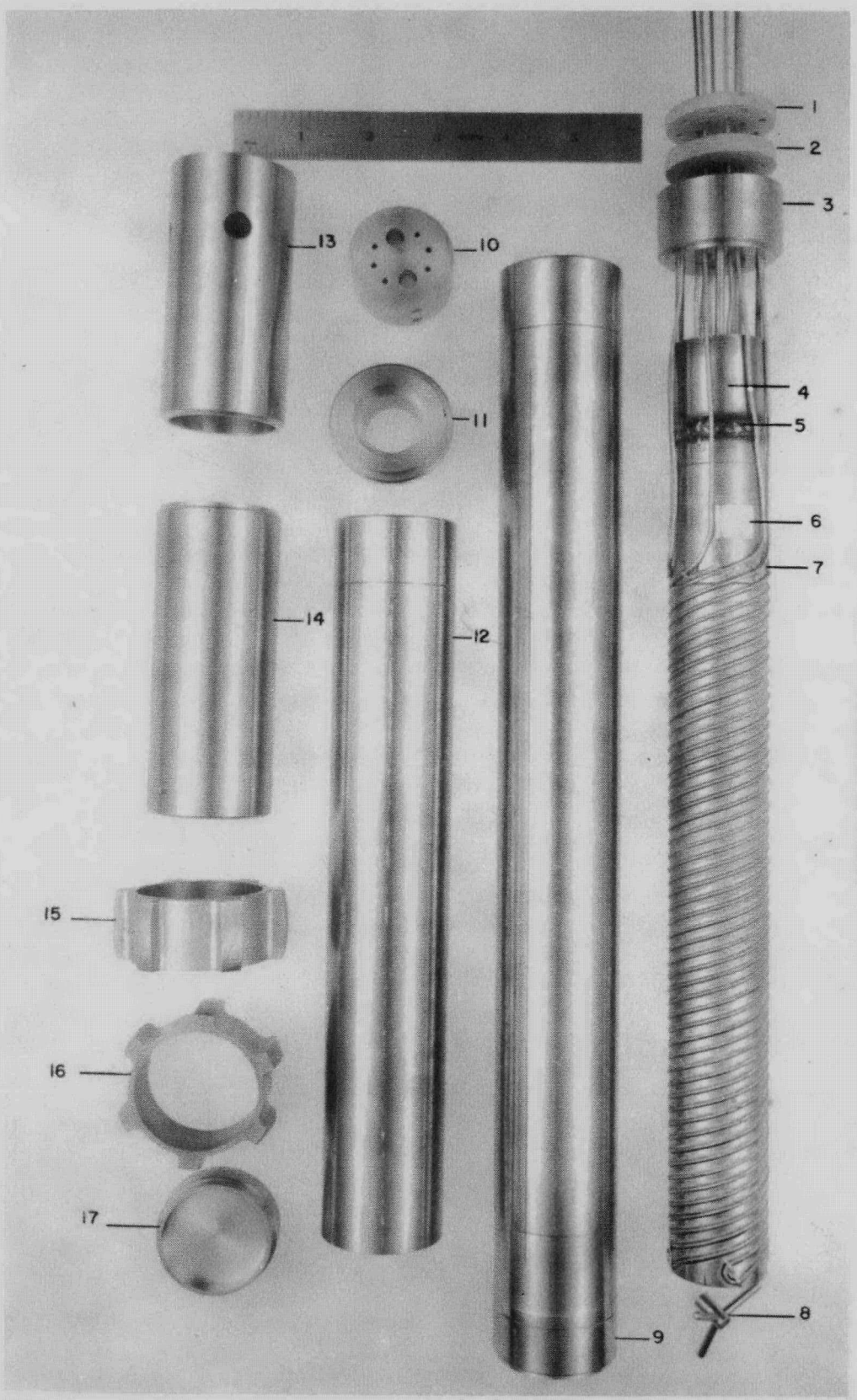
The UO_2 specimens were fabricated in a manner quite similar to that used in preparation of the UN specimens. The UO_2 was obtained from a standard hydride production run of 93.16 per cent enriched uranium. The stainless steel was identical to that used in the UN specimens and was blended with 30 w/o UO_2 of minus 100 plus 200 mesh. Coupons of this composition were pressed at 15 tsi and sintered in a hydrogen atmosphere overnight at 1600 F. The temperature was then raised at a rate of 170 F per hour to 2300 F and held for 2 hr. The coupons were cooled in the furnace cold zone and then coined at 50 tsi. The remainder of the UO_2 specimen fabrication was identical to that of the UN specimens except that the hot rolling was performed at 2200 F with an initial 40 per cent reduction followed by 20 per cent reductions per pass. Control specimens of both UN and UO_2 were sectioned for metallographic examinations. The physical dimensions and density of each specimen were measured prior to irradiation.

Capsule Design and Specimen Encapsulation

In order to obtain good comparison between the radiation stability of UN and UO_2 dispersions, a capsule was designed which would permit both the measurement and control of specimen temperatures. To accomplish these requirements, electric-resistance heaters and six thermocouples were incorporated in the design. The capsule itself was of a double-walled, NaK-filled, high-temperature type, designed to maintain specimen surface temperatures of 1650 F ± 50, in a specified neutron flux (see Figure 1).

The outer capsule shell, or water-contacting wall, was made of stainless steel and was approximately 0.1 in. thick with an outside diameter of 1.750 in. The outer capsule was separated from the inner capsule by an 0.068-in.-wide helium annulus when at operating temperature. The gas annulus was uniform over the capsule length and served as the primary heat-transfer barrier. Four nickel-cobalt dosimeters, placed 90 deg apart, were affixed in grooves along the outside of the outer shell. Two nickel-cobalt dosimeters were extended the length of the specimens in the inner capsule.

The inner capsule was machined from a solid block of Type 304 stainless steel to eliminate the need of a bottom weld. The outside diameter of the inner body was 0.699 in. at the operating temperature and was supported and centered in the outer shell by four short axial rods at the top and a spider at the bottom. This arrangement maintained concentricity without imposing axial restraint on the inner capsule, thus permitting relative motion resulting from differential thermal expansion between the inner and outer shells as a result of heating. Two 1-kw heaters sheathed with 1/16-in. of stainless steel and packed with MgO were brazed into spiral grooves machined into the outside diameter of the inner capsule. To achieve good heat transfer, approximately one-half of the



1. Lavite disk
2. Lavite disk
3. Second thermocouple seal
4. First seal
5. Weld
6. Fission boss
7. Heaters
8. Centering pin
9. Outer shell
10. Saddle clamp
11. Lead tube junction
12. Long extension
13. Top spacer
14. Short extension
15. Centering ring
16. Centering ring
17. Bottom plug

N56687

FIGURE 1. COMPOSITE PARTS OF CAPSULE BMI-28-1

heater diameter was imbedded in the capsule wall. The six stainless steel-sheathed Chromel-Alumel thermocouples penetrated the top of the inner capsule through a braze seal and were positioned at approximate midface locations of each specimen, Figure 2. The top specimen was thermocoupled on both faces. Also, a thermocouple was positioned in the NaK bath midway between the center of the top specimen face and the capsule wall. The thermocouples positioned next to the specimens were expected to record temperatures only 20 to 30 F below the actual temperature of the specimen surfaces.

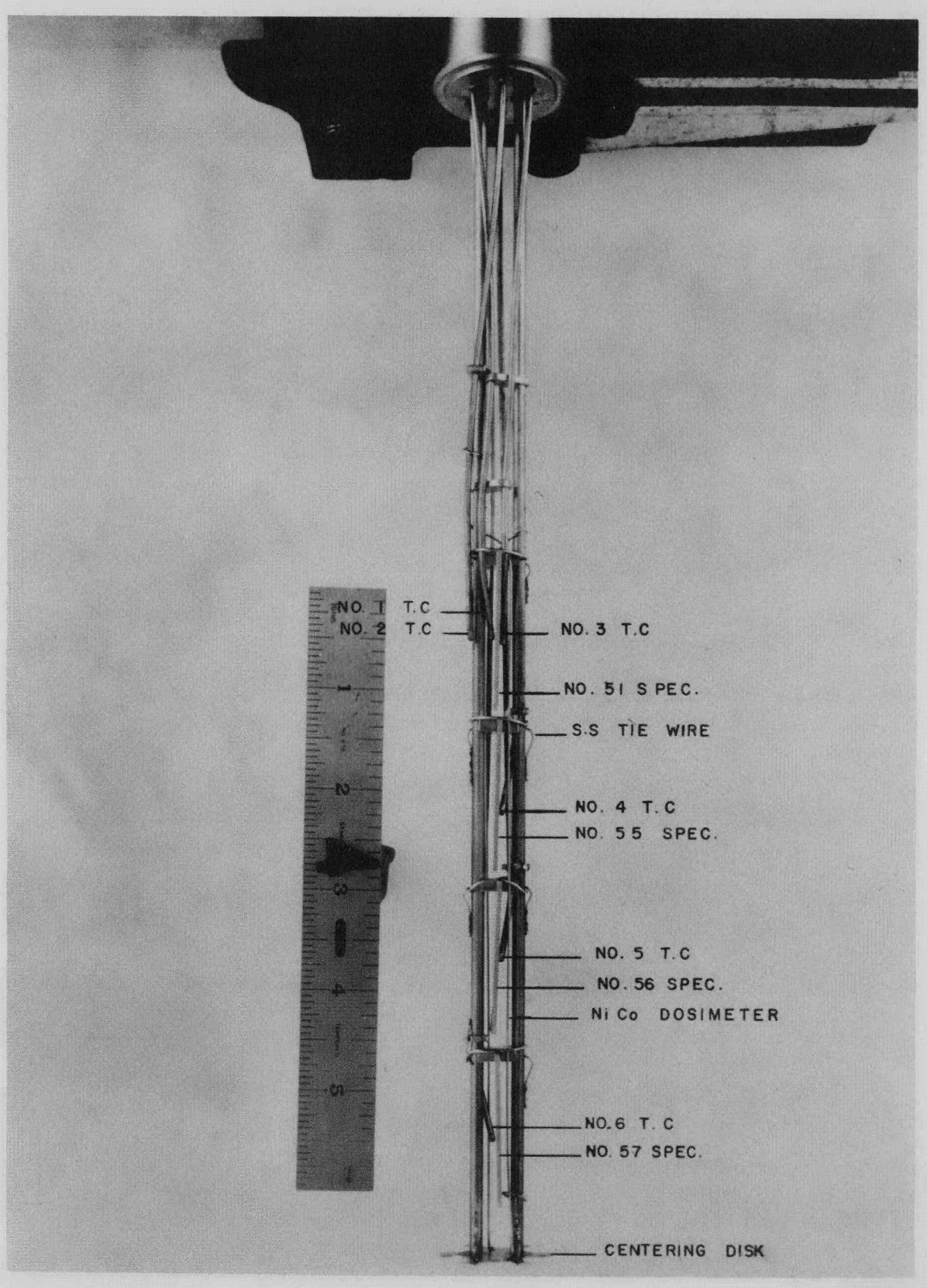
Two regions of major importance were studied in consideration of heat-transfer conditions within the capsule. These regions were the NaK bath between the specimens and the inner capsule and the gas annulus between the inner and outer capsules. A temperature study conducted with the use of an electrical analog computer during capsule design indicated an expected temperature drop of 150 F in the NaK between the specimen surfaces and the inner capsule wall and a drop of 50 F across the width of the specimen at operating temperature. The helium annulus between the inner and outer capsules served as the primary thermal barrier and was expected to maintain a temperature differential of about 1300 F between the inner and outer capsule wall at operating conditions. The computation of these temperature drops included considerations for the effects of changes in thermal conductivity, radiation losses, and thermal expansion.

The four specimens were suspended in the capsule on hangers located between two vertical rods centered in the inner capsule. Approximately 100 cm^3 of NaK was loaded into the inner capsule under a helium atmosphere. The NaK was purified in a tilting furnace at 1700 F for 200 hr using zirconium foil as an oxygen getter. The final weld on the outer capsule was performed in a helium atmosphere. The completed capsule was then checked and found free of leaks.

Irradiation History

The irradiation of the capsule was performed in a single-hole A-piece in Position A-40-NE in the MTR. The quoted unperturbed peak flux for this position was 1.1×10^{14} nv. The quoted flux was based on the neutron flux measured in an adjacent position during preceding cycles. The capsule was inserted into the reactor on April 10, 1959, MTR Cycle 120. Upon initial startup, temperatures of the specimens were within the desired range without the use of auxiliary heat from the resistance heaters. Throughout the entire irradiation, consisting of almost four MTR cycles, it was necessary to supply auxiliary heat (20 to 30 w) on only two occasions, both of which occurred during the first cycle of operation.

From the reactor temperature charts, Figures 3, 4, 5, and 6, it is noted that Thermocouple 2, which measured the NaK temperature opposite the top specimen in the capsule, failed at 1360 F during reactor startup for the first cycle. The remaining thermocouples all performed satisfactorily during the remainder of this cycle. The temperatures observed on either side of the top specimen differed by only 20 F during the first cycle. On one occasion, this variation increased to about 40 F which was most likely due to thermocouple movement caused by vibration of the capsule produced by the flow of cooling water. During the four reactor scrams and one reactor shutdown that occurred during this cycle, the specimen temperatures dropped to reactor ambient temperature.



N56691

FIGURE 2. SIDE VIEW OF THE SPECIMEN ASSEMBLY PRIOR TO INSERTION IN CAPSULE BMI-28-1 SHOWING THE POSITIONS AND IDENTIFICATIONS OF THE DOSIMETERS, THERMOCOUPLES AND SPECIMENS

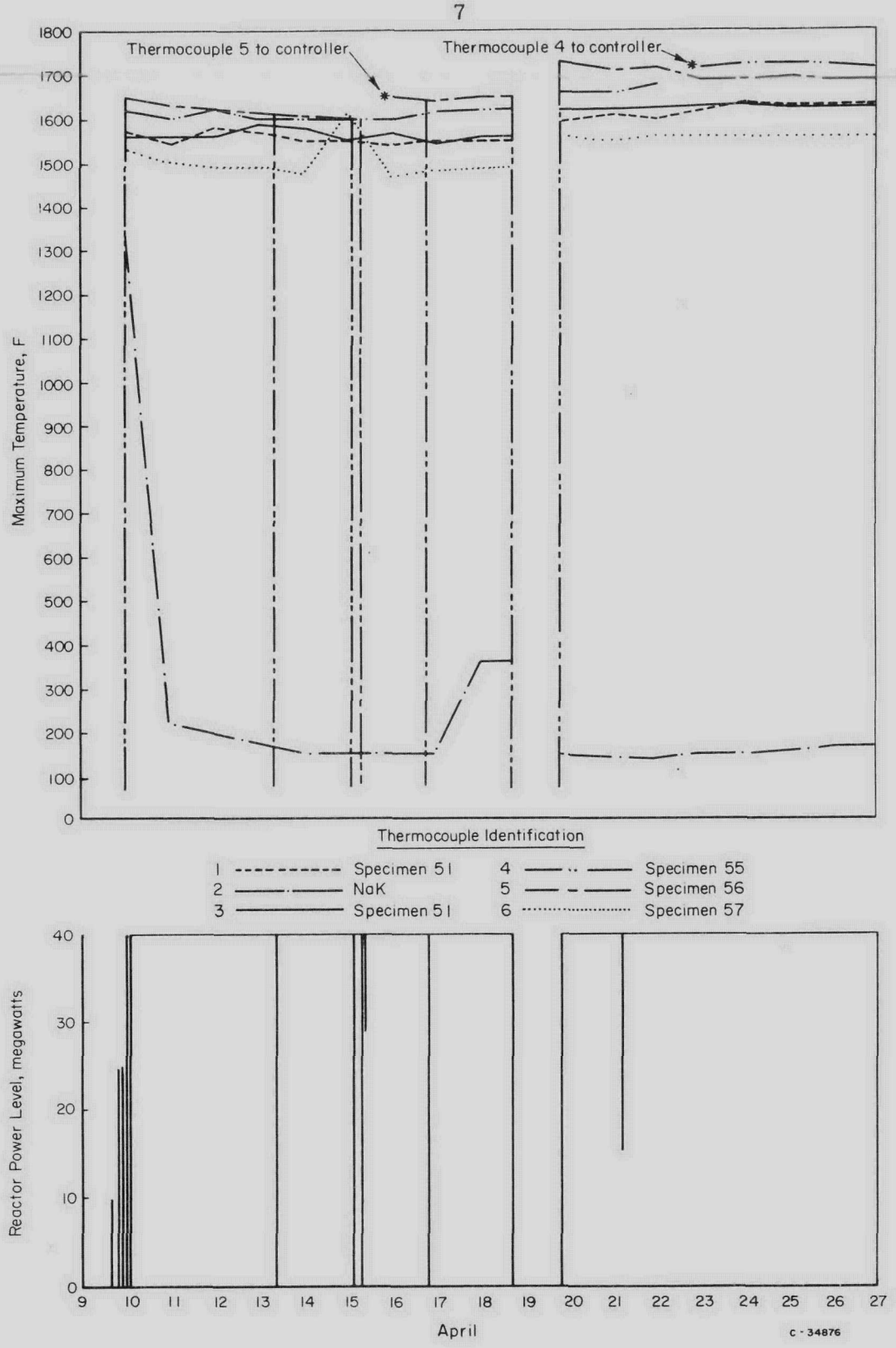


FIGURE 3. TEMPERATURE HISTORY OF CAPSULE BMI-28-1 DURING MTR CYCLE 120

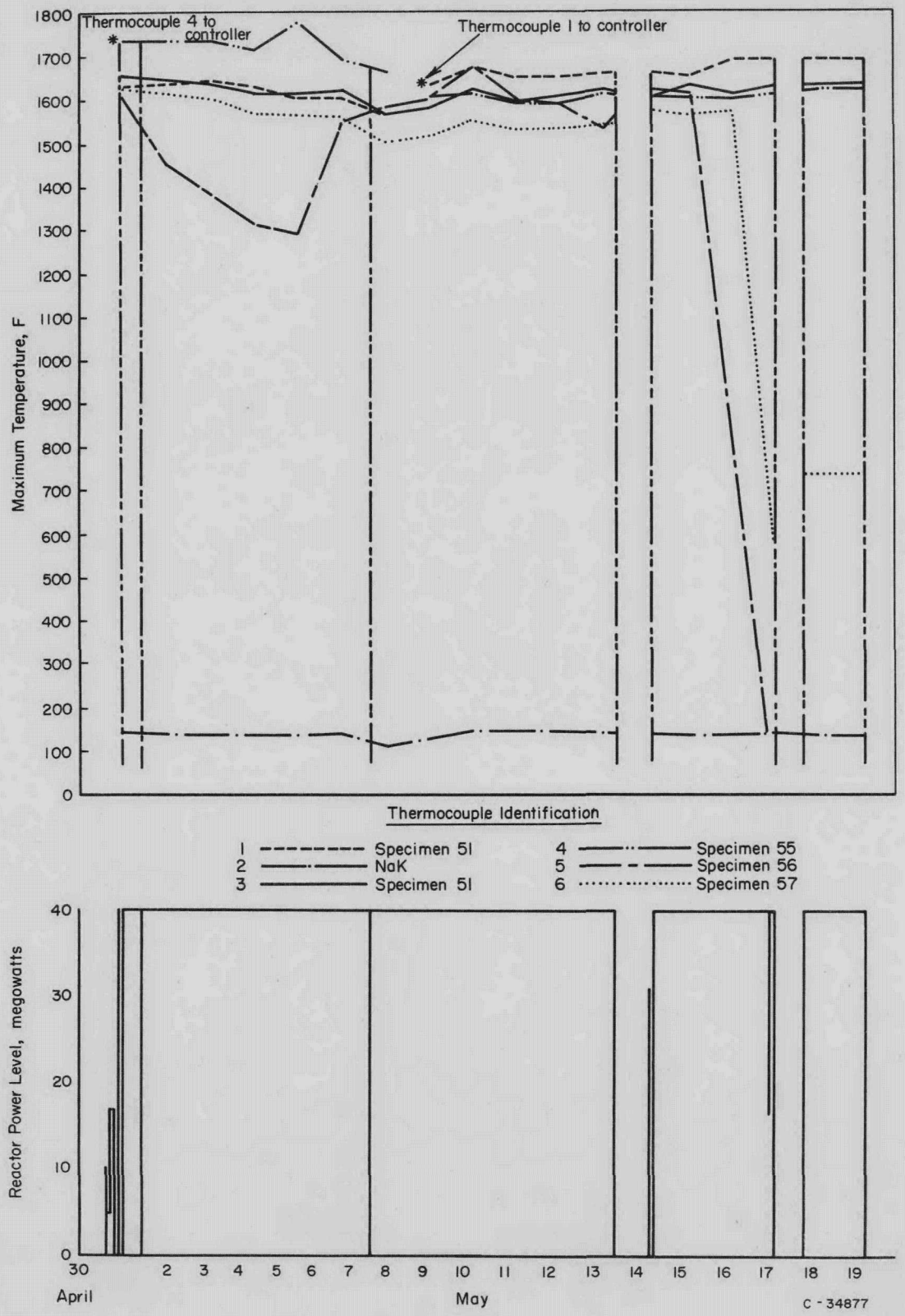


FIGURE 4. TEMPERATURE HISTORY OF CAPSULE BMI-28-1 DURING MTR CYCLE 121

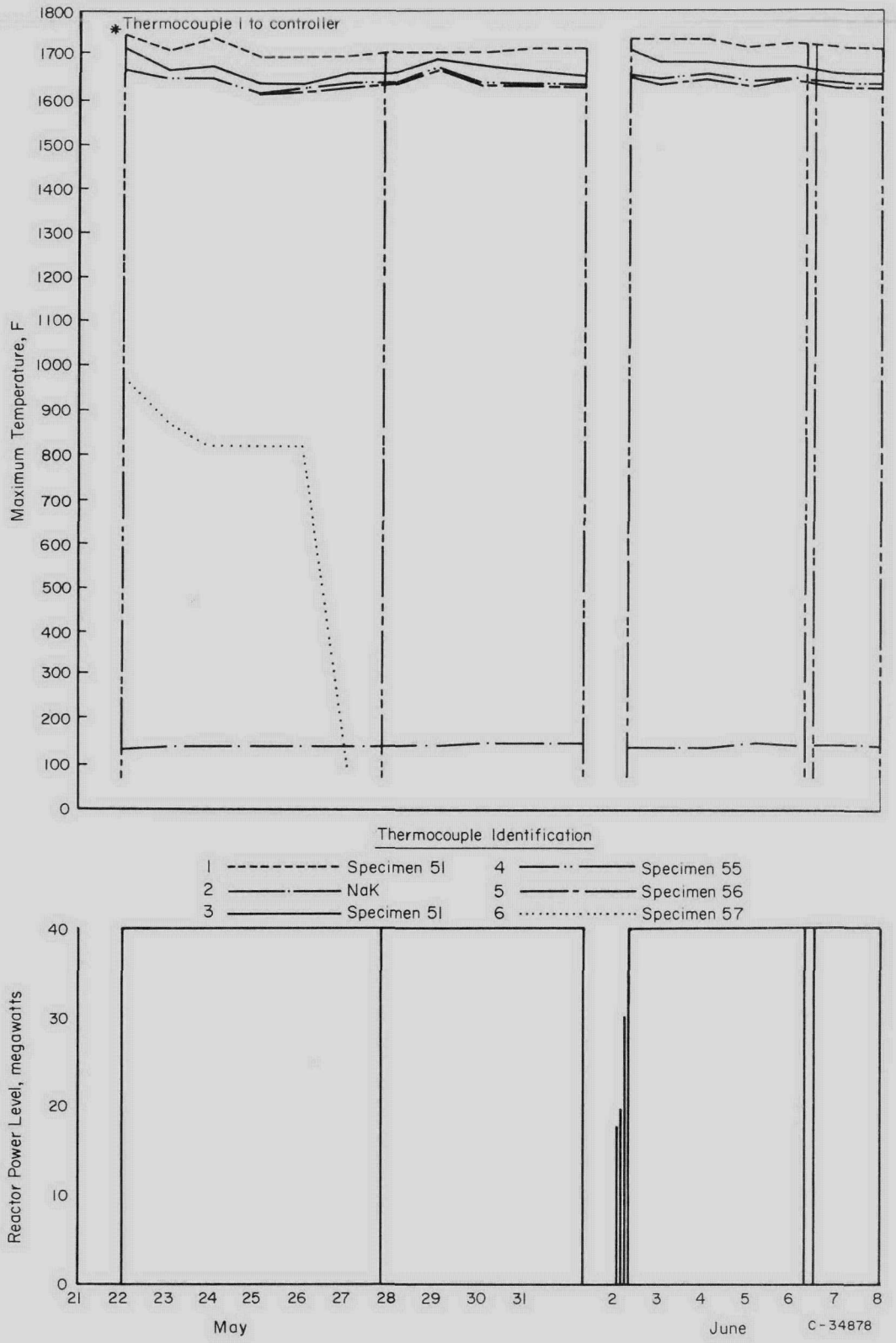


FIGURE 5. TEMPERATURE HISTORY OF CAPSULE BMI-28-1 DURING MTR CYCLE 122

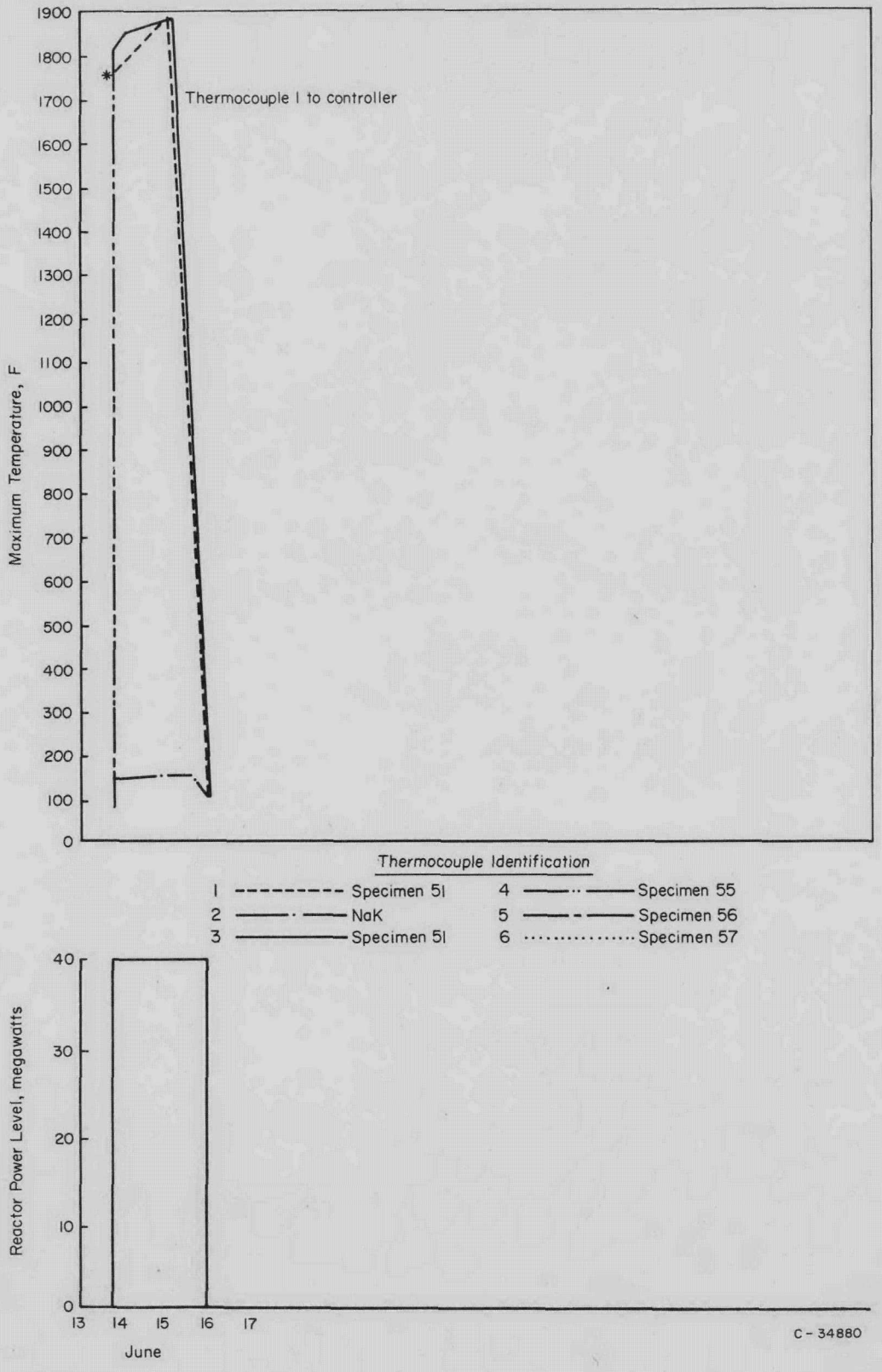


FIGURE 6. TEMPERATURE HISTORY OF CAPSULE BMI-28-1 DURING MTR CYCLE 123

Upon startup of the second cycle, MTR Cycle 121, Thermocouple 5, positioned adjacent to Specimen 56, recorded temperatures as low as 1300 F during the first 4 days of operation. However, normal operation of this thermocouple was resumed until the fourteenth day, at which time failure occurred. Thermocouple 6 began to operate erratically near the end of this cycle.

Upon startup for the third cycle, MTR Cycle 122, Thermocouple 5 failed to operate. However, on the third day of normal reactor operation, this thermocouple began to function properly and continued to do so throughout the entire cycle. Thermocouple 6, positioned adjacent to Specimen 57, continued to operate erratically and failed about 5 days after the reactor startup.

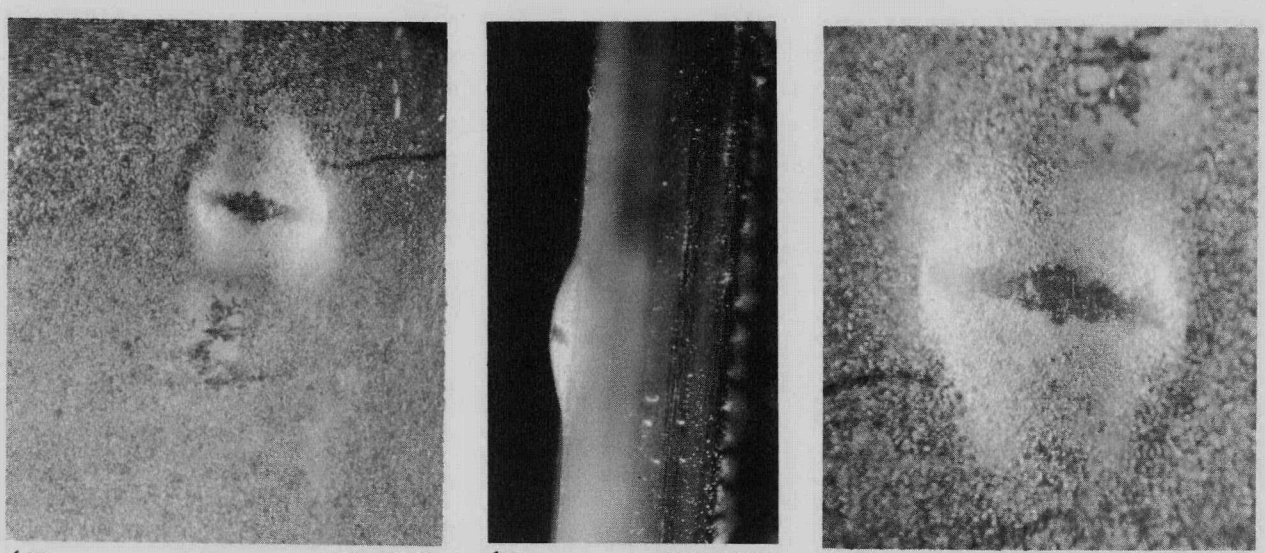
Upon startup for the fourth cycle, MTR Cycle 123, only Thermocouples 1 and 3 responded. Since the measured specimen temperatures were excessively high, apparently as a result of increased neutron flux, the capsule was discharged after only 2 days of operation in Cycle 123. The irradiation of the capsule was terminated before the target burnup of 12 a/o of the uranium-235 had been attained because it was felt that continued operation at temperatures in excess of 1800 F would seriously affect the specimens and confuse interpretation of the results. It was estimated that the specimen burnup was approximately 7.2 a/o of the uranium-235, which was high enough to be in the range of practical interest. The capsule was discharged on June 16, 1959, during a reactor scram.

Postirradiation Examination of Specimens

The opening of the capsule and the examination of the specimens were undertaken about 8 months after the irradiation was completed. The proposed examination was to include a measurement of the quantity of fission gas released from the specimens, measurements of specimen density and dimensions, visual inspection of the specimen surfaces, analytical determination of fuel burnup, analysis of dosimeter wires, and metallographic examination of the fuel core and cladding from selected specimens. Experimental details and the results of each phase of the postirradiation examination are discussed separately below.

Storage of Capsule BMI-28-1

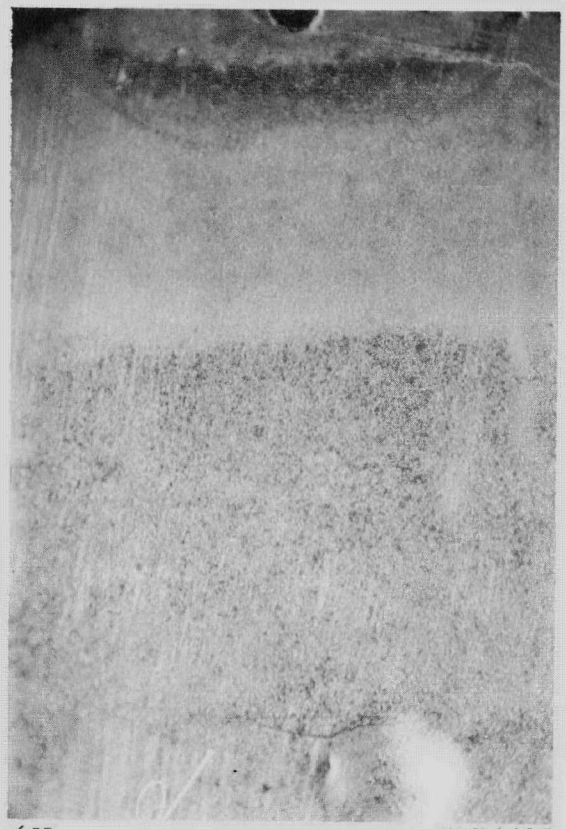
Capsule BMI-28-1 was discharged from the MTR on June 16, 1959, during MTR Cycle 123. The capsule was stored under water at the MTR until it was loaded into a cask during the first week of July. The dimensions of the interior cavity of the cask were about 6 in. in diameter by 48 in. long and no heat-transfer medium other than air was used to transfer fission-product-decay heat from the capsule. Experience with many such shipments has shown that sufficient heat is removed by convection and by direct contact between the capsule and cask to prevent the occurrence of excessive specimen temperatures. Shipments made directly after removal of a capsule from the reactor would, of course, present a more severe problem. The capsule was received at the Hot-Cell Facility on July 16 whereupon it was unloaded and placed in storage in static air in the hot cell. The capsule was removed from storage about the middle of February, 1960. It is believed that neither shipment or storage of the capsule created any problems in regard to the production of significant temperatures as a result of the radioactive decay of fission products.



6X HC4428 6X HC4433 12X HC4430

FIGURE 7. CONDITION OF BLISTERED AREA ON SPECIMEN 51 (UO_2)

The specimen was irradiated at 1500 to 1880 F to a burnup of 3.6 a/o uranium-235.



6X HC4435

12X HC4436

FIGURE 8. CONDITION OF BLISTERED AREA ON SPECIMEN 55 (UN)

The specimen was irradiated at 1575 to >1800 F to a burnup of 4.8 a/o uranium-235. Note the rough surface of the cladding over the fueled area and the apparent crack near the blister. Thorough examination of the blistered area failed to establish definitely whether the mark near the blister was a scratch or crack.

6X HC4440

12X HC4444

FIGURE 9. CONDITION OF BLISTERED AREA ON SPECIMEN 56 (UN)

The specimen was irradiated at 1590 to >1850 F to a burnup of 5.0 a/o uranium-235. The blister occurred near the top end of the specimen. The mark near the blister is believed to be a surface scratch.

6X HC4447

12X HC4449

FIGURE 10. CONDITION OF BLISTERED AREA OF SPECIMEN 57 (UN)

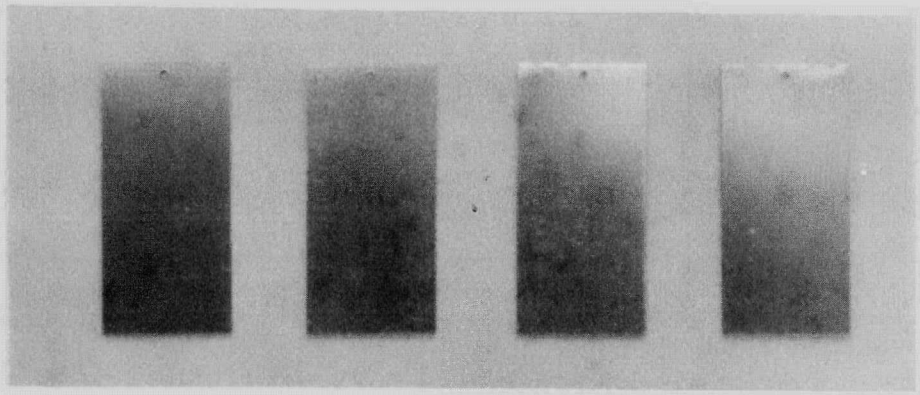
The specimen was irradiated at 1460 to >1750 F to a burnup of 4.3 a/o uranium-235. This blister occurred near the edge of the fueled core. The cause of the light parallel marks on the specimen surface is unknown.

Capsule Opening and Fission-Gas Sampling

The outer capsule shell was removed using a remotely operated power pipe cutter. An attempt was made to puncture the inner capsule shell to permit sampling of the gases released from the specimens. The gas sampling was unsuccessful because of the failure of a seal between the sampling system and the capsule which permitted gases contained in the capsule to leak to the atmosphere. The opening of the inner capsule was accomplished by cutting through the top of the capsule with the pipe cutter. The capsule was then lowered into a butyl alcohol bath to permit reaction of the NaK with the alcohol. After the NaK-alcohol reaction was complete, samples of the sodium-potassium butoxide solution were obtained for fission-product analysis, and the specimen-holder assembly was removed from the inner capsule. The dosimeters and specimens were removed from the hanger assembly. The dosimeters were sectioned into lengths corresponding to the position of each specimen within the capsule. The specimens were cleaned and identified in preparation for visual examinations.

Visual Examination

After the specimens were removed from the capsule, they were rinsed in fresh butyl alcohol and dried. Each specimen was then thoroughly examined at magnifications up to 12X with the aid of a stereomacroscope. A small blister was observed on one surface of each specimen. Photographs of the blistered area on each specimen were prepared, and are shown as Figures 7, 8, 9, and 10. In some cases, Figure 7, an apparent crack was observed in the cladding near the blister on the UO_2 dispersion specimen. A thorough examination of the specimen surface failed to positively identify the mark as either a crack or scratch. The cladding surface on each specimen, directly over the fueled core, was observed to be considerably roughened as compared with the surface of the nonfueled stainless steel frame around the edges of each specimen. The condition of the specimens before irradiation is shown in Figure 11 for comparison purposes.



1X N55133

FIGURE 11. APPEARANCE OF SPECIMENS 51 (UO_2), 55 (UN), 56 (UN), AND 57 (UN) PRIOR TO IRRADIATION IN CAPSULE BMI-28-1

Analysis of the NaK

Samples of the solution remaining after the NaK had reacted with the butyl alcohol were analyzed for cesium-137 content. The results of the analysis indicated that about 4.6×10^{16} atoms of cesium-137 (33-year half-life) were present in the NaK which corresponded to about 2.9×10^{-6} g of uranium-235. This quantity of uranium-235 could have appeared from three different sources: uranium contamination on the surfaces of the specimens prior to irradiation; exposure of the fuel core through cracks in the cladding of the specimens; and diffusion of cesium-137 through the cladding.

The specimens were cleaned prior to irradiation by immersion in a boiling HNO_3 solution for three separate 30-min periods. Each solution was then analyzed for uranium. The results indicated that the quantity of uranium removed from the specimen surfaces by the third treatment was about 10 μg or less. If the quantity of uranium-235 found in the NaK is adjusted for burnup, then the amount of uranium found in the NaK would correspond to the quantity removed from the surface of the unirradiated specimen during the third treatment in the boiling HNO_3 solution. It is therefore probable that the presence of cesium-137 in the NaK was a result of uranium surface contamination and possibly diffusion through the cladding rather than leakage through cladding cracks. In other studies where similar specimens were irradiated at temperatures in the 1500 F range, considerably larger amounts of fission products escaped from specimens with small cladding cracks than were observed in this irradiation.

Neutron Dosimetry

Neutron dosimeters fabricated from nickel-0.6 w/o cobalt were positioned on the outer capsule wall and also adjacent to the specimens. The inner dosimeter wire was located about 1/4 in. from the specimen surface as shown in Figure 2. This wire was cut into sections the length of which corresponded to the adjacent specimen. Each section was then radiochemically analyzed to determine the quantity of cobalt-60 formed during the irradiation. The thermal-neutron flux to which the dosimeters were exposed was then calculated. The effective neutron flux incident on the fuel specimens was then calculated using the semiempirical method of W. B. Lewis[2] and a Battelle correlation factor of 0.7. The effective thermal-neutron flux calculated in this manner is presented in Table 1.

Physical Dimensions

The physical dimensions and density of each specimen were measured before and after irradiation. Dimensions were measured with standard friction-thimble micrometers to at least ± 0.0005 in. Densities were measured by standard immersion techniques in carbon tetrachloride at 25 C to at least ± 0.03 g per cm^3. The data are presented in Table 2.

Burnup Analyses

The fuel burnup of each specimen was determined by three different methods: (1) neutron dosimetry; (2) radiochemical analysis for cesium-137; (3) isotopic analysis The results obtained from all three methods are presented in Table 1. In the case of the

TABLE 1. IRRADIATION CONDITIONS AND EFFECTS

Specimen	Effective Thermal-Neutron Flux From Dosimetry, 10^{13} nv	Specimen Surface Temperature Range (From Reactor Temperature Charts), F	Burnup by Isotopic Analyses, a/o uranium-235	Burnup by Cesium-137 Analyses, a/o uranium-235	Burnup by Dosimetry, a/o uranium-235
51 (UO_2)	3.5	1500-1880[a]	3.6	3.5	5.3
55 (UN)	3.1	1575-1650[b]	4.8	5.3	6.2
56 (UN)	3.5	1590-1680[c]	5.0	4.8	5.5
57 (UN)	2.8	1460-1590[d]	4.3	5.0	4.9

(a) The two thermocouples for Specimen 51 operated properly during all of the irradiation. The maximum temperature occurred during the fourth cycle whereupon the capsule was discharged from the MTR.

(b) The thermocouple for Specimen 55 operated properly for three MTR cycles. The maximum temperature could have reached 1900 to 2000 F during the fourth cycle.

(c) The thermocouple for Specimen 56 operated properly for one MTR cycle. The maximum temperature could have reached 1900 to 2000 F during the fourth cycle.

(d) The thermocouple for Specimen 57 operated properly for two MTR cycles. The maximum temperature could have reached 1800 to 1900 F during the fourth cycle.

TABLE 2. CHANGES IN PHYSICAL DIMENSIONS OF SPECIMENS IRRADIATED IN CAPSULE BMI-28-1

Specimen	Preirradiation Dimensions		Postirradiation Dimensions		Dimensional Change, per cent		Thickness at Blister, in.	Change in Thickness at Blister, per cent
	Thickness, in.	Density, g per cm^3	Thickness, in.	Density, g per cm^3	Thickness	Density		
51 (UO_2)	0.0461	8.11	0.0462	8.01	0	-1.2	0.0598	30
55 (UN)	0.0476	8.22	0.0468	8.11	0	-1.3	0.0614	29
56 (UN)	0.0478	8.22	0.0465	8.10	0	-1.5	0.0598	25
57 (UN)	0.0463	8.22	0.0461	8.17	0	-0.6	0.0567	23

radiochemical and isotopic analyses, small cross sections removed from the approximate longitudinal center of the specimens were analyzed. The sections used for the cesium-137 determination and isotopic analyses were shipped to the Phillips Petroleum Company, Chemical Processing Plant, at the NRTS where they were dissolved and analyzed. The results of the isotopic analyses are presented in Table 3. The relative burnup, Table 1, was calculated from the relationship

$$\text{Relative burnup} = \frac{(1 + \alpha)(E_0 - E_1)}{E_0 (1 + \alpha - E_1)},$$

where

α = the ratio of cross sections for fission and capture in uranium-235 = 0.185 for the MTR

E_0 = fraction of uranium-235 present before irradiation

E_1 = fraction of uranium-235 present after irradiation.

TABLE 3. RESULTS OF ISOTOPIC ANALYSES OF SPECIMENS IRRADIATED IN CAPSULE BMI-28-1

Specimen	Fraction of Uranium Isotope Present, per cent			
	Uranium-234	Uranium-235	Uranium-236	Uranium-238
51 (UO_2)	0.93	92.23	1.21	5.64
52 (UO_2, unirradiated)	0.95	92.97	0.52	5.56
55 (UN)	1.03	91.90	1.05	6.02
56 (UN)	1.01	91.87	1.08	6.04
57 (UN)	0.99	92.02	1.05	5.94
58 (UN, unirradiated)	1.00	92.92	0.30	5.78

Metallographic Examination

Transverse cross sections of selected unirradiated specimens were mounted in Bakelite for metallographic preparation. These were prepared by grinding through 240, 400, and 600-grit SiC paper and then polishing on red felt with 0 to 2-μ diamond abrasive, on black Forstmann's cloth with a Linde B and 3 per cent CrO_3 abrasive water slurry and finally on Microcloth with Linde B abrasive in water. The specimens were examined in the as-polished condition and after swab etching with a solution of 20 cm^3 of glycerine, 20 cm^3 of HCl, and 5 cm^3 of HNO_3.

The irradiated sections were mounted in Bakelite and ground through 600-grit SiC paper using kerosene as a lubricant. Each specimen was then polished on Syntron vibratory polishers using water slurries of 1, 0.3, and 0.1-μ alumina on Microcloth.

The specimens were then examined at magnifications up to 500X in the as-polished condition and after swab etching with the solution used on the unirradiated specimens.

A structure representative of the 30 w/o UO_2 dispersion is shown in Figure 12 prior to irradiation. The structure of the material after irradiation is shown in Figure 13. In general, no structural changes were detected by light microscopy in the UO_2 and stainless steel that could have resulted from the effects of irradiation or reaction between the UO_2 and stainless steel. However, a study of the contact areas between the UO_2 particles and matrix by electron microscopy indicated that a phase or zone surrounding the unirradiated UO_2 particles had disappeared during irradiation, Figure 14. This was apparently a zone of reaction between excess oxygen in the UO_2 and constituents of the stainless steel matrix and appeared to follow grain boundaries leading away from the UO_2 particles into the matrix. The zone was apparently formed during fabrication and its disappearance during irradiation could be due to diffusion and fission-fragment bombardment effects.

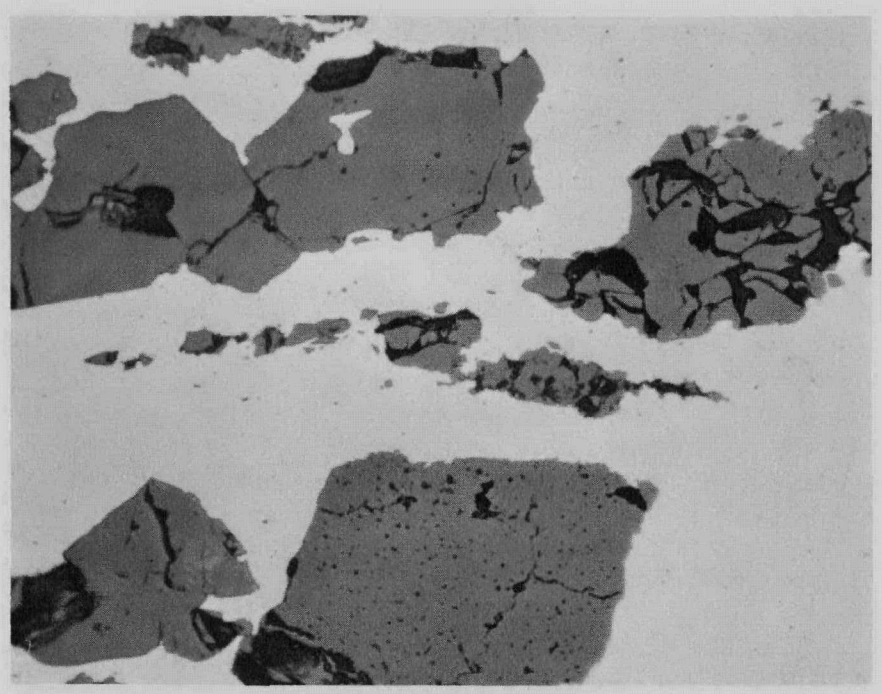
The typical appearance of the microstructure of UN dispersions in Type 318 stainless steel before irradiation is shown in Figure 15. A dark gray-colored reaction zone was observed on the surface of the UN particles, which was apparently formed during fabrication. The UN particles did not appear porous. The gray reaction zone still appeared around the UN particles after irradiation and porosity was observed, Figures 16 and 17. A small zone in the matrix around each fuel particle was rapidly attacked by the etchant after irradiation. This compares with the results reported by others[3] in the case of irradiated UO_2 dispersions.

The contact area between the UN particles and the matrix was also studied by electron microscopy. A slight porosity or reaction zone was noted between the UN particles and the matrix before irradiation, Figure 18. The recessed area around the fuel particles after irradiation as produced by the etchant was also apparent.

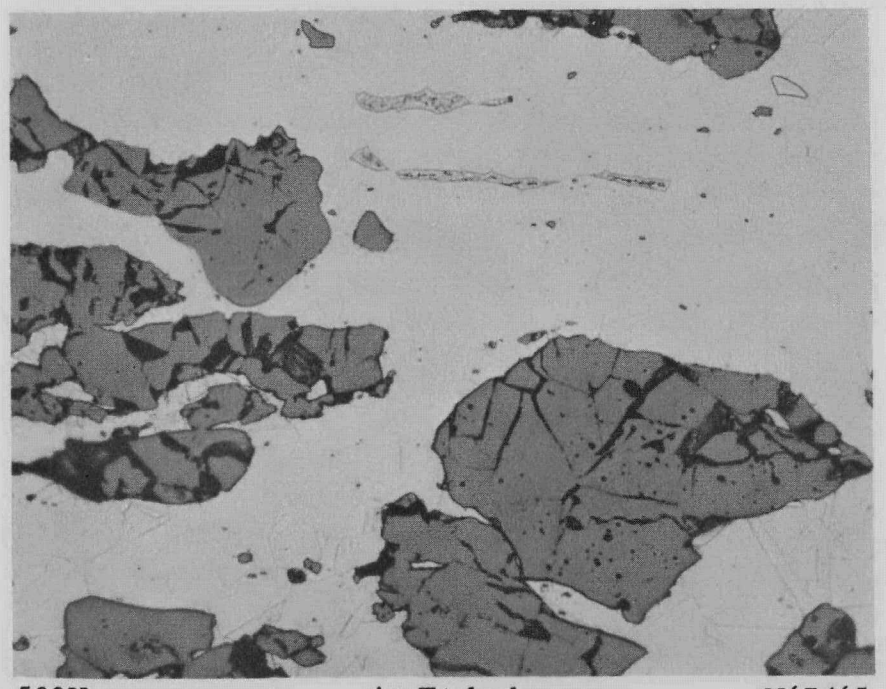
The typical microstructure of an area directly beneath a blister is shown in Figure 19. The separation obviously occurred in the fuel matrix rather than at the core-cladding bond. The crack also appears to follow along the edges of fuel particles. This type of blistering has been observed in other irradiations of dispersion and alloy fuel materials.

DISCUSSION

The formation of small blisters on the surface of both the UO_2 and UN dispersion specimens is a phenomenon that has been observed in other irradiations of similar materials at Battelle. There does not seem to be a rational explanation for the blister formation with regard to radiation damage or fission-gas agglomeration. There was no observable swelling in the UO_2 or UN particles, although porosity did increase in the UN particles during irradiation. There was no observable damage to the matrix. Metallographic inspection of the blistered areas merely indicated that a fissure had occurred in the core material slightly below the core-cladding bond zone. Accumulation of gases and/or thermal stresses then combined to enlarge the fissure into the blister or pimple formation. It does not appear likely that sufficient fission gas would have escaped from the fuel particles and agglomerated in the matrix to cause such a large expansion.



500X As Polished N67464

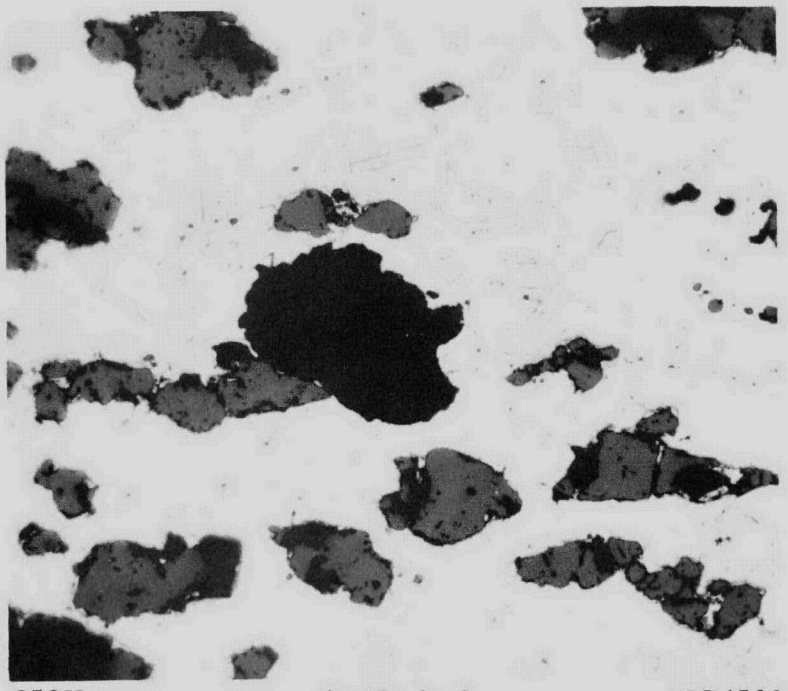


500X As Etched N67465

FIGURE 12. TYPICAL MICROSTRUCTURE OF UNIRRADIATED SPECIMEN OF 30 w/o UO_2 DISPERSED IN TYPE 318 STAINLESS STEEL

Swab etched with 20 cm^3 glycerine, 20 cm^3 HCl, and 5 cm^3 of HNO_3 solution.

250X As Polished HC4531



250X As Etched HC4510

FIGURE 13. TYPICAL MICROSTRUCTURE OF 30 w/o UO_2 DISPERSION IN TYPE 318 STAINLESS STEEL AFTER IRRADIATION, SPECIMEN 51

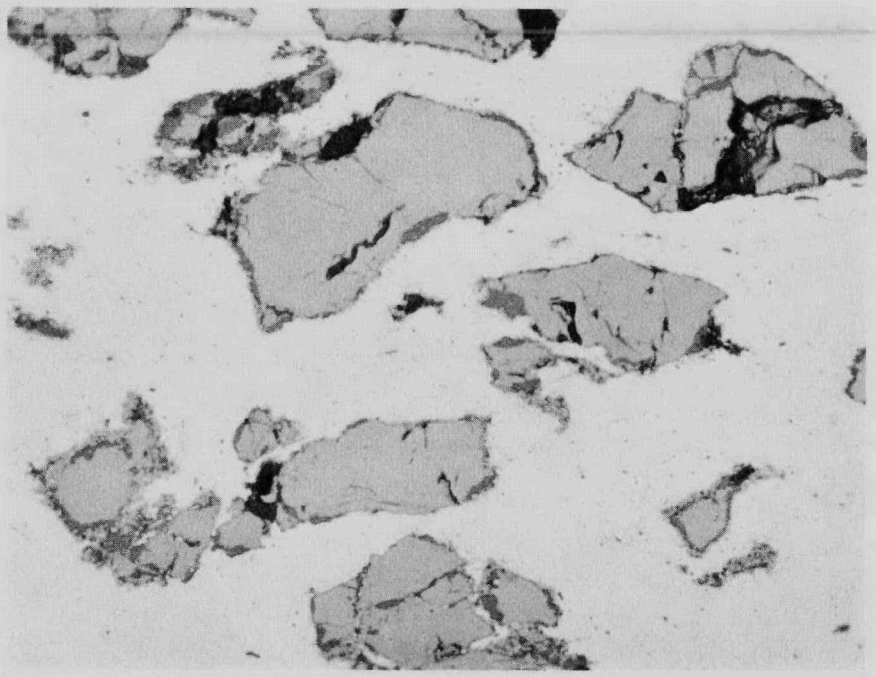
Burnup was 3.6 w/o of uranium-235.

6000X J324
Positive Replica of Unirradiated Specimen 52

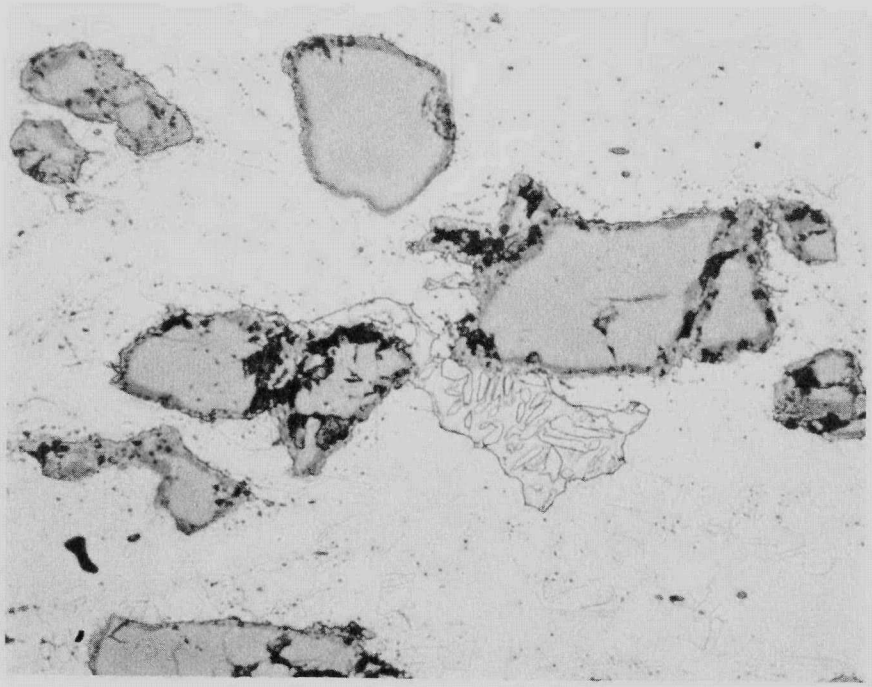
6000X J266
Negative Replica of Irradiated Specimen 51

FIGURE 14. STRUCTURE OF CONTACT ZONE BETWEEN A UO_2 PARTICLE AND THE STAINLESS STEEL MATRIX IS SHOWN BEFORE AND AFTER IRRADIATION

Note that the phase surrounding the UO_2 particle in the unirradiated condition does not appear in the irradiated specimen. This phase is believed to be a reaction zone formed during fabrication between the matrix and the excess oxygen in the UO_2 particles.



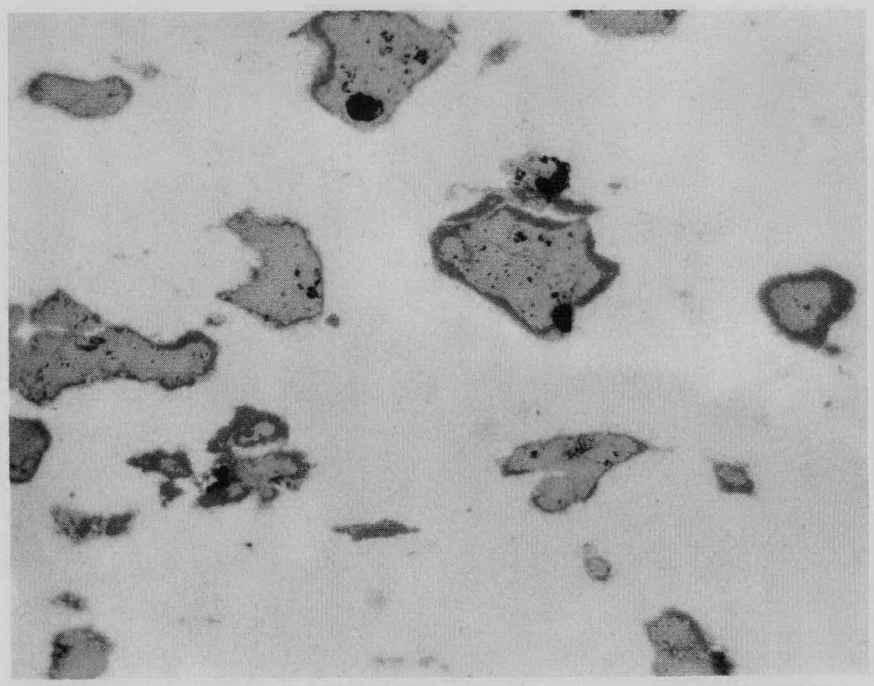
500X As Polished N67470



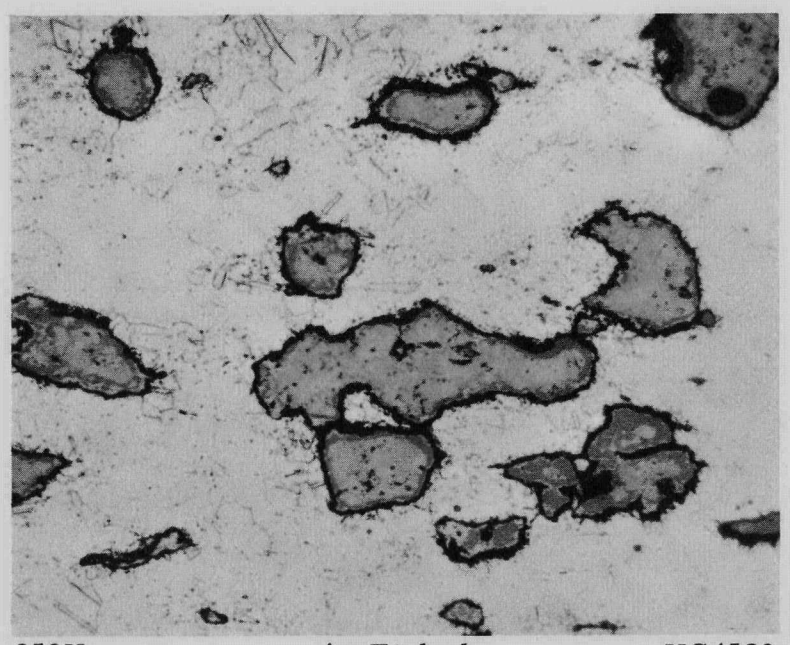
500X As Etched N67468

FIGURE 15. TYPICAL MICROSTRUCTURE OF UNIRRADIATED 28 w/o UN DISPERSIONS IN TYPE 318 STAINLESS STEEL

Note the appearance of the dark gray reaction zone at the surface of the particles.



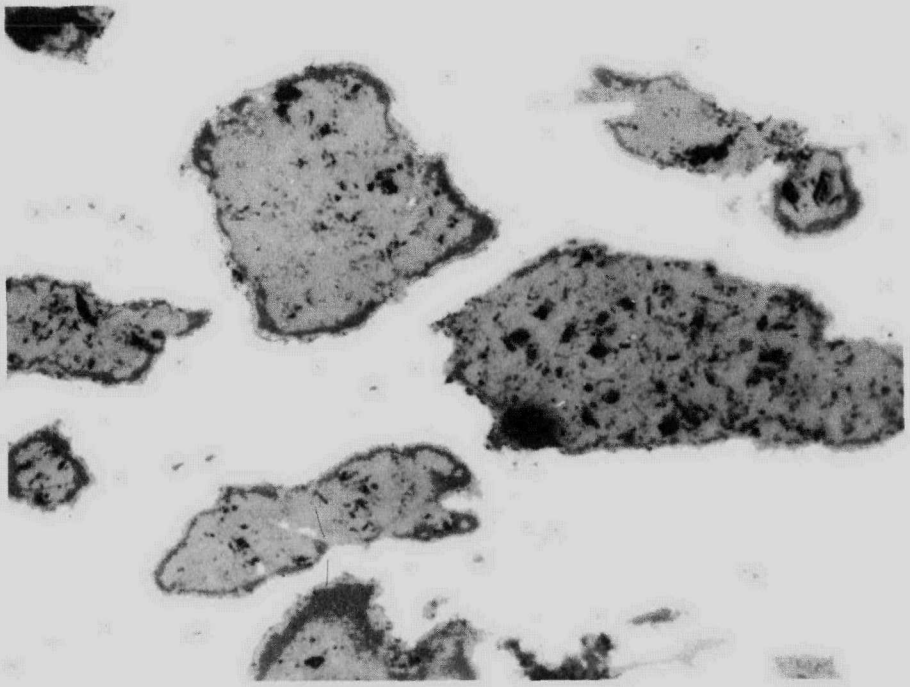
250X As Polished HC4515



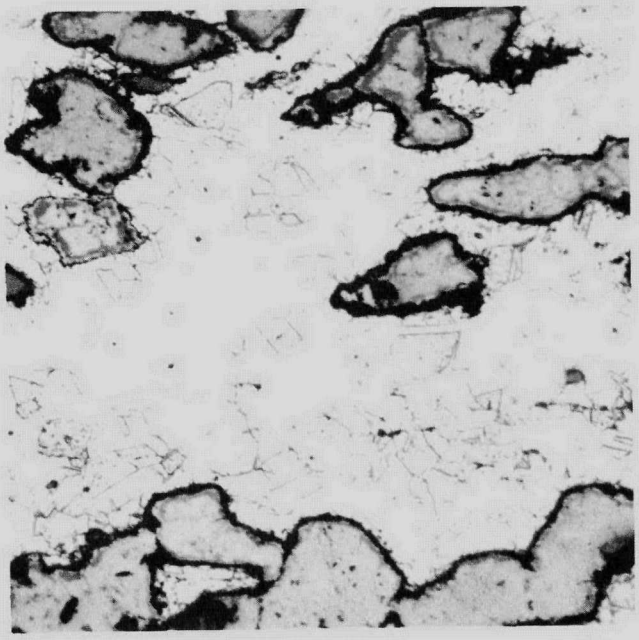
250X As Etched HC4520

FIGURE 16. TYPICAL MICROSTRUCTURE OF IRRADIATED 28 w/o UN DISPERSED IN TYPE 318 STAINLESS STEEL, SPECIMEN 55

Note the porosity formed in the UN and the continued presence of the gray zone around the UN particles. During etching, it was found that a thin zone of the matrix surrounding the fuel particles was rapidly attacked and removed by the etchant. Burnup was 4.8 a/o of uranium-235.



500X As Polished HC4536

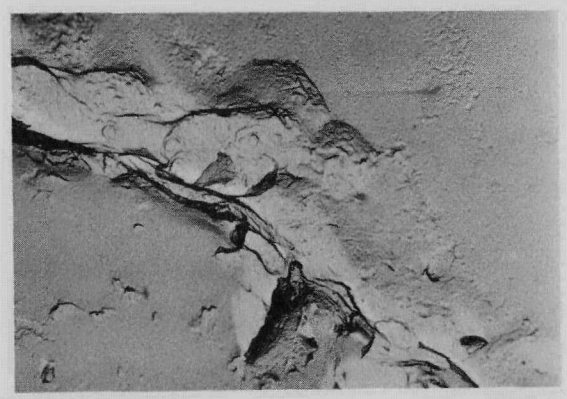


250X As Etched HC4524

FIGURE 17. TYPICAL MICROSTRUCTURE OF IRRADIATED 28 w/o UN DISPERSED IN TYPE 318 STAINLESS STEEL, SPECIMEN 56

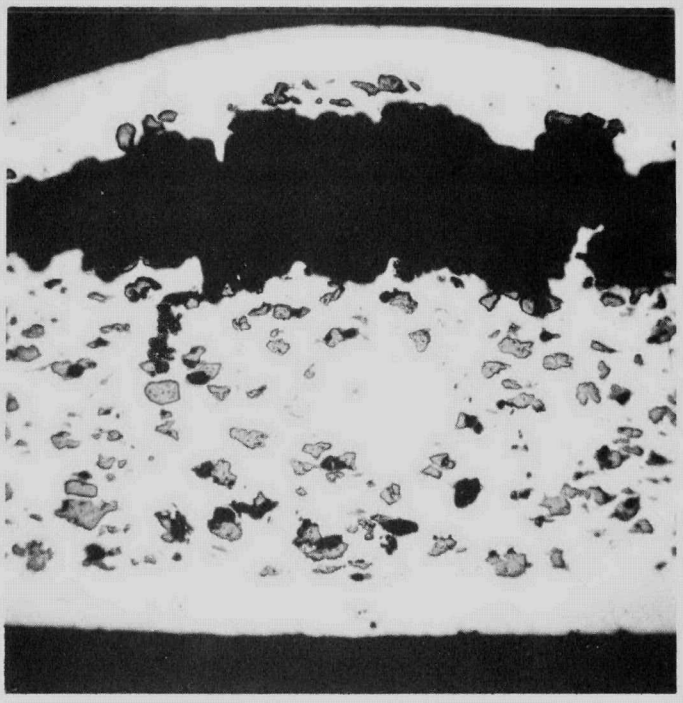
Note the porosity in the UN particles and the continued presence of the gray zone around each particle in the as-polished condition. Etching the specimen resulted in the removal of a thin layer of the matrix surrounding the fuel particles. Burnup was 5.0 a/o uranium-235.

6000X J330
Positive Replica of Unirradiated Specimen 58

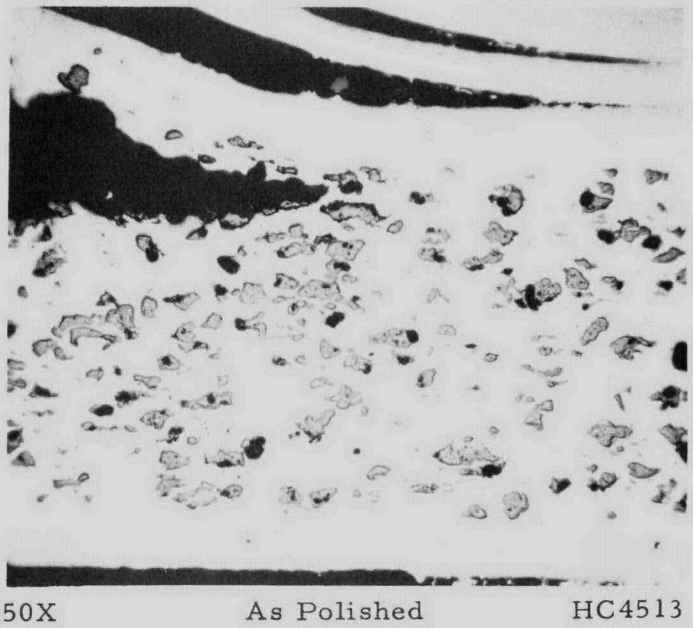


6000X J275
Negative Replica of Irradiated Specimen 56

FIGURE 18. TYPICAL MICROSTRUCTURE OF IRRADIATED AND UNIRRADIATED 28 w/o UN DISPERSED IN TYPE 318 STAINLESS STEEL AS OBSERVED BY ELECTRON MICROSCOPY



50X As Polished HC4512



50X As Polished HC4513

FIGURE 19. AREA UNDER BLISTER ON SPECIMEN 55

Note that the separation occurred in the core.

The most likely explanation therefore probably lies somewhere in the techniques of fabrication of these materials.

The cladding surface directly over the fueled core was observed to be roughened when compared with the surface located over the unfueled portion. This probably illustrates a slightly more rapid corrosion of the area over the fueled core since this area would be expected to experience higher temperatures than those occurring outside the fueled area. During metallographic examinations, the surfaces of the specimens were observed to be slightly pitted but there was no observable intergranular attack. There was no positive identification of cracks in the cladding by either visual or metallographic examinations. The results of the analysis of the NaK for cesium-137 indicate that cracks did not occur in the cladding during irradiation.

Measurements of physical dimensions indicated that little swelling occurred in the specimens during irradiation except at the location of the blisters. A decrease in density was noted, principally as a result of the blister formation.

Measurements of the specimen temperature during irradiation indicated that temperatures were maintained at fairly constant levels, in the range of 1500 to 1750 F, for the first three MTR cycles. Some of the indicated thermocouple readings were erratic but this is attributed to intermittent shorts in the thermocouples rather than fluctuations in specimen temperature. An increase in specimen temperatures to the 1800 to 1900 F region was noted for Specimen 51 during the startup for the fourth MTR cycle. It is estimated that temperatures of Specimens 55 and 56 could have reached 1900 to 2000 F during this period and Specimen 57 could have reached 1800 to 1900 F. Operation at these temperatures continued for the first 2 days of the cycle whereupon the capsule was discharged. It is believed that shifting of reactor fuel elements or other experiments during the Cycle 123 shutdown increased the neutron flux incident on the specimens and produced the higher temperatures. It is probable that the blisters in the specimens were formed during this period of high temperature.

Fuel burnups (fission plus absorption), determined by three different methods, were reasonably similar. However, the results of the isotopic analyses are considered to be the most accurate.

Examination of the microstructure of the UO_2 dispersions by light microscopy indicated that the UO_2 particles were slightly fragmented prior to irradiation. The distribution of the fuel particles within the matrix appeared to be uniform without noticeable stringering or other defects. The matrix appeared sound and no reaction between the UO_2 particles and the Type 318 stainless steel matrix was observed. There was only a very slight porosity visible in the UO_2 particles. After irradiation, neither the fuel particles or the matrix appeared changed, except for the blisters discussed earlier.

Examination of the UO_2 dispersions by electron microscopy before irradiation indicated the existence of a very small separate and distinct zone around the larger fuel particles and continuing outward into the matrix along grain boundaries. This zone was apparently the result of reactions between the constituents of the stainless steel matrix and excess oxygen in the UO_2. Similar examinations of these specimens after irradiation indicated that this zone had apparently disappeared, possibly as a result of the heavy bombardment of this area by fission fragments and diffusion.

A study of the microstructure of unirradiated UN dispersions by light microscopy indicated that the fuel particles were evenly distributed without any evidence of stringering. Only very occasional fractures were observed in the fuel particles. There was no indication of any porosity in the fuel particles and the Type 318 stainless steel matrix appeared sound. A gray area was observed on the surface of the UN particles which may have resulted from gettering of oxygen from the matrix by the UN. After irradiation, the gray area was still visible. A slight porosity was also observed in the irradiated fuel particles which could have resulted from expansion of fission gases. The matrix appeared to be in good condition, except for the blisters as discussed earlier. Chemical etching of the irradiated specimens produced a pitted and eroded area in the matrix surrounding the fuel particles. The rapid attack of this area by the etchant was probably caused by a reaction between the stainless steel constituents and free nitrogen produced from the fission of UN and the damage caused by fission fragments attenuated in the small volume of matrix surrounding each fuel particle.

Examination of the microstructure of the UN dispersions by electron microscopy did not indicate any differences between the unirradiated and irradiated condition except for the noted zone around each fuel particle.

CONCLUSIONS

Although it is difficult to evaluate the relative worth of UN dispersions in Type 318 stainless steel as compared with UO_2 dispersions in the same material after the irradiation of only four specimens, the results of this study indicate, in general, that the UN dispersions withstood irradiation at temperatures of 1500 to 2000 F and at burnups of 3.5 to 5.0 a/o of the uranium-235 at least as well as the UO_2 dispersions. Although a slight amount of porosity was observed in the UN particles, there is not enough evidence available to determine if this is the forewarning of swelling in the fuel. Additional irradiations should be performed in order to fully evaluate this possibility.

The major advantages of using UN as a dispersion fuel in stainless steel in comparison with UO_2, lie in the increased uranium density and the favorable properties of UN during fabrication. If the radiation stability of the two fuel dispersions is reasonably similar, then the advantages of using UN become much greater.

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(2) Lewis, W. B., "Flux Perturbations by Material Under Irradiation", Nucleonics, 13 (10), 82 (October, 1955).

(3) Barney, W. K., and Wempler, B. D., "Metallography of Irradiated UO_2-Containing Fuel Elements", KAPL-1836 (January, 1958).