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EVALUATION OF IRRADIATED EXPERIMENTAL OMR FUEL ELEMENTS I.

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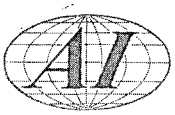


ABSTRACT

As part of a program to develop an economical fuel cycle for an Organic Moderated Reactor (OMR), experimental fuel elements are being irradiated in the Organic Moderated Reactor Experiment (OMRE). The first two experimental fuel elements have been removed from the OMRE, examined, and evaluated. These extended-surface, plate-type fuel elements had finned aluminum cladding, metallurgically bonded to flat, uranium alloy fuel plates. A 0.0005 in. nickel layer was used to prevent interdiffusion between the aluminum and uranium. The fuel core alloys irradiated were U - 3.5% Mo and U - 3.5% Mo - 0.5% Si.

These two experimental elements were removed from the reactor, after one of the elements released fission products, and cladding temperatures increased on both fuel elements. The maximum measured cladding temperatue was 780°F. The maximum fuel burnup was later determined to be 0.23 at.% U.

Hot cell examination revealed that the coolant inlet ends of the fuel elements had filtered particulate matter out of the coolant, which blocked the ends of the coolant channels. Restriction of the coolant flow through the elements caused partial melting of the cladding in one fuel element, and a blister on one of the fuel plates from the other fuel element. Dimensions were taken on three of the fuel plates. Metallographic sections from the blistered fuel plates were examined, and burnup profiles were determined for the three plates measured. The fuel had good dimensional stability, which is significant, since the melted cladding and the core microstructure evidenced temperatures substantially above the maximum temperatures recorded by thermocouples. Nothing was observed that would indicate the fuel elements would not have functioned properly in the same coolant, if the coolant were free of particulate matter of sizes that could be trapped.

I. INTRODUCTION

Organic cooled and moderated reactors are promising systems for the production of economic power. The Organic Moderator Reactor Experiment (OMRE) has demonstrated the feasibility of using hydrocarbons as the moderator-coolant, and provides a test facility for investigating the behavior of organic moderator-coolants under power reactor conditions.[1,2] Since the OMRE is principally a test of the organic moderator-coolant, a fuel element type of proven radiation stability and fabrication feasibility was selected for the first core loadings. These plate-type fuel elements of stainless steel-clad, highly enriched UO_2 - stainless steel cermet fuel, are not economically feasible for power reactors.[3] As part of a program to develop an economic fuel cycle for an Organic Moderated Reactor (OMR),[4] experimental fuel elements of advanced designs are being irradiated in the OMRE. This report covers the evaluation of the first two experimental fuel elements removed for hot cell examination.

The two fuel elements irradiated were of the plate type, with an extended-surface or "finned" aluminum cladding, metallurgically bonded to a uranium alloy core. The design is similar to the one originally proposed for the 45-Mwt Piqua OMR, presently under construction. The final Piqua fuel element design utilizes the same materials, but has a cylindrical configuration. Aluminum was chosen as the cladding material because of its low absorption cross section for thermal neutrons, compatibility with the organic coolant,[6,7] ability to be bonded to the uranium alloy core, and ease of fabrication. The low mechanical strength of aluminum at elevated temperature is compensated for by the design, in which the aluminum is treated as a skin which is not highly stressed in the fuel element assembly. The fins on the aluminum cladding increase the surface area for the transfer of heat to the organic coolant.

The cladding is metallurgically bonded to the core with a thin intermediate nickel layer.[8] The nickel layer acts as a barrier to prevent interdiffusion of the aluminum and uranium.

The fuel core alloys were selected on the basis of mechanical property tests, thermal cycling results, and irradiation data from other sites. A reference

alloy of U - 3.5 wt % Mo* was selected, on the basis of the available irradiation stability information.[9] Ternary additions of small amount of silicon or aluminum were found to substantially increase the creep strength of the reference alloy.[10,11] Both of the first two experimental fuel elements irradiated in the OMRE contain U - 3.5% Mo and U- 3.5% Mo - 0.5% Si fuel alloys. Other experimental fuel elements (not discussed in this report) in the OMRE contain U - 3.5% Mo - 0.1% Al alloy fuel.

After a fission product release occurred in the OMRE and temperature and flow instrumentation indicated that the first two experimental fuel elements were malfunctioning, both experimental fuel elements were removed from the reactor for hot cell examination. The objective of the examination and subsequent evaluation was to determine the reason for and nature of the failure of the elements. Although the integrated exposure had been less than that originally planned, the dimensional stability of the alloy fuel and the integrity of the nickel bond were of great interest. The burnup distribution within the fuel plates was felt to be of importance in interpreting the results and in designing future, similar fuel elements.

* % signifies weight % except when stated otherwise.

II. DESIGN AND FABRICATION OF EXPERIMENTAL FUEL ELEMENTS

A. DESIGN

The two experimental fuel elements were of the type originally proposed for use in the Piqua OMR. The design, maximum, fuel-surface temperature for the fuel elements was 750°F. The maximum, design, fuel-center temperature was approximately 770°F.

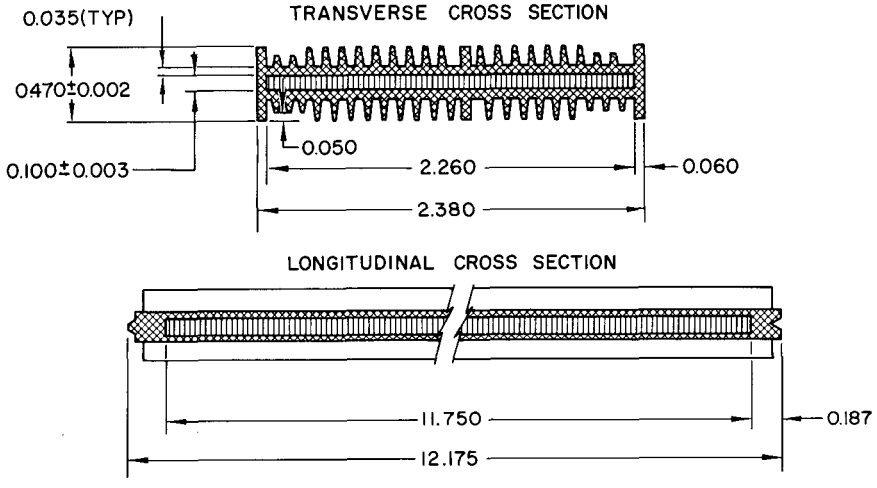
Both fuel elements consisted of cast-plate, uranium alloy, fuel cores with pressure-bonded, finned, aluminum cladding.[8] The plates were stacked, five high, to form a nearly square array, and enclosed in a stainless-steel shroud box. The ends of the box contained standard OMRE fittings, allowing the elements to be substituted for standard fuel elements in the OMRE. Both experimental fuel elements were instrumented to measure cladding temperatures and coolant flow rates, while in operation.

The fuel element that was designed to provide rapid fuel burnup, HB-1, contained a 12-in. center section, consisting of five, 8%-enriched, uranium alloy fuel plates and nonfueled, 1%-boron steel end sections. Three of the fuel plates were U - 3.5% Mo alloy and two were U - 3.5% Mo - 0.5% Si alloy. Figure 1 is a cross section of the 12-in. fueled portion of the element, showing the composition of the fuel plates, the location of thermocouples, and the dimensions of the element.

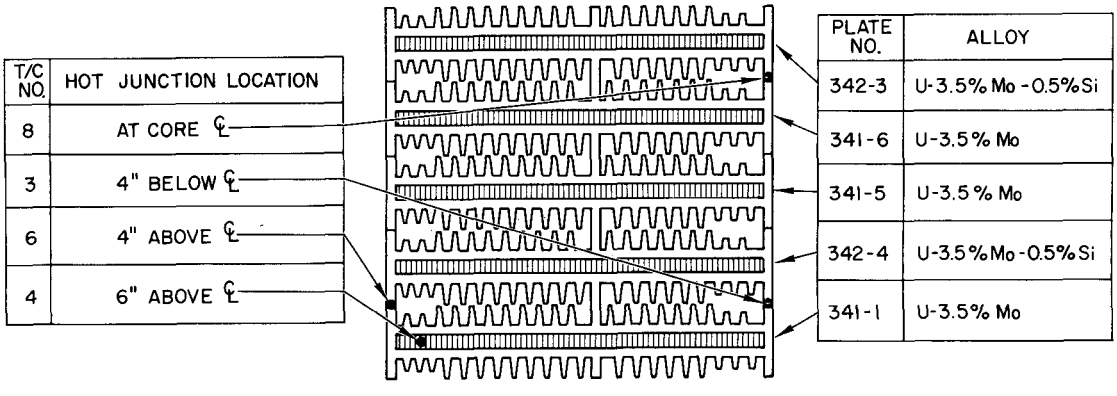
The fuel element that was designed to provide heat transfer as well as fuel burnup information, HT-1, contained three sections, with five, 12-in. long, 4%-enriched, uranium fuel plates per section. The plates in the upper and lower sections contained U - 3.5% Mo alloy fuel. The fuel plates in the center contained U - 3.5% Mo - 0.5% Si alloy fuel. Figure 2 is a cross section of the fueled portion of the fuel element, showing the thermocouple locations and the dimensions of the element.

B. FABRICATION[8]

The core and cladding components used to fabricate a finished fuel plate are shown in Figure 3.

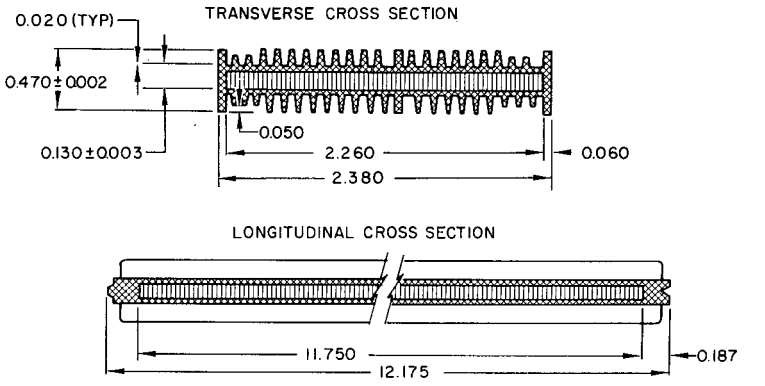


a. Cross Sections of Fuel Plates.

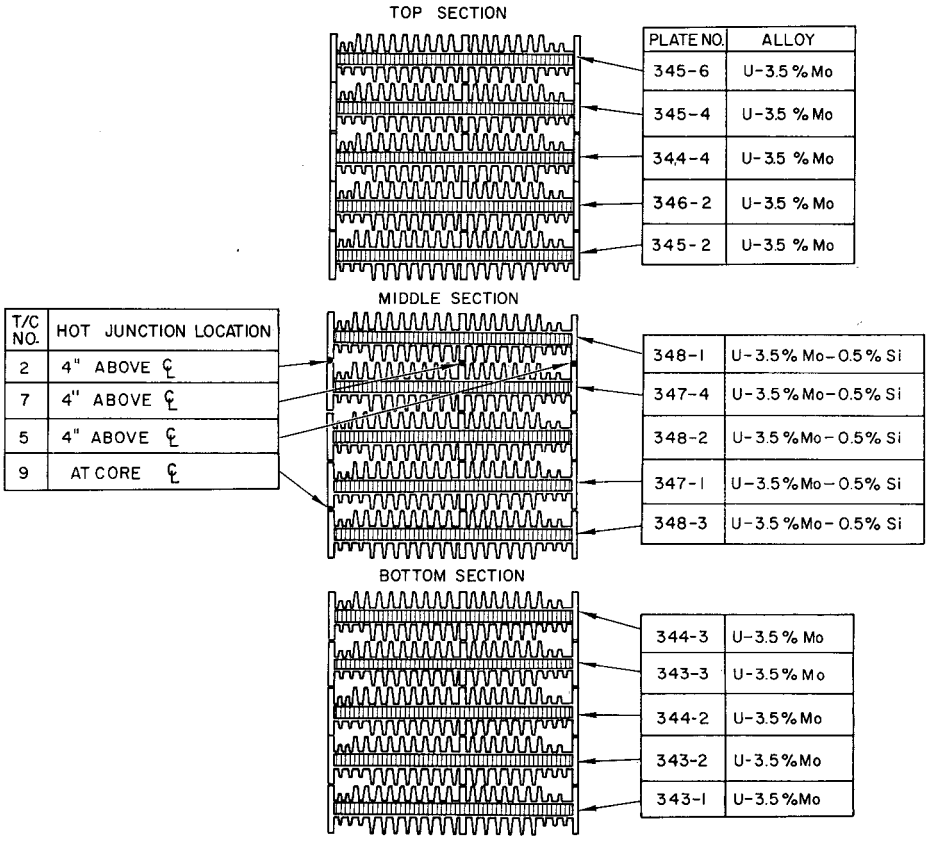


b. Fuel Plate and Thermocouple Location and Designation.

Figure 1. Cross Section of HB-1



a. Cross Sections of Fuel Plates



b. Fuel Plate and Thermocouple Location and Designation

Figure 2. Cross Section of HT-1

The uranium alloy cores for the fuel plates were vacuum induction melted and cast into graphite molds. All plates were cast to the nominal as-cast thickness, 0.130-in., required for the HT-1 fuel element. All the plates were then machined to the length and width specifications shown in Figures 1 and 2. The plates for the HB-1 element were machined to 0.100 ● 0.002-in. thickness.

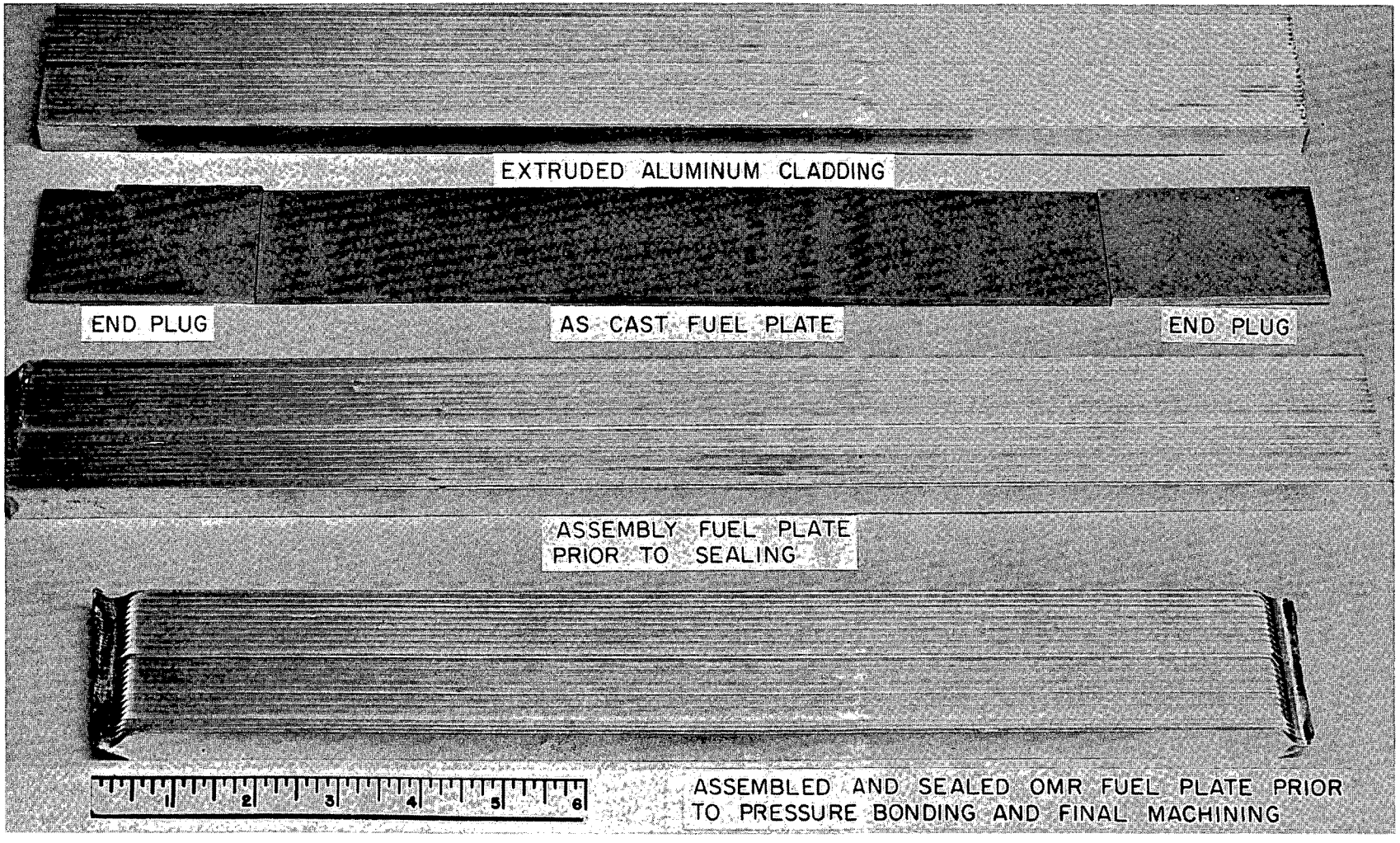


Figure 3. Fuel Plate Assembly Sequence

The aluminum cladding was a flat, extended surface, extruded tube of Harvey Aluminum Company 8030 reactor grade aluminum (1100 F aluminum with controlled impurities). The HB-1 element cladding was fabricated from as-extruded tubing. For the HT-1 fuel element, the inside dimensions of the as-extruded tubing were increased 0.030 in. by Chem-Milling. Sections of cladding were cut to length, and end plugs of 1100 aluminum were machined to fit the inside dimensions of the cladding. A cleaning procedure that included degreasing, an alkaline etch, and nitric acid cleaning was used on the aluminum components.

The uranium alloy core plates were electroplated with 0.0005 in. of nickel and the aluminum components were cleaned at the same time. If the nickel-plated cores were allowed to stand for any length of time after plating, a definite oxide layer would form at the uranium-alloy—nickel interface.

The core and the end plugs were loaded into the cladding. One end plug, fitted with a length of evacuation tube, was Heliarc welded to the cladding to form a leak-tight closure. The other end was then sealed by a "hot knife operation". This operation consisted of pressing a portion of the cladding containing the end plug between two hot punches (800 to 1000°F) until the aluminum between the punches was extruded to a thin section. The excess was cut off with a pair of shears.

The assemblies were then outgassed at 1000°F after evacuating to 5 to 10 μ to assure removal of air, entrapped solutions, and adsorbed hydrogen. Outgassing time varied from 4 hr to overnight. Longer outgassing times resulted in assemblies which were less likely to form cladding blisters after hot pressing. After outgassing, the end of the assembly containing the evacuation tube was sealed by the hot knife technique.

The outgassed and sealed assemblies were bonded by hot isostatic pressing. The pressing was done with either helium or argon at 8000 psi and 1000°F for 10 min. If incomplete bonding occurred, it evidenced itself as visually apparent blisters in the cladding.

The bonded plates were machined to length and inspected. The as-fabricated thickness dimension, as measured across the fins, ranged from 0.465 to 0.478 in. The length, width, and thickness across the flat runner dimensions were within the tolerances shown in Figures 1 and 2. All plates were subjected to a 250°F,

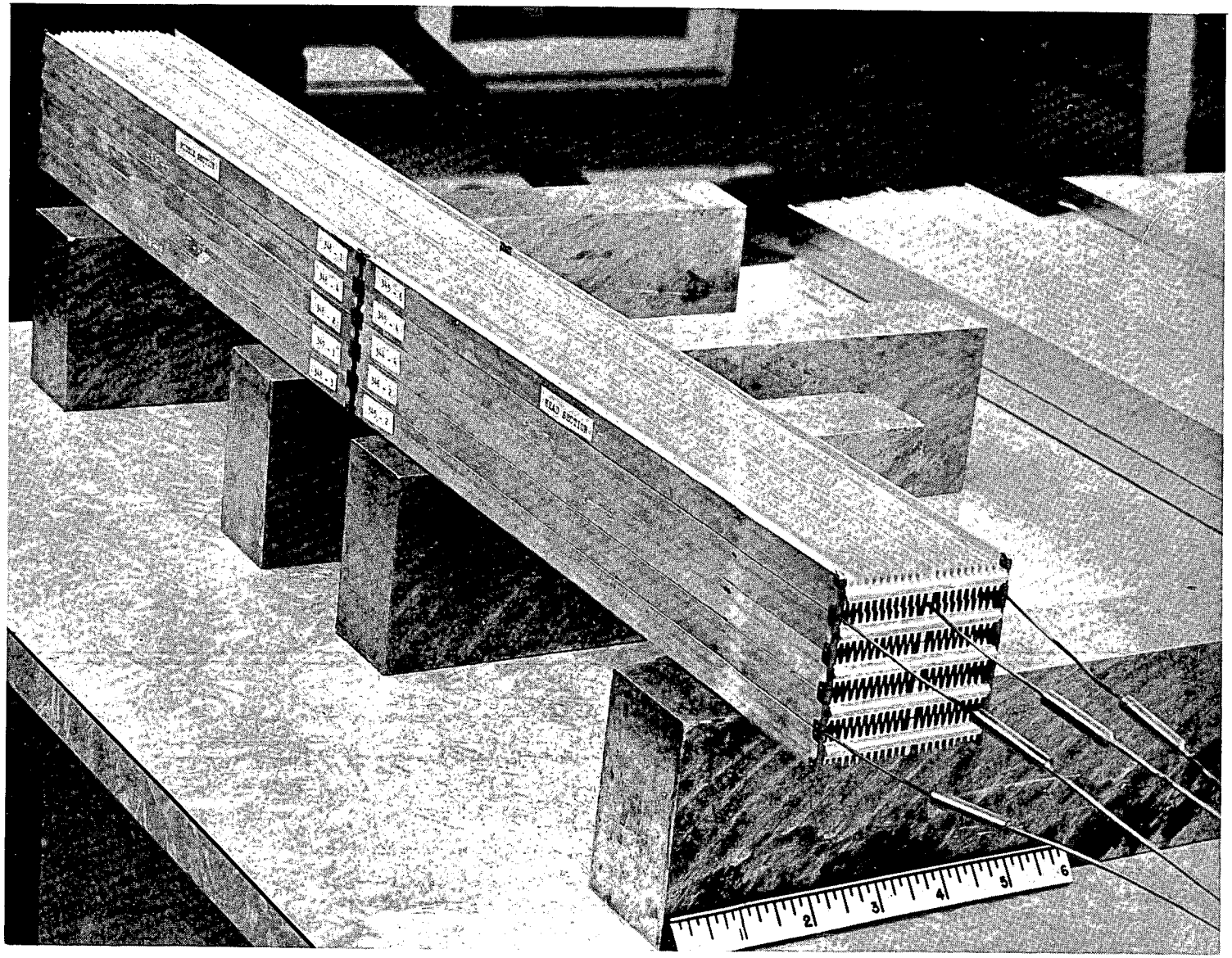


Figure 4. Partially Assembled HT-1 Fuel Element
(Note Thermocouple Leads at Outlet End)

24 hr, 15 psi water autoclave test, prior to final assembly. One plate, 344-2, which had several cladding blisters, was used in the HT-1 element.

Finished plates were assembled into five-plate subassemblies, simply by stacking the plates one on the other and clipping the group together with stainless steel clips. Each experimental fuel element contained four thermocouples to measure the temperature of the aluminum cladding, and one thermocouple to measure the coolant outlet temperature. The stainless steel-clad, Chromel-Alumel thermocouples were positioned between grooves milled into flat portions where two fuel plates rested together. The locations of the thermocouple hot junctions are shown in Figures 1 and 2. The partially assembled HT-1 fuel element, with thermocouples installed, is shown in Figure 4. The subassemblies were mounted inside a 0.050-in., Type 304 stainless steel shroud box. The coolant inlet end of the HT-1 element is shown in Figure 5. End hardware, designed to fit the OMRE grid plates, was welded to the shroud box as the final fabrication step. The bottom end fitting was modified to contain a hot wire anemometer. The anemometer was located in, and measured coolant flow rate through, the inlet end of the fuel element.

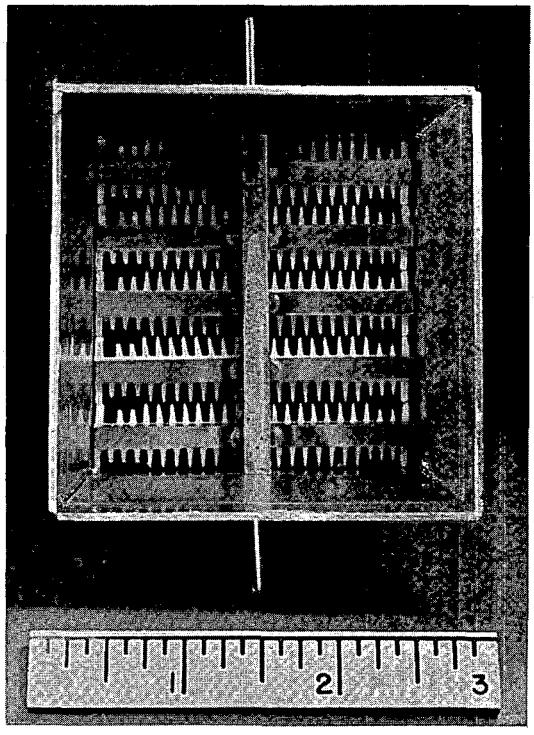
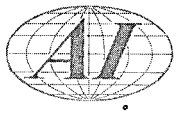


Figure 5. Inlet End of HT-1



III. IRRADIATION HISTORY

A. OMRE CONDITIONS[2]

The experimental fuel elements were irradiated in standard fuel element positions in the OMRE. During the irradiation, the primary reactor fuel consisted of plate type, stainless steel clad, UO_2-stainless steel cermet fuel elements. The evaluation of these OMRE fuel elements has been covered in another report.[12]

The OMRE operates with coolant flow up through the core. The average thermal neutron flux in the core is approximately 2×10^{13} nv. Radiation and high temperatures cause decomposition of the polyphenyl moderator-coolant. This damage results in the formation of multi-ring, high-boiling compounds and also gases, mainly hydrogen, methane, ethane, and smaller amounts of other light hydrocarbons. The high-boiling compounds are detrimental to the heat transfer properties of the coolant, and are removed from the OMRE by batch distillation. The still bottoms, referred to as high boilers (HB), are removed; and the distillate and fresh makeup are returned to the system. By this method, the HB content of the coolant can be maintained at any desired level. The composition of the OMRE coolant has not been constant during operation of the reactor. Variations in coolant composition have been introduced as part of the organic testing programs. The initial coolant composition was 16% diphenyl, 46% orthoterphenyl, 32% metaterphenyl, and 6% paraterphenyl.

B. OPERATIONAL HISTORY

The two experimental fuel elements were loaded in the OMRE during July of 1958. The element (HT-1) designed to provide heat transfer, as well as burnup information, was loaded in Core Position 6. It was planned to leave this element in the OMRE for a period of eight months to one year. The other element (HB-1), designed to provide fuel burnup information more rapidly, was loaded in Core Position 11. It was planned to operate this HB-1 element in the OMRE for a period of four to six months. The arrangement of the OMRE core, after the experimental fuel elements were loaded, is shown in Figure 6.

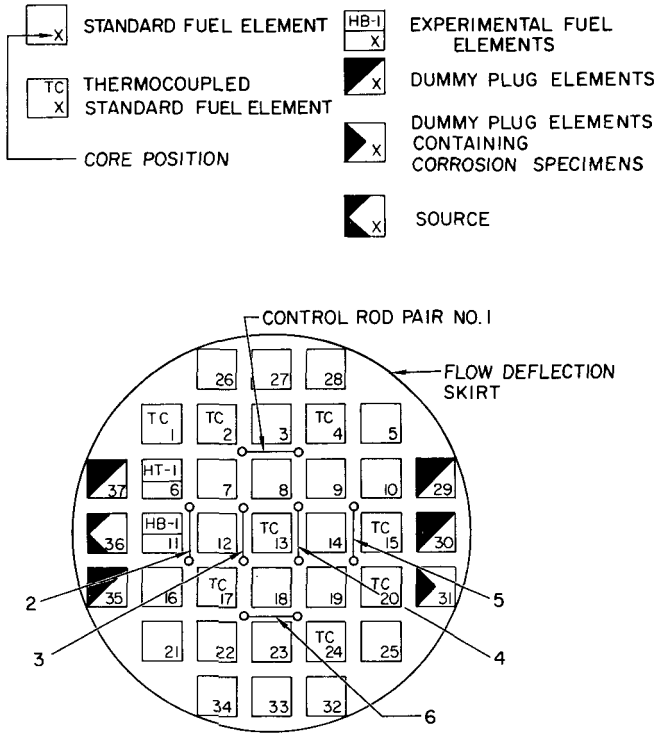


Figure 6. Location of Experimental Fuel Elements in the OMRE Core

(Control Rod Pairs 1, 2, 5, and 6 Normally Withdrawn During Operation)

During the reactor startup after loading the experimental fuel elements, difficulties were experienced with one of the standard OMRE fuel elements. Standard Fuel Element 30, which was moved to Core Position 1 during the shutdown, had failed to latch in the reactor gridplate. Subsequent partial loss of coolant flow caused the fuel element to overheat. After 8 Mwd of reactor operation, the element was latched, and the thermocouple readings on the element returned to normal. When the first core loading of OMRE standard fuel element fuel elements was removed for reprocessing, Element 30 was found to be badly distorted, due to massive carbonaceous deposits which deformed the fuel element.[12]

The conditions in the OMRE core during the irradiation of the two experimental fuel elements are represented in Figure 7. From July 24 to September 18, the reactor accumulated 235 Mwd at 600°F inlet temperature and a power level of 5.5 to 6.0 Mwt. HB content varied from 30 to 40% during the run, with most of the operation at 40%. Higher coolant activity than previously experienced,

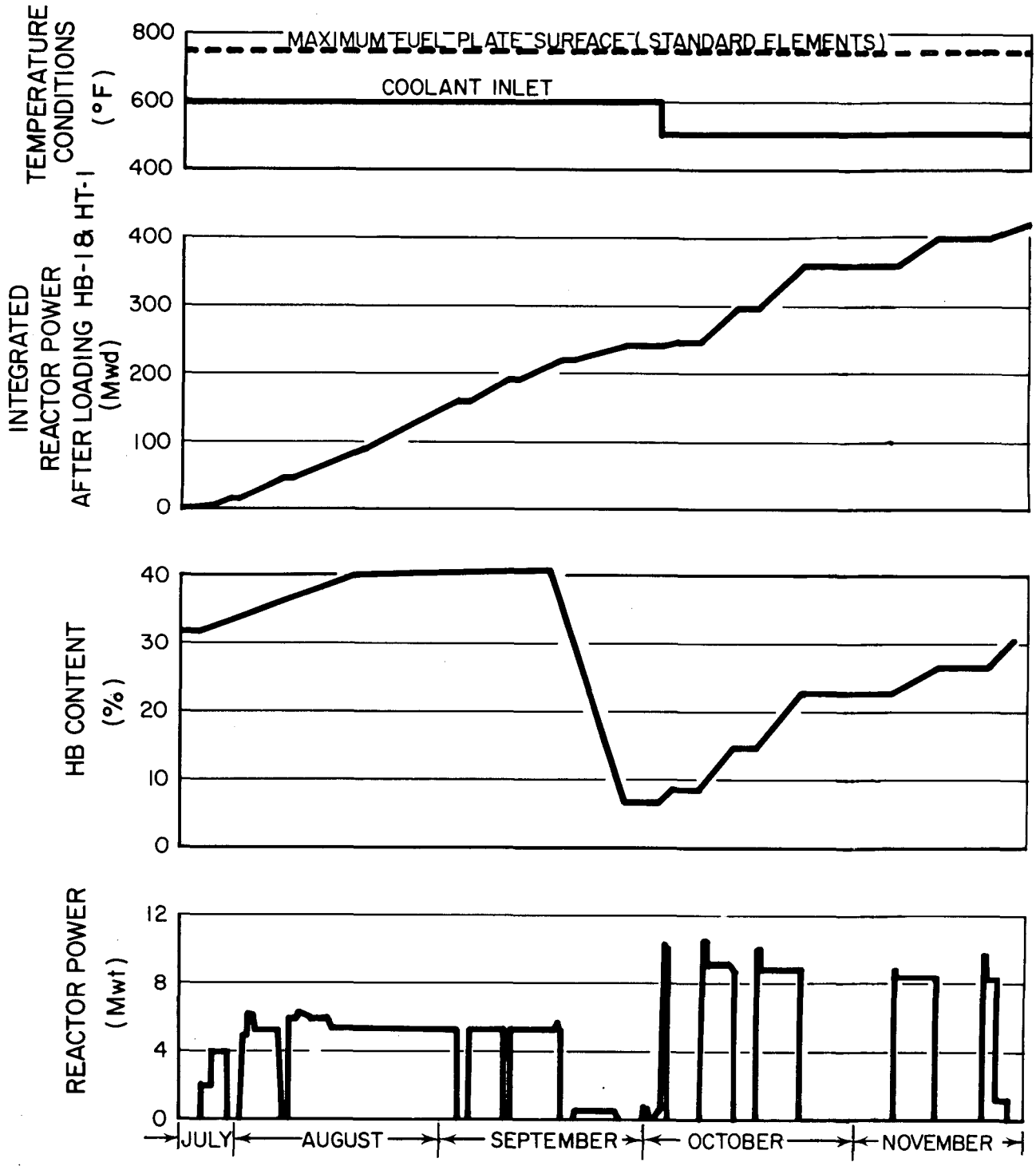


Figure 7. OMRE Operating Conditions During July through November 1958

principally due to Mn^{56}, was noted during this period. The Mn was attributed to residual rust in the system and rust from the low carbon steel drums used for shipping the organic coolant. Since the high activity impurities in the coolant stay behind with the still bottoms during removal of HB from the coolant, the HB content of the coolant was lowered from 40 to 8%. From September 18 to October 1, the OMRE was either shut down or at a low power, during reduction of the HB content of the coolant. On October 1, a run with 10 to 8.5 Mw power level at 500°F coolant inlet temperature was initiated. The purification system was shut down, and the HB content was permitted to increase, with the intention of leveling off at 30%. A sample of coolant, taken on October 10, contained a very small amount of fission product activity. At 10:43 A. M. on October 23, after 346 Mwd reactor operation with the experimental fuel elements, a significant quantity of fission products was released to the OMRE coolant. The reactor was scrammed immediately and the off-gas system sealed.

The temperature and flow conditions recorded for the HB-1 element are shown in Figure 8. The two thermocouple readings plotted, Number 6 and Number 8, recorded 40 and 15°F increases respectively, for a 2-min period prior to the coolant activity increase. The remainder of the thermocouples on HB-1, and the thermocouples on HT-1, read temperatures less than those shown for Thermocouple Number 6. The location of the thermocouples in the fuel elements is shown in Figures 1 and 2. Fuel temperatures had been gradually increasing during the irradiation. However, the maximum reading thermocouple, Number 8, was only slightly higher than the design maximum surface temperature of 750°F. Table I compares the thermocouple readings from the fuel elements at two dates during the power run at 10.5 to 8.5 Mw. The temperatures increased, even though the power level had decreased, with the exception of Thermocouple Number 8, which had shown an increase earlier in the irradiation.

There is considerable doubt about the accuracy of the anemometer readings. The anemometer had been calibrated out of pile and, based on measured flow vs pressure drop characteristics, the flow through the HB-1 element should have been approximately 110 gpm. As shown in Figure 8, the anemometer never indicated more than 60 gpm. Over most of the exposure of the HB-1 element, the readings were between 20 and 40 gpm. The tendency was toward decreasing flow readings with continued exposure.

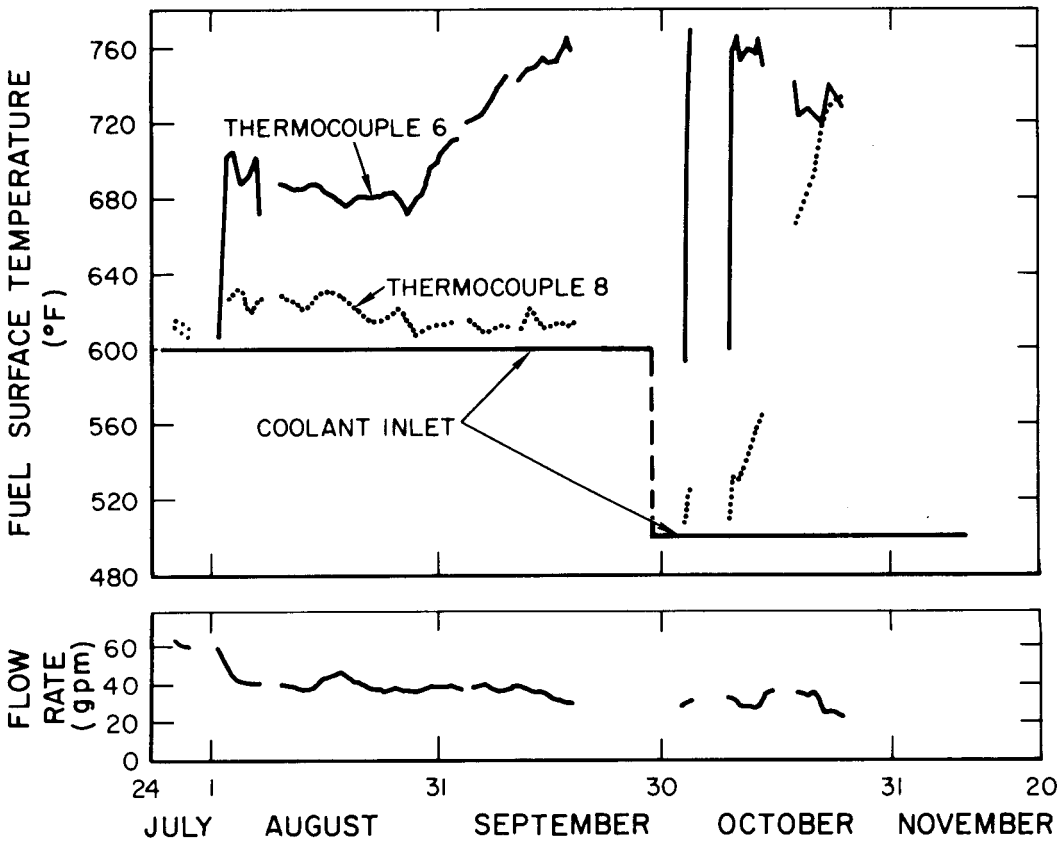


Figure 8. Temperature and Flow Conditions in HB-1

TABLE I

INCREASE IN EXPERIMENTAL FUEL CLADDING TEMPERATURES DURING IRRADIATION

Fuel Element	Thermo-couple No.	Hot Junction Location (inches from core centerline)	Temperature (°F)	
			Oct. 3, 10.5 Mw	Oct. 13, 8.5 Mw
HB-1	3	4 below	547	556
HB-1	4	6 above	590	618
HB-1	6	4 above	760	760
HB-1	8	centerline	522	566
HT-1	2	at centerline	582	588
HT-1	5	4 above	not functioning	
HT-1	7	4 above	600	630
HT-1	9	at centerline	618	679

Based on the temperature and flow information, it seemed likely that the HB-1 element was the source of the fission products. However, due to the detected release of only gaseous and highly volatile fission products, it was not certain what type of failure had occurred. Therefore, it was decided to operate the reactor again, at low power, and observe the results. On October 29, the reactor was run at 350 kw for 3 hr with the coolant at 500°F. The activity, 2 in. from a 10-in. coolant line, increased from 140 to 240 mr/hr. On October 30, the reactor was operated at 1.6 Mw for 6 hr at 500°F. The radiation level increased to an equilibrium value of approximately 400 mr/hr. The temperature indications from HB-1 were significantly higher than expected from previous experience. As operation continued, the temperatures continued to increase and showed pronounced fluctuations. Figure 9 shows the temperatures from HB-1 during the 6-hr run. The temperatures scattered more than is shown by the relatively few points plotted; for example, Thermocouple 8 increased from 615 to 645°F between 21:15 and 21:32, and then dropped down to 600°F within 1 min. During this run none of the temperatures in HT-1 exceeded 550°F. Based upon the above data, it was decided to remove HB-1 for hot cell examination.

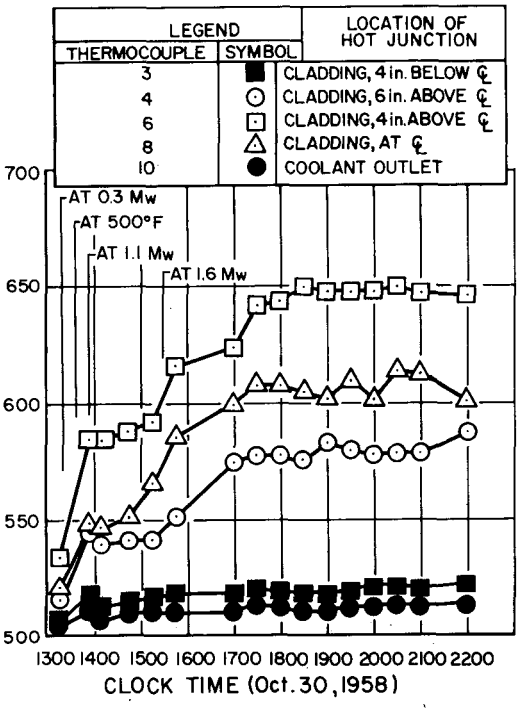


Figure 9. Temperatures in HB-1 After the Fission Product Release

The decay in radiation level, 2 in. from the 10-in. coolant line, was plotted after the October 23 fission product release. Extrapolating back to the time of the fission break resulted in levels between 50 and 70 R/hr, which imply a release of several hundred to one thousand curies of activity. Gamma spectral analysis of the coolant indicated the presence of isotopes of xenon, iodine, barium, lanthanum, tellurium, krypton, and rubidium. A special analysis, to detect the presence of zirconium and niobium, was negative.

Based on coolant samples and blanket gas samples, the amount of activity released at the time of the fission break was calculated to be that shown in Table II. Most of the iodine released was retained in the organic coolant. Analysis of the OMRE coolant showed a uranium concentration of 1.7×10^{-8} gram of uranium per gram of coolant, or approximately 350 mg uranium in the entire organic system.

TABLE II

CALCULATED ACTIVITY OF SPECIFIC ISOTOPES RELEASED, BASED ON COOLANT AND BLANKET GAS SAMPLES

Isotope	Curies
I^{131}	27.4
I^{133}	70.4
Ba^{140} - La^{140}	2.1
Xe^{133}	37.9
Xe^{135}	48.6

Power operation was initiated again on November 8. Temperature readings on the HT-1 element continually increased. When a maximum temperature of 781°F was reached with Thermocouple Number 9, the reactor power was decreased from 8.5 to 5.9 Mw. The reactor was shut down on November 13, 1958, to remove the HT-1 element. Reactor operation with the HT-1 element totaled 384 Mwd. During the run, some fresh fission product activity was detected, but no massive releases occurred.

IV. POSTIRRADIATION PROCEDURES AND RESULTS

Hot cell examinations of the two experimental fuel elements were conducted to determine the reason for the failure, as evidenced by the fission product release in the OMRE, and to determine the extent of damage to the elements. Although the exposure time had been less than that originally planned, the dimensional stability of the alloy fuel and the integrity of the nickel bond were of great interest. The burnup distribution within the fuel plates was felt to be of importance in interpreting the results and in designing similar fuel elements.

A. SHIPPING OF FUEL ELEMENTS

The experimental fuel elements were shipped for hot cell examination in special shipping casks, designed to handle OMRE fuel elements. These casks contained isopropyl diphenyl as a heat transfer medium between the walls of the cask and the fuel element. A circulating water system was provided to cool the walls of the cask. Both experimental fuel elements were bent at one end during loading into the shipping casks. Insufficient space for the cut-off thermocouple lead tube caused the fuel elements to cock. The misaligned fuel elements then bent when the cask lid was closed. The shipping casks have been reworked to prevent this type of damage from occuring in the future.

B. DIASSEMBLY AND VISUAL OBSERVATIONS

1. High Burnup Fuel Element, HB-1

Initial viewing of the outside of the fuel element revealed no damage other than the bend near the inlet end, which had occurred during closing of the shipping cask. The entire element was covered with a glossy, dark coating of residue. The residue had a reddish appearance and was somewhat viscous. Over several hours, the coating appeared to dry and harden, particularly over the fueled portion of the element. By use of Tempilsticks (crayon-type materials of known melting points) the temperature of the shroud box over the fueled portion was determined to be between 125 and 150°F.

The experimental fuel elements were disassembled, using a milling machine with a vertical head. A holding fixture was bolted to the mill table to position the element. This fixture supported the entire length of the element, and

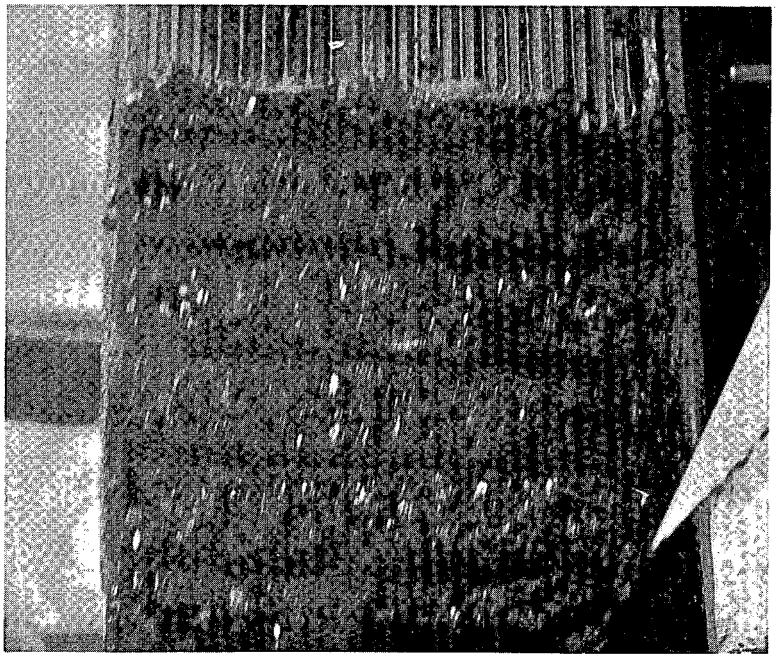


Figure 10. Inlet End of HB-1

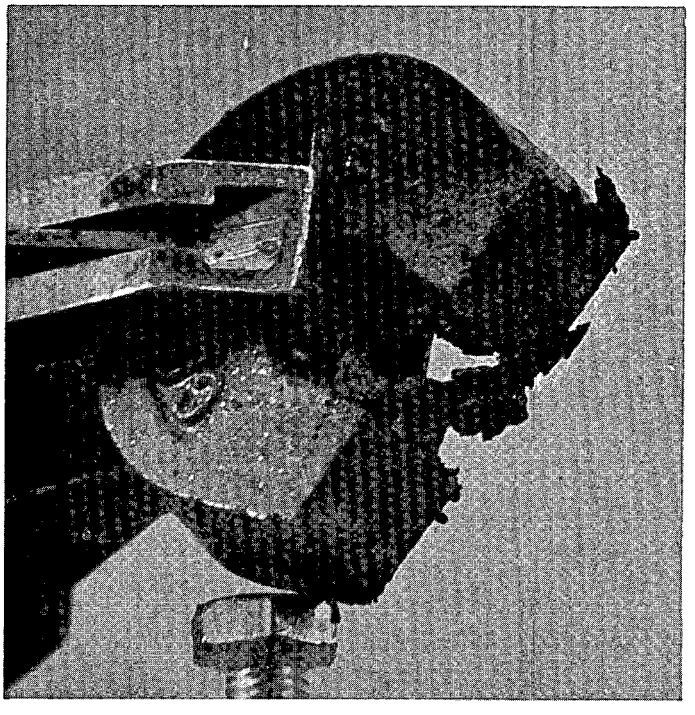
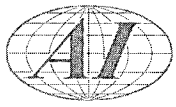


Figure 11. Magnetic Material Blocking Inlet of HB-1



was adjustable to compensate for the limited mill table travel. The shroud box was milled open longitudinally, along two diagonally opposite corners. The two cuts were joined at each end of the box by transverse cuts, and the angle-shaped piece of box was removed. The flat edges of the fuel plates, and the edges of the boron steel plates of the HB-1 element, were seen to be covered with a hard, flaky deposit. The deposit could be flaked off and pulverized. The faces of the boron steel plates had only a light coating of residue. The exposed, finned, cladding surfaces of the outer fuel plates were partly coated with a heavy residue. The fuel plate surfaces were examined in greater detail after disassembly.

The boron steel plates at the inlet end of the assembly were removed as a unit. The coolant channels through this portion of the assembly were free from any heavy deposits. The inlet end of the fueled portion, which was largely blocked by a hard granular deposit, is shown in Figure 10. The deposit had the appearance of chunks of material that had been filtered out by the aluminum fins of the extended surface cladding. The material was magnetic, to the extent that small pieces jumped approximately 1/2 in. to a magnet. Figure 11 shows a sample of the deposit adhering to a magnet. At this point in disassembly, the stacked fuel plates showed no gross distortion.

The boron steel plates on the outlet end of the assembly were removed, one at a time. A large metallic deposit was noted between the third and fourth steel plates. The deposit was attached to Fuel Plate 341-6, as shown in Figure 12. The metal was relatively soft when scratched, and had the appearance of a solidified, large, molten drop. Smaller metallic pieces were noted between the fourth and fifth steel plates. The deposits were not attached to the steel plates. Figure 13 shows the outlet end of the fueled section. With the exception of the globs of metal, the coolant channels were relatively free from blockage.

The five fuel plates were separated and examined individually. It was apparent that partial melting of the cladding had occured on four of the five plates. In addition to, and apparently associated with, the melted areas, some areas of the cladding were coated with a dark deposit. In some areas, the deposit completely filled the space between adjacent fins. Figure 14 shows typical fuel plate surfaces. The side of the plates where most of the melting occurred was facing Core Position 16. The majority of the melting occurred between the 5th and 4th,

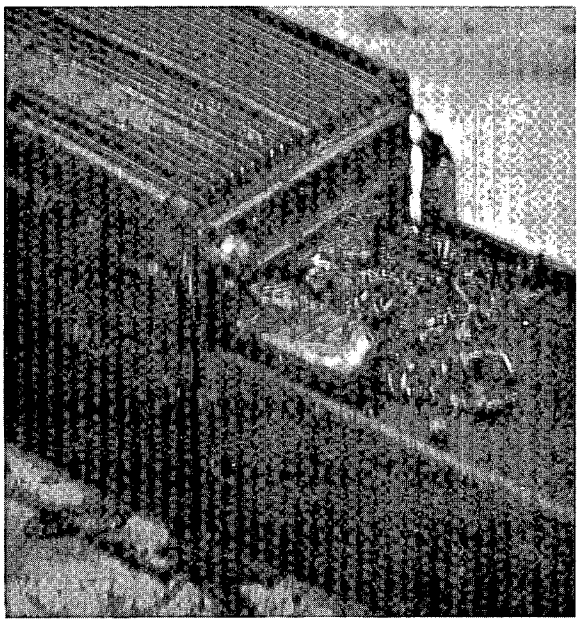


Figure 12. Metallic Deposit at Outlet End of HB-1

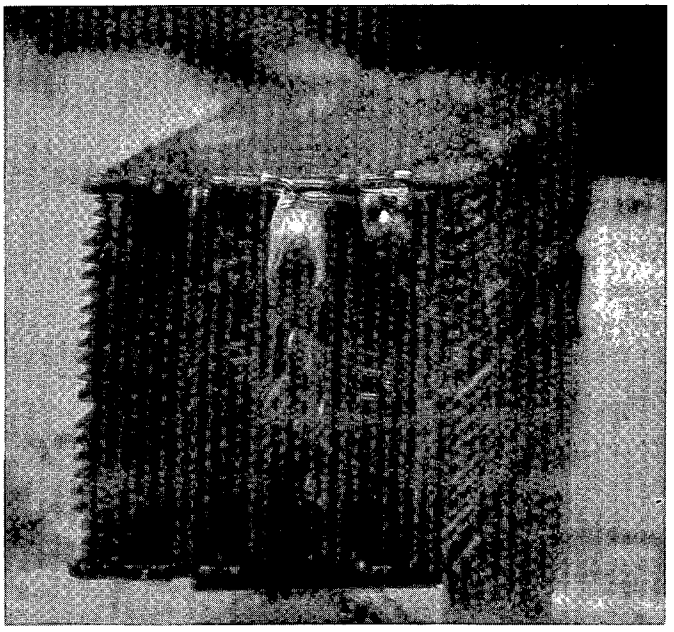
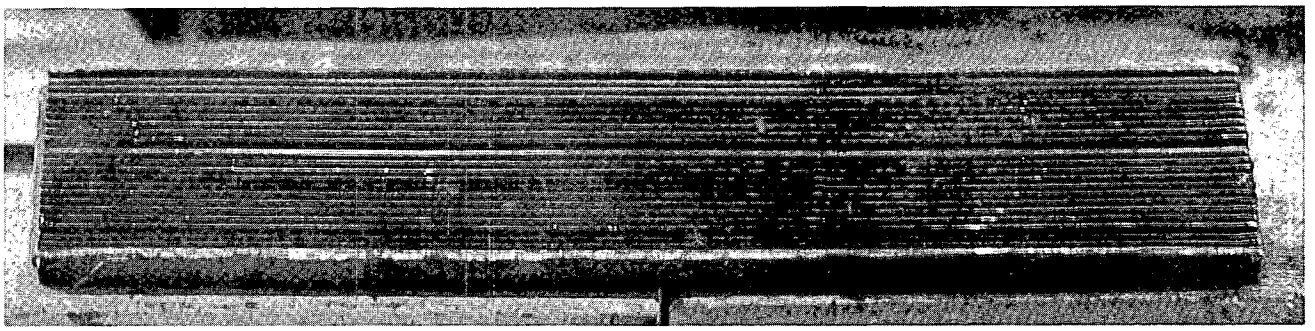
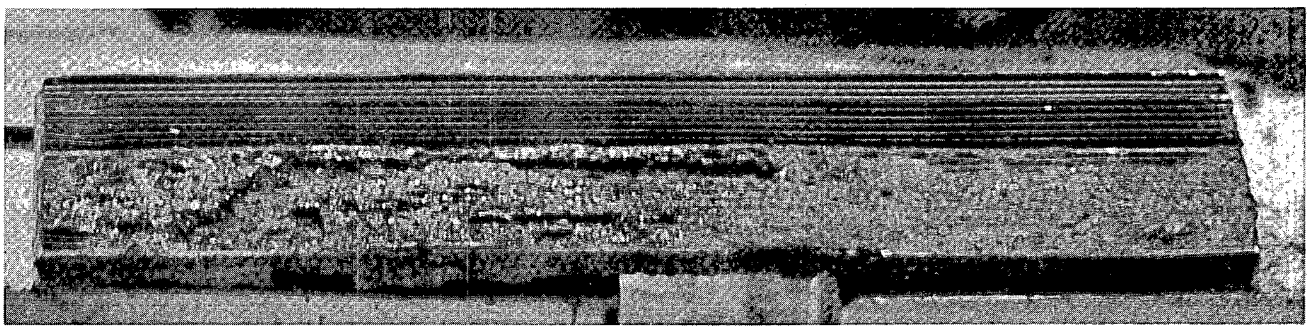


Figure 13. Outlet End of HB-1
(Note Metallic Deposit)



a. Plate 341-1 — Undamaged Plate



b. Plate 341-5 — Plate With Partially Melted Cladding, Typical of 4 of the 5 Plates in HB-1

Figure 14. Fuel Plates from HB-1

the 4th and 3rd, and the 3rd and 2nd fuel plates, as numbered from the side facing the center of the OMRE core. The fuel plate from the side of the element closest to the center of the core appeared undamaged, and had only scattered deposits. The thermocouples located closest to melted areas were Thermocouples Number 6 and 8, but neither of these actually impinged on an area where melting had occurred. It was visually estimated that about 40 $in.^2$ of cladding were at least partially melted.

2. Heat Transfer Element, HT-1

The HT-1 fuel element was examined and disassembled in the same manner as the HB-1 fuel element. With the exception of the damage incurred while loading the fuel element into the shipping cask, the outside of the fuel element appeared in good condition. The outside of the fuel box was coated with a dark, glossy film. As with the HB-1 fuel element, the film dried to a harder deposit, in a few hours, over the fueled portion of the fuel element. The temperature of

the outside of the fuel element was determined, by the use of Tempilsticks, to be less than 125°F.

The shroud box was milled open, as with the HB-1 element. No gross distortion of the three subassemblies of fuel plates was noted. The flat sides of the fuel plates, perpendicular to the extended surface portion, were coated with a dark, flaky, hard deposit, similar in texture to the deposits in the same location on the HB-1 fuel element. The inlet end of the lower fuel section, shown in Figure 15, was largely blocked by a hard, granular deposit. The outlet end of the fuel element, and the interfaces between the three fueled sections, had only occasional deposits.

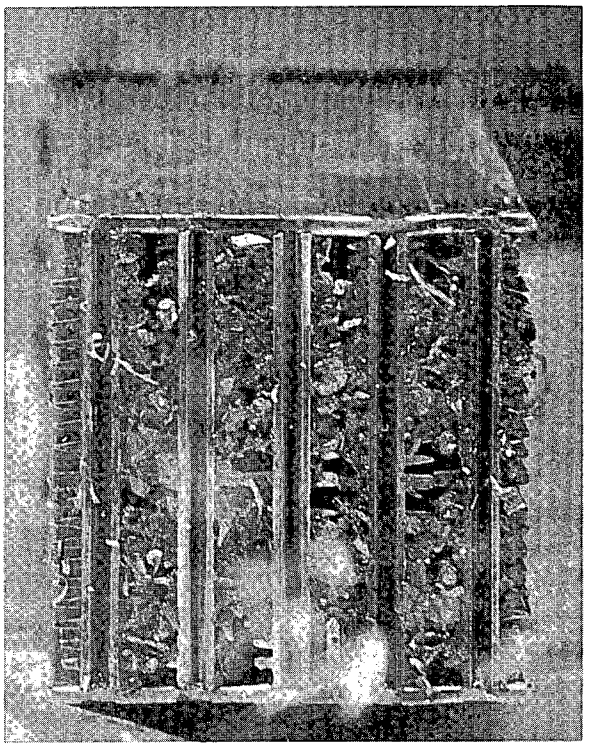
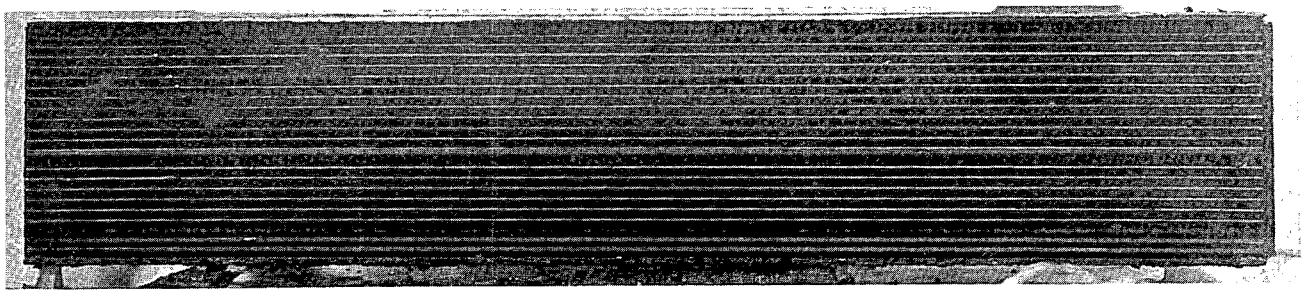
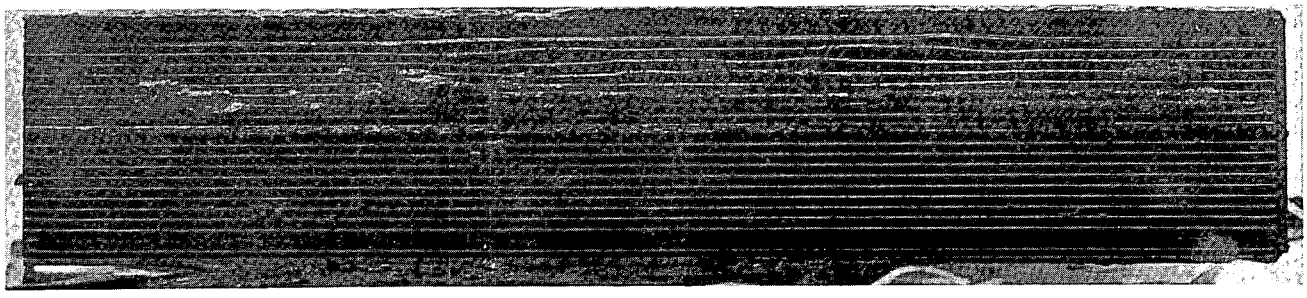


Figure 15. Inlet End of HT-1

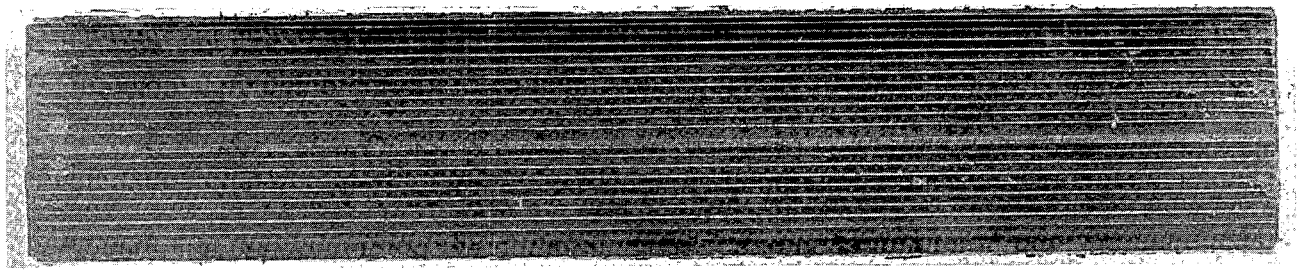
The fuel plates were separated and examined individually. No signs of cladding melting were observed. Occasional, heavy, dark deposits were found on the cladding, which filled the space between adjoining fins. One plate was found to have a cladding blister, about 6 in. long by 1 in. wide, on both sides of the plate. This blistered plate, Number 347-1, and other typical plates are shown in Figure 16. Plate 347-1 was from the center section of the fuel element,



a. Plate 348-3 — Plate From Center Section of HT-1, Closest Plate to the Center of the Core



b. Plate 347-1 — Blistered Plate From Center Section of HT-1, Adjacent to Plate 348-3 (The blister is evidenced by the wavy fins in the top half of the photo)



c. Plate 344-1 — Plate From Upper Section of HT-1

Figure 16. Fuel Plates From HT-1

and was the second plate from the edge of the element closest to the center of the reactor core. The center plate from the center section evidenced some distortion of the cladding fins in the same location as the blister on adjacent Plate 347-1, but the area had not raised, as on Plate 347-1. The only other deformed areas noted were small blisters on the center plate from the lower fueled section. These blisters had been noted during fabrication, and appeared to be the same size as before irradiation. The deposits on the finned cladding surfaces were most pronounced on fuel plates from the lower and center sections. The five plates from the upper section were noticeably more free from deposits. The deposits on the finned surfaces were relatively soft and could be readily removed. Part of some of the deposits had a matted, fibrous appearance.

C. DIMENSIONAL MEASUREMENTS

Dimensional measurements were taken on selected fuel plates, in order to determine the dimensional stability of the core material. It was assumed that any change in dimensions of the core would be reflected as changes in dimensions of the cladding. The undamaged U - 3.5% Mo plate from the HB-1 element, Plate Number 341-1, the blistered U - 3.5% Mo - 0.5% Si plate from the center section of the HB-1 element, Plate Number 347-1, and an undamaged U - 3.5% Mo - 0.5% Si plate next to 347-1 in the center section of the HT-1 element, Plate Number 348-3, were selected for detailed dimensional analysis. The thickness and width measurements were taken with flat anvil micrometers. The length measurements were taken with gauge block and dial indicator. Figure 17 shows the locations measured. Before measuring, all superficial deposits were wiped from the plate surfaces. A dark, very adherent coating remained on the plates. Light probing with a sharp tool, which exposed bright aluminum underneath, indicated that the coating was of negligible thickness, except at the plate ends. The ends were carefully scraped before measuring. Table III gives the results of the measurements on the three plates. Table IV compares the average dimensions found with the preirradiation measurements. The thickness across the flat runners was used in determining the average postirradiation thickness. The thickness across the fins was not averaged, due

TABLE III

POSTIRRADIATION FUEL PLATE MEASUREMENTS
(Measurements in inches)

Dimension	Position*	Plate 341-1†	Plate 348-3§	Plate 347-1**
Thickness on Flats	1	0.473	0.473	††
	2	0.460	0.482	††
	3	0.467	0.474	††
	4	††	††	0.474
	5	0.472	0.467	0.485
	6	††	††	0.498
	7	0.472	0.476	††
	8	0.468	0.476	††
	9	0.468	0.470	††
Thickness on Fins	10	0.461	0.480	0.493§§
	11	0.465	0.483	0.554§§
	12	0.461	0.482	0.588§§
	13	0.465	0.475	0.546§§
	14	0.460	0.480	0.494
	15	0.465	0.478	††
	16	0.459	0.481	††
	17	0.468	0.479	0.485
	18	0.460	0.479	††
	19	0.460	0.476	††
Width	a	2.382	2.386	††
	b	2.383	2.388	††
	c	2.380	2.383	††
Length	I	12.259	12.247	12.243
	II	12.248	12.262	12.248
	III	12.229	12.266	12.245

* Location of measurements shown in Figure 17
† Plate 341–1 was a U – 3.5% Mo plate from HB–1
§ Plate 348–3 was a U – 3.5% Mo – 0.5% Si plate from HT–1
** Plate 347–1 was the blistered U–3.5% Mo – 0.5% Si plate from HT–1
†† Measurement not taken
§§ Region of visible blister

to the spread in preirradiation measurements. The only significant changes were small increases in plate lengths for all three plates and increases in thickness on the blistered area of Plate 347-1.

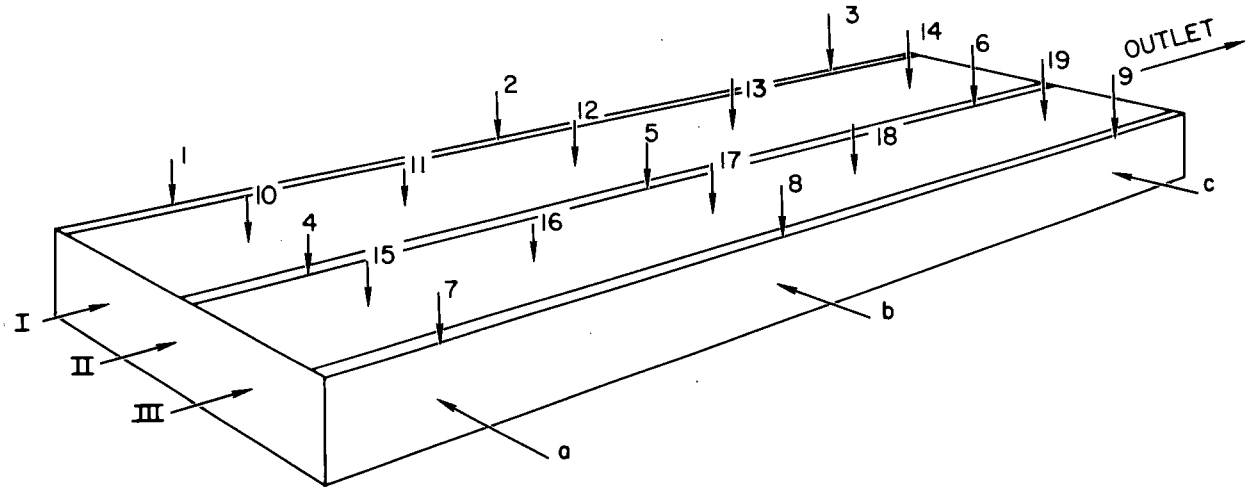


Figure 17. Location of Plate Measurements

TABLE IV

CHANGE IN DIMENSIONS OF FUEL PLATES DURING IRRADIATION
(Measurements in inches)

	Thickness*	Width	Length
Original Dimensions			
Average	0.470	2.380	12.175
Fabrication Tolerance	±0.001	±0.001	±0.001
Plate 341-1			
Average	0.469	2.382	12.245
Average Change	-0.001	+0.002	+0.070
Plate 348-3			
Average	0.474	2.386	12.259
Average Change	+0.004	+0.006	+0.084
Plate 347-1			
Average	†	§	12.245
Average Change			+0.070

* Flat areas only
† Not averaged due to blister
§ Not taken

D. CHEMICAL ANALYSES

It was desirable to know the composition of the material blocking the inlet ends of the fuel plates, and the metallic-appearing material that apparently flowed out of the outlet end of the HB-1 fuel element. Unfortunately, the radioactivity of the samples, which ran as high as 10 R/hr at the surface of the sample bottles, limited the size of the samples and the number of analyses. The samples analyzed were from the HB-1 element. Samples from the inlet end of the fuel element had much lower activity than the samples from the other areas.

The samples were extracted with solvents for polyphenyls, and the extracted portions analyzed by gas chromatography and ultraviolet spectroscopy. The analyses of the extracted portions, which amounted to 17 to 20% of the material blocking the inlet end of the element and 3.9% of the material from the outlet end, are given in Table V, along with the composition of the OMRE coolant at the time the HB-1 fuel element was removed from the reactor. Even if all of the dissolved material not accounted for is assumed to be HB, the amount present is quite small. It is apparent that the major soluble organic portion of the deposits is isopropyl diphenyl from the shipping cask.

The following additional information was obtained concerning the material blocking the inlet end of the fuel element: Fe^{59} and Cr^{51} accounted for the major amount of the identifiable activity present. Iron was identified as the major metallic constituent present, by an X-ray fluorescence determination, chromium and nickel were identified in lesser amounts. Firing a sample resulted in 16% ash. X-ray diffraction showed no identifiable pattern.

After extraction, the sample of residue, found between the boron steel plates at the outlet of the HT-1 fuel element, appeared as a shiny, metallic-appearing chunk and some fine black powder. The shiny, metallic chunk was analyzed chemically as 92.1% aluminum, 7.3% uranium, and 1.0% iron. The powder was analyzed chemically as 87.0% aluminum, 7.3% uranium, and 1.0% iron. A large number of metallic fission products were identified by radiochemical analysis in these metallic samples.

E. BURNUP ANALYSES

Three fuel plates were sent to Battelle Memorial Institute for burnup analysis and metallographic examination.[13] The three plates previously measured to

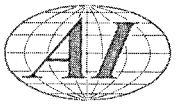
TABLE V

COMPOSITION OF SOLUBLE ORGANIC FRACTION OF SAMPLES FROM HB-1

Sample No.	Location	Solvent	Extracted (%)	Organic Analysis of Extracted Portion (%)				
				Isopropyl Diphenyl	Ortho-terphenyl	Meta-terphenyl	Para-terphenyl	Unaccounted For
1	Inlet End	methylene chloride	20	greater than 90% unpolymerized polyphenyls				
11	Inlet End	tetrahydrofuran	17	71	6.5	4	1	17.5
3	Outlet End	tetrahydrofuran	3.9	84	8	-	-	8

Analysis of OMRE Coolant Sample Taken 2 Days After HB-1 was Removed

	%
Diphenyl	6.9
Orthoterphenyl	31.8
Metaterphenyl	24.4
Paraterphenyl	3.3
HB (by distillation)	24.0
Unaccounted for (by difference)	9.6



determine dimensional changes due to irradiation were selected. The three plates were: (1) Number 341-1 from HB-1, U - 3.5% Mo, an undamaged plate from the side of the element closest to the center of the OMRE core; (2) Number 348-3 from HT-1, U - 3.5% Mo - 0.5% Si, an undamaged plate from the center section of the element and from the side of the element closest to the center of the OMRE core; (3) Number 347-1 from HT-1, U - 3.5% Mo - 0.5% Si, a blistered plate adjacent to Plate 348-3.

Profiles of the relative intensity of gamma radiation emitted from the three plates were used to establish burnup profiles. For this measurement, the fuel plates were moved past a 1/8-in. diameter hole through the hot cell wall. The radiation coming through the hole was detected with a sodium iodide scintillation crystal. The pulses from the crystal were recorded with a pulse height analyzer. Only gamma energies between 700 and 750 kev were recorded. On each of the three plates, three longitudinal and four transverse scans were made. Two 1/4-in.-square samples were cut from the longitudinal centerline of each plate. The six samples were dissolved and analyzed for cesium-137 and uranium content to determine the atom percent uranium burnup. By plotting the gamma intensity, recorded at the sampled areas before sectioning, against analyzed atom percent uranium burnup, a correlation was established between the relative gamma intensity scans and the burnup. The correlations are shown in Figures 18 and 19. Separate curves were used for the two samples from the HB-1 plate and the four samples from the HT-1 plates because of the differences in core thickness between the two elements. By relabeling the gamma scans in terms of burnup, it was possible to plot burnup contours for the fuel plates. The contours for the three plates are shown in Figure 20.

The average burnup for the three plates was calculated by determining the area in the profile plots between increments of burnup, multiplying the area by the average burnup in the area, summing the results over the total area of the profile, and dividing the sum by the total area of the profile. The average burnups obtained by this method, and the maximum burnup from the scans obtained for each plate, are shown in Table VI.

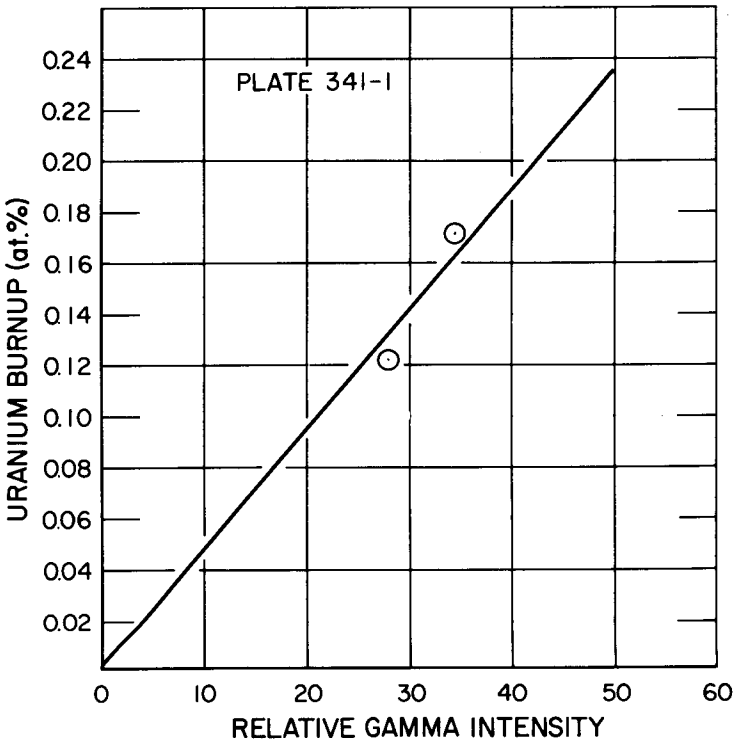


Figure 18. Correlation of Radiochemical Burnup with Gamma Intensity for HB-1

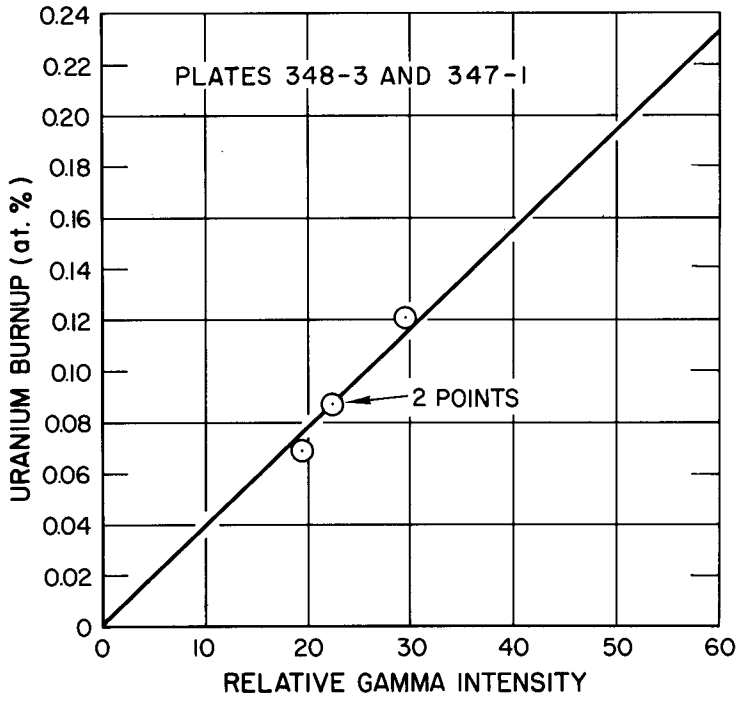
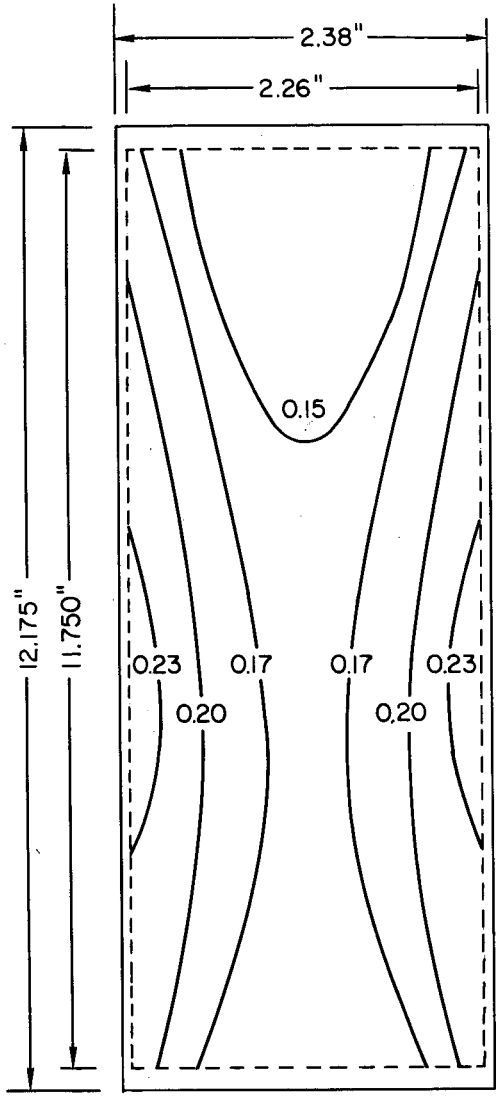
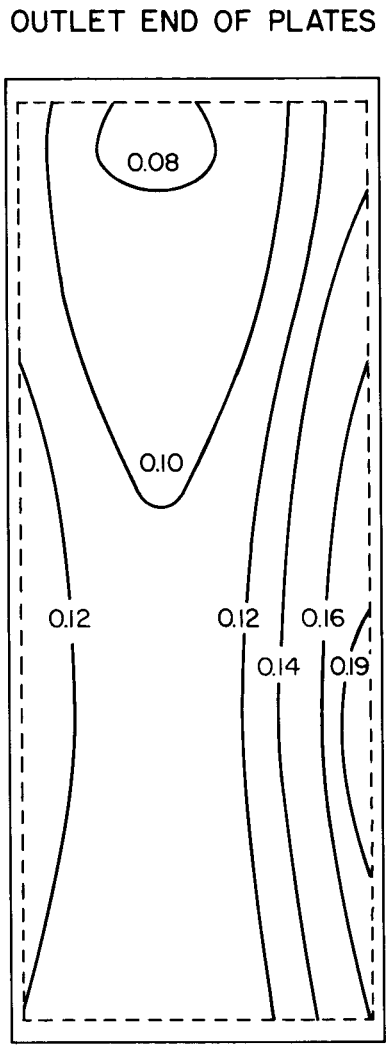


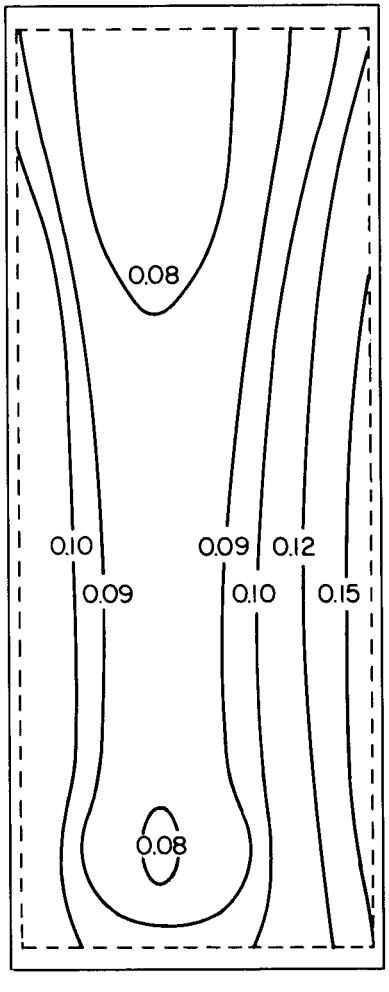
Figure 19. Correlation of Radiochemical Burnup with Gamma Intensity for HT-1



a. Plate 341-1, U – 3.5% Mo, from HB-1 – Plate Closest to Center of Reactor Core

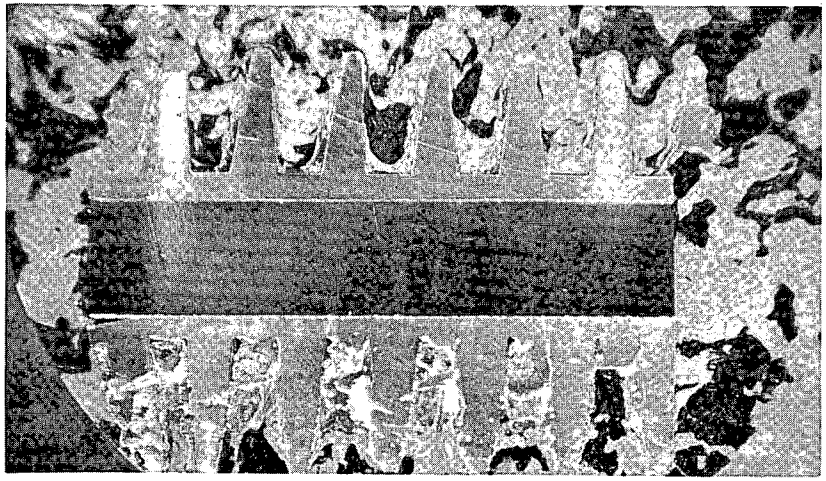


b. Plate 348-3, U – 3.5% Mo – 0.5% Si, from Center Section of HT-1 – Plate Closest to Center of Reactor Core



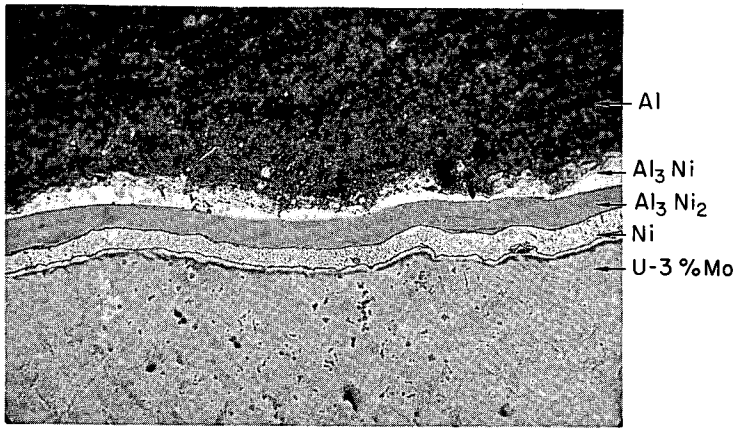
c. Plate 347-1, U – 3.5% Mo – 0.5% Si, from Center Section of HT-1 – Plate Adjacent to 348-3

Figure 20. Burnup Profiles (Burnup in at.% U)



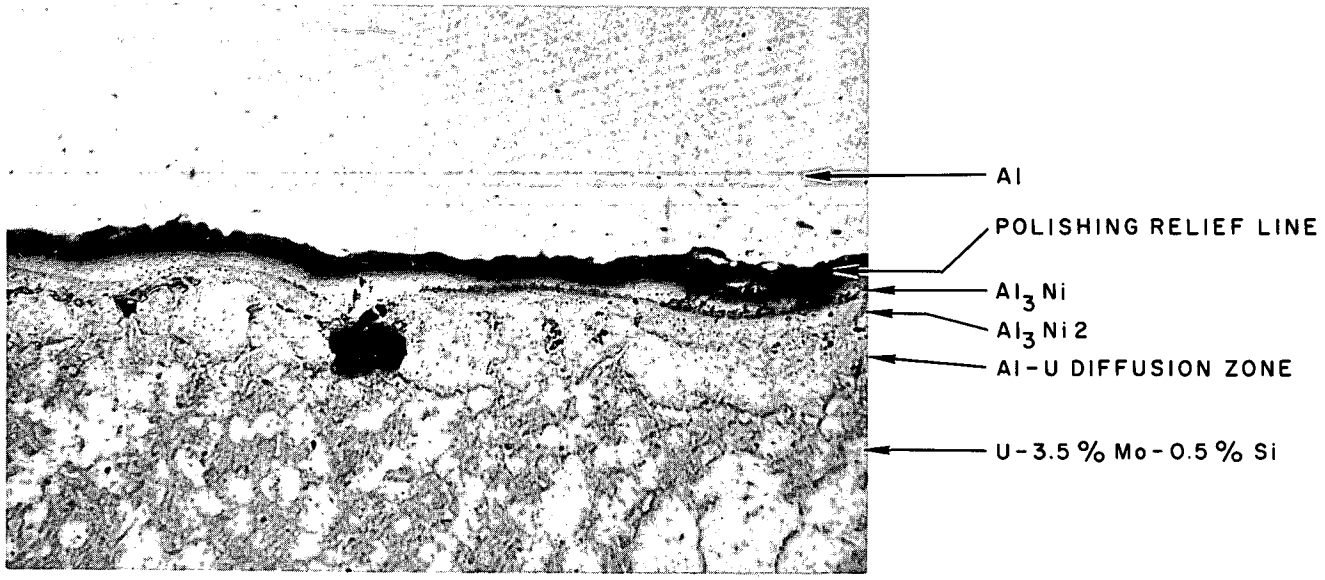
As Polished 4X

Figure 21. Cross Section of Unblistered Area of Plate 347-1



Etchant: $NaCN$, $(NH_4)_2S_2O_8$ 500X

Figure 22. Microstructure of As-Fabricated Bond



Heat Etched 270X

Figure 23. Microstructure of Bond in Unblistered Area of Plate 347-1

TABLE VI

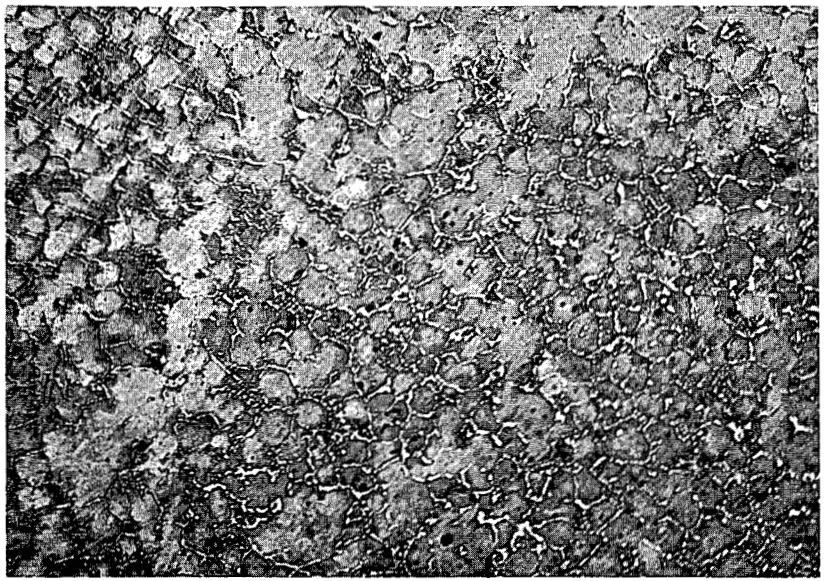
BURNUP OF FUEL PLATES

Plate Number	Element	Alloy	Maximum Burnup (at.% U)	Average Burnup (at.% U)
341-1	HB-1	U - 3.5% Mo	0.23	0.18
348-3	HT-1	U - 3.5% Mo - 0.5% Si	0.19	0.12
347-1	HT-1	U - 3.5% Mo - 0.5% Si	0.15	0.10

F. METALLOGRAPHIC EXAMINATION

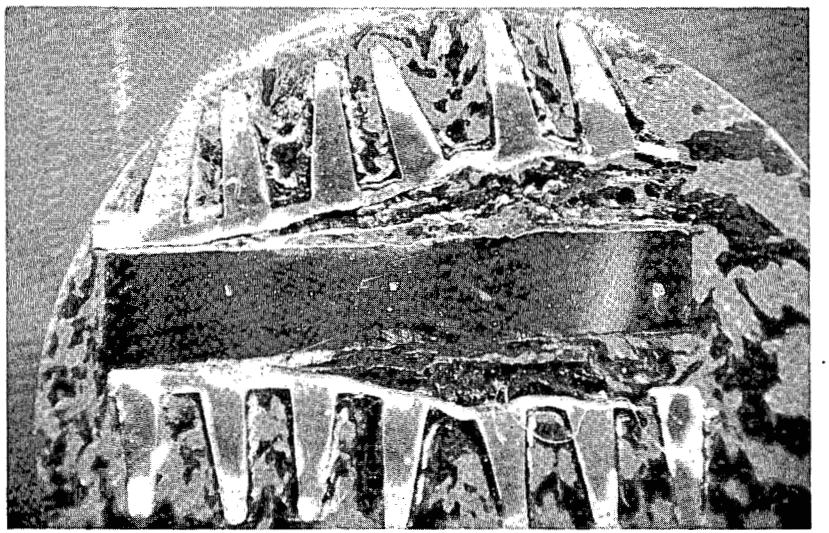
Metallographic sections were cut from the blistered Plate 347-1, from the HT-1 fuel element.[13] The two specimens examined represented a cut across the width of the plate, 4 in. from the outlet end, perpendicular to the plate longitudinal centerline. One specimen represents the blistered side of the plate, and the other specimen represents the adjacent unblistered side. The specimens were mounted in Bakelite, ground with 600 grit silicon carbide papers and polished on Syntron vibratory machines using 1, 0.3, and 0.1 μ alumina on microcloths. The specimens were examined in the as-polished condition, after heat etching with an infrared lamp, and after electrolytic etching. Electrolytic etching was done at 20 volts in four parts each of phosphoric acid, ethyl alcohol, and ethylene glycol.

Figure 21 is a photomacrograph of the unblistered region. The bond between the core and cladding was intact, except for a small split at one edge, which probably occurred during sectioning. The microstructure of the bond area showed almost complete consumption of the nickel bond by the aluminum, to form brittle Al-Ni intermetallic compounds. Some diffusion of aluminum into the uranium core was evident. Figure 22 is a typical photomicrograph of an unirradiated "as-fabricated" bond. Figure 23 is a photomicrograph of a typical bond area from the unblistered specimen, with the probable components identified. Identification of the compounds was based on previous experience with Al-Ni-U diffusion bonds.[8] The dark band due to relief polishing contained no cracks or voids. The microstructure of the core alloy in the unblistered section was very similar to the unirradiated structure, and is shown in Figure 24.



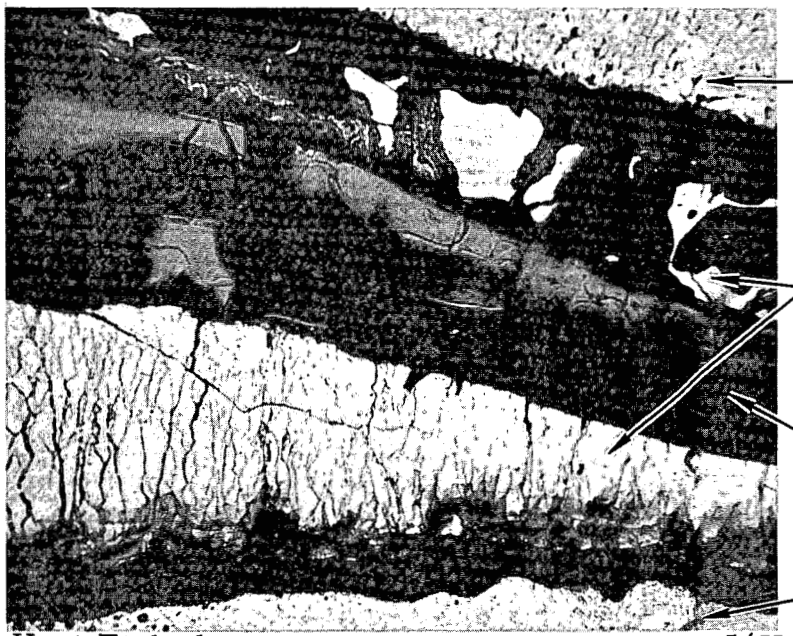
Electropolished 270X

Figure 24. Microstructure of Fuel Core Alloy in Unblistered Area of Plate 347-1



As Polished 4X

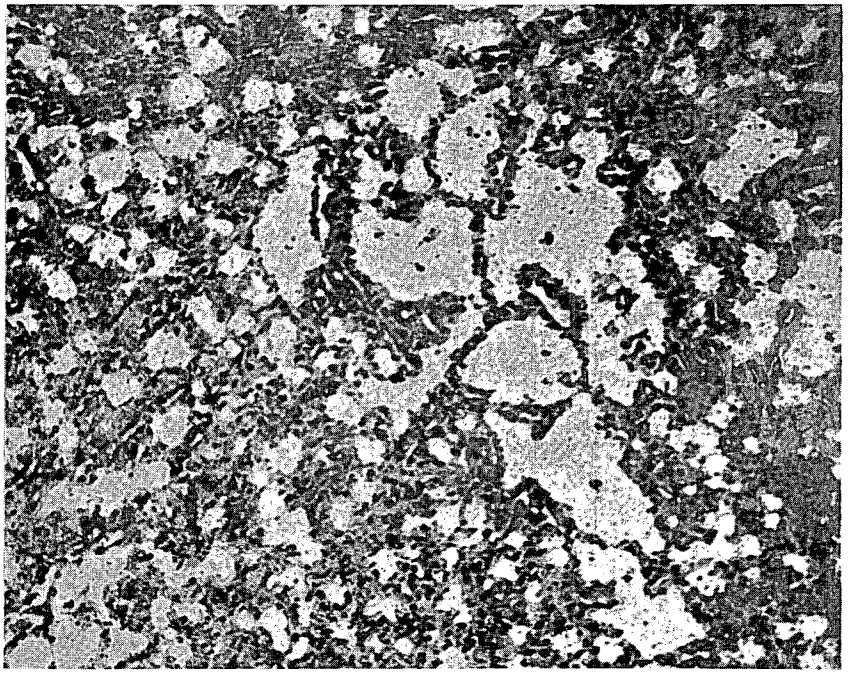
Figure 25. Cross Section of Blistered Area of Plate 347-1



Heat Etched 56X

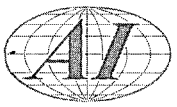
Figure 26. Microstructure of Blistered Area of Plate 347-1

Figure 25 is a photomacrograph of the blistered section. Both sides of the fuel plate were blistered. The space between the cladding and the core is apparently filled with coolant decomposition products and U-Al and Al-Ni intermetallic compounds. One small hole was observed in the cladding. The cladding varied in thickness from 0.020 in. to zero at the break. The Al-Ni compounds contained many cracks. The core material showed no evidence of gross attack by the organic coolant. Figure 26 is a photomicrograph of the blistered area, with the probable components identified. The microstructure of the core alloy was not homogeneous in the region of the blister. Figure 27 shows the affected region.



Electropolished 270X

Figure 27. Microstructure of Fuel Core Alloy in the Blistered Area of Plate 347-1



V. DISCUSSION OF RESULTS

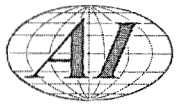
A. VISUAL INSPECTION OF THE EXPERIMENTAL FUEL ELEMENTS

Hot cell examination of both fuel elements revealed that the inlet ends of the coolant channels were practically completely blocked by material filtered out of the coolant. The filtering action of the aluminum fins on the fuel plates was readily apparent when the inlet ends of the fuel elements (Figures 10 and 15) were compared with the outlet ends (Figure 13). The presence of particulate matter in the OMRE, at the time of the experiment, has been separately established. Hot cell examination of a standard OMRE fuel element, removed from the OMRE, revealed several pieces of particulate matter stuck on the inlet ends of the fuel plates.[12] The flat plates in the standard OMRE fuel element are spaced 0.134 in. apart. One of the pieces was analyzed as a piece of rock. Additional evidence of particulate matter is the high radioactivity noted during the 40% HB run. The bulk of this activity was attributed to rust introduced into the residual in the system.

Subsequent to the damage to the HB-1 and HT-1 fuel elements, provisions were made to remove particulate impurities from the OMRE coolant. Filtration and centrifuging of the coolant, and distillation of coolant makeup are presently being performed. Experimental fuel elements presently operating in the OMRE have shown no signs of blockage of coolant flow.

From visual inspection of the HB-1 experimental fuel element, it is apparent that part of the cladding melted, exposing bare uranium alloy core. The melting occurred on the side of the fuel element facing OMRE Core Position 16, and was most pronounced on the third and fourth plates, counting from the side closest to the center of the core. The plate that was closest to the center of the core, and should therefore be the highest power-output plate, was undamaged. The amount of plugging in individual coolant channels was apparently a greater factor in the distribution of the damage than the relative power output of the plates. None of the cladding thermocouples were located at, or in the immediate vicinity of, the melted areas.

On the HT-1 fuel element, the only damage observed was a blister on one fuel plate. As with the HB-1 fuel element, the fuel plate closest to the center of



the core, and therefore the one with the supposedly highest rate of power output, was undamaged. The overheating that resulted in cladding melting in the HB-1 fuel element, and the blister in the HT-1 fuel element, was apparently of a localized nature, depending on the degree of blockage of individual coolant channels. The HB-1 fuel element appeared to have the greatest amount of blocking material at the inlet end. The greater blocking and the higher power output account for earlier failure of the HB-1 element. No evidence of chemical reaction was observed between the coolant and the melted Al-U alloy or the exposed core alloy.

B. DIMENSIONAL STABILITY OF THE FUEL MATERIAL

The only significant change in dimensions of the fuel plates was a small increase in length. The small average changes, shown in Table III, for width and thickness are close to the preirradiation tolerances and are probably less than the accuracy of the hot cell measurements. The 0.070 to 0.084-in. average length increases are believed to be real. Due to the small magnitude of the change (0.6 to 0.7%) and the uncertainties in the irradiation thermal histories of the plates, no conclusions can be drawn as to differences in the relative stability of the two alloys tested. The lack of measurable swelling, as evidenced by increases in width or thickness, is encouraging; particularly since visual observations and metallographic examination indicated localized temperatures in excess of the thermocouple readings.

C. CHEMICAL ANALYSES

Although the analytical results are somewhat scanty, due to the high activity of the samples and the difficulty in obtaining representative samples, some significant information was obtained. The low activity of samples from the blocking material, compared with samples from the rest of the element, indicates that the blocking material did not originate in the element during failure. All of the samples contained only small amounts of coolant. The soluble organic portions of the coolant showed no significant buildup of HB. The incomplete analysis of the organic-insoluble portion of the blocking material, which showed 16% ash with Fe as the principal metal constituent, and 64% unidentified, is similar to the analysis obtained on carbonaceous deposits in damaged OMRE Fuel Element Number 30. These carbonaceous deposits, which very likely had flaked off to

some extent, had an insoluble composition of approximately 75% carbon, 3% hydrogen, and 23% ash. Since the blocking deposits appeared particulate, rather than as a homogeneous deposit, it is likely that they contained a mixture of a variety of foreign materials in the coolant, such as iron oxide from residual rust and makeup coolant and carbonaceous material from the damaged standard element.

The composition of the metallic material (~90% Al) locates it close to the 1180°F eutectic at the aluminum end of the U-Al compositional diagram.[14] The constituents of an alloy of this composition would be U-Al_4 and Al.

D. BURNUP ANALYSES

The burnup distributions shown in Figure 20 and Table VI are similar to those expected. The maximum burnup was located on the edge of the fuel plates, slightly below the vertical centerline of the reactor. Doubling the enrichment in the HB-1 element, over that in the HT-1 fuel element, resulted in an increase of only approximately 50% in burnup, due to the greater flux depression of the more highly enriched uranium and boron steel sections in the HB-1 fuel element. It is significant that the regions of greatest damage in the experimental fuel elements do not correspond to areas of highest burnup.

E. METALLOGRAPHIC EXAMINATION

The postirradiation microstructure of the cladding-to-core bond and of the fuel core showed signs of higher-than-anticipated temperatures in the HT-1 fuel element. In the blistered area, high temperatures were evidenced by the formation of massive intermetallic compounds and the uneven core microstructure, indicative of partial alpha-to-gamma transformation. In this area, a substantial portion of the aluminum cladding had been converted to brittle intermetallic compounds. Fracture of these compounds apparently allowed coolant to enter the bond layer, decompose, and enlarge the blistered area. From the appearance of the core microstructure, it was estimated that the maximum center fuel temperature in the blistered area was between 1200 and 1300°F. The insulating effect of the blister would account for these temperatures.

In the adjacent unblistered area, overheating was evidenced by practically complete consumption of the 0.0005-in. nickel diffusion barrier and some uranium-

aluminum interdiffusion. However, the bond between cladding and fuel was still intact, and no uranium had diffused to the surface of the cladding. Due to the complex temperature history of the fuel element, the consumption of the free nickel bond in the unblistered area cannot be analyzed by time-at-temperature relationships. Recent unpublished information predicts that the 0.0005-in. nickel layer would be completely consumed, as aluminum-nickel intermetallic compounds, during the time of the irradiation (~3000 hr), if the bond temperature were 700°F. Since the recorded cladding temperatures, during the early part of the irradiation, were less than 700°F, most of the compound formation probably took place during final phases of the irradiation, when the temperature increased rapidly, due to coolant channel blockage. In order to provide a greater margin of safety, the nickel bond thickness has been increased to 0.001 in. for the Piqua fuel element design.

VI. SUMMARY AND CONCLUSIONS

Two experimental fuel elements have been irradiated in the OMRE and evaluated. The experimental fuel element designs were based on aluminum-clad, uranium-alloy fuel plates. The finned, aluminum cladding was metallurgically bonded to the core. A 0.0005-in. nickel layer was used to promote diffusion bonding and to prevent interdiffusion between the alumium and uranium. The fuel core alloys irradiated were U - 3.5% Mo and U - 3.5% Mo - 0.5% Si.

Both experimental fuel elements in the OMRE were exposed to abnormal operating conditions and were damaged, for reasons not intrinsic to the fuel element itself. Part of the cladding on the HB-1 fuel element melted, releasing fission products into the reactor coolant. Due to this failure, the experimental elements were removed from the reactor before achieving their planned exposures. Measured temperatures in the cladding generally increased throughout the irradiation and reached a maximum of 770°F in the HB-1 fuel element and 780°F in the HT-1 fuel element. The maximum burnups achieved were 0.23 at.% uranium in the HB-1 fuel element and 0.19 at.% uranium in the HT-1 fuel element.

The fuel elements were disassembled, and all 20 fuel plates were examined individually, to determine the reason for failure and the extent of damage. One U - 3.5% Mo fuel plate, with 0.18 average and 0.23 maximum at.% uranium burnup, and two U - 3.5% Mo - 0.5% Si fuel plates, one with 0.19 maximum and 0.12 average and the other with 0.15 maximum and 0.10 average at.% uranium burnups, were measured to determine the dimensional stability of the fuel alloys. Burnup profiles were established for these three fuel plates by scanning the plates for relative gamma intensity and correlating the gamma intensities with radiochemically analyzed burnup. A blistered area and an undamaged area on a U - 3.5% Mo - 0.5% Si fuel plate were investigated metallographically.

The following conclusions were derived from the irradiation and the evaluation:

a) Particulate material in the OMRE coolant was trapped by the aluminum cladding fins at the inlet ends of both experimental fuel elements.

This foreign material restricted the flow of coolant past the fuel plates and caused the fuel plates to overheat. The overheating caused partial melting of the aluminum cladding and some alloying of the aluminum with uranium from the core, in four of the five plates in the higher enrichment HB-1 fuel element. Damage in the HT-1 fuel element was limited to a blister separation of the core and cladding on one fuel plate. Nothing was observed that would indicate the fuel elements would not have functioned properly in the same coolant, if free of particulate matter of sizes that could be trapped.

b) Under the irradiation conditions experienced by the measured plates, both the U - 3.5% Mo and the U - 3.5% Mo - 0.5% Si alloy showed good dimensional stability. Metallographic observations of the core microstructure in the blistered region, and the melting of the aluminum cladding, showed the localized temperature conditions experienced by the fuel plates to be considerably in excess of the 750°F design temperatures and the 780°F maximum thermocouple temperature. The lack of observable fuel swelling, under these conditions, is encouraging for the use of these alloys as OMR fuels, at considerably higher burnups than those experienced in the test.

c) The 0.0005-in. nickel layer may be inadequate to prevent interdiffusion between the aluminum cladding and the uranium alloy core, particularly if fuel surface temperatures exceed 750°F.

d) No gross reactions occurred between the organic coolant and the uranium alloy core materials, at temperatures up to the melting point of aluminum. This conclusion, which is based on visual and metallographic examination, is substantiated by the fact that metallic fission products were not found in the coolant after the fission break.

VII. FUTURE WORK

Additional experimental fuel element irradiations are in progress, and others are planned, as part of the Organic Reactor program. Two experimental fuel elements, similar in design to HB-1, with U - 3.5% Mo - 0.1% Al alloy fuel are presently in the OMRE. Also in the OMRE is a circular, finned, aluminum clad, U - 3.5% Mo - 0.1% Al fuel, experimental fuel element which is a prototype of the present Piqua design. Irradiation of a UO_2-fueled, experimental fuel element is planned for the near future. Also in the advanced stages of planning is a full OMRE core loading of finned, aluminum clad, uranium alloy fuel.

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