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IRRADIATION OF AN ALUMINUM ALLOY-CLAD,
ALUMINUM-URANIUM ALLOY-FUELED PLATE

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10H	4X
10I	6X
10J	6X
10K	6X
10L	6X
13	4X
14	4X
15	4X
16	11X top, 4X bottom

IRRADIATION OF AN ALUMINUM ALLOY-CLAD, ALUMINUM-URANIUM ALLOY-FUELED PLATE

by

A. P. Gavin and C. C. Crothers

I. SUMMARY

A penetration of the cladding of an aluminum alloy-clad, aluminum-uranium-cored fuel plate undergoing irradiation in the Argonne high-pressure water loop in the MTR caused the irradiation to be discontinued after a maximum burnup of 58% of the original 2.2 atom % of total U^{235} core atoms. The plate was irradiated at 380 psig with a loop water temperature of 420°F for the equivalent of 156 full-power days of MTR operation. Local boiling was allowed in the region of highest flux; the maximum heat flux was 690,000 Btu/(hr)(ft²) initially and 260,000 Btu/(hr)(ft²) at the end of the irradiation.

The cladding failure was first indicated by a high activity level in a sample of loop water taken for pH measurements. Further investigation verified the presence of fission products in the loop water, and the element was immediately removed from the high-flux zone by pulling it back in the pressure tube. A check of the loop water activity monitor charts indicated that the element was probably irradiated in the high-flux position for at least two days after the initial penetration of the cladding occurred. Loop water activity at no time reached a level high enough to set off the high-level alarm, and no appreciable permanent contamination of the loop was noted.

Postirradiation examination revealed the presence of two blister-type defects on one side of the 16-inch long element at a point approximately four and one-half inches from the end which was in the highest flux position in the radial irradiation facility. Both blisters were filled with corrosion products. From the appearance of a cross section taken through them, it was concluded that local corrosive attack caused penetration of the cladding at these points. The local corrosion which produced the cladding penetration is attributed to operation of the test loop with water of high pH for a period of seven days prior to the detection of the rupture.

Swelling was observed in the core in the portion of the plate which was subjected to the highest neutron flux. The swelling, which was attributed to the combination of high burnup and high fuel temperatures,

was not the cause of the failure of the cladding. It is estimated that the fuel reached temperatures in the neighborhood of 1000°F as a result of operation at high heat flux and buildup of a heavy oxide scale layer.

A corrosion rate of 10 to 15 mils/yr was observed on the irradiated plate. This high rate is attributed mainly to the small ratio of aluminum surface area to loop water volume which existed. Operation with water of high pH for a short period of time also caused accelerated corrosion, but this factor was not considered to account for more than a small percentage of the corrosion loss observed.

II. INTRODUCTION

The experiment described in this report, designated MTR-W11, was designed to determine the effect of high burnup on an aluminum alloy-clad, aluminum-uranium alloy core plate of the type manufactured at Argonne for the Argonne Low Power Reactor (ALPR). The irradiation temperature selected (420°F) was the same as that of the ALPR. With the temperature set at this value, calculations were made to determine the lowest possible pressure which would allow the maximum possible local boiling on the plate and still assure a margin of safety against burnout. The value finally agreed upon by the MTR Safety Committee and Argonne personnel was 380 psig.

The maximum burnup which the ALPR core is expected to reach is 2% of the total core atoms. Calculations indicated that an irradiation period of 13 cycles in the ANL-2 Loop would produce a burnup of this magnitude in the portion of the plate which was to be exposed to the highest flux.

Since rupture of the cladding was not contemplated, no definite procedure to be followed in event of such an occurrence was set up. Fortunately, many experiments with fissionable material in loop water have been conducted by various people at the MTR, and equipment and procedures were readily available to obtain a considerable amount of pertinent information in the short period of time the ruptured element was in the high-flux zone.

III. TEST SPECIMENS

The test specimens were manufactured by the Argonne Metallurgy Division using the same procedures and materials used in the manufacture of the ALPR core.^(1,2,3) Figure 1 is a drawing of the miniature plates used in this irradiation.

The cladding material was M388 aluminum alloy, which consists of 2S aluminum with 1% nickel added. The core material was an alloy consisting of 17.5 w/o uranium, 2 w/o nickel, the remainder being 2S aluminum. The uranium content was enriched in U^{235} to 93%.

The ALPR plates are constructed of punched "picture frames" and core blanks, which are assembled with the side plates, elemental silicon being used as a bonding agent on the surfaces to be bonded. The assembly is first hot pressed at 600°C and 1000 to 2000 psi, and is then reduced to finished thickness by rolling. Three hot passes at 550°C and a final cold pass are employed. After the exact location of the core section is determined by radiographing, the plates are trimmed to final dimensions.

In order to produce the miniature plates for this experiment, it was necessary to procure a set of smaller dies for punching the "picture frames" and core blanks. With this exception the process and equipment used was the same as that used on the full-sized plates.

The bonding integrity was checked by annealing at 550°C for one hour and applying a 100% ultrasonic transmission test. No blistering or unbonded areas were detected.

A complete set of length, width, and thickness measurements was made on the two plates to be used as the in and out-of-pile specimens for the irradiation experiment.

Since the holding cartridge to be used in this experiment had been used in a previous irradiation, it was necessary to load the W11 in-fuel plate into it remotely in a cave. Both the in and out-of-pile plates in their holding cartridges were then shipped to the MTR to be irradiated.

Fig. 1
Drawing of Test Specimen

IV. IRRADIATION

A. ANL-2 Loop

The test plate was irradiated in the ANL-2 Loop⁽⁴⁾ in the Materials Testing Reactor at the National Reactor Testing Station in Idaho. This loop is located in a radial beam hole through the graphite reflector of the reactor. The tip of the pressure tube is located near the edge of the reactor core in a cylindrical well in the reactor tank. Due to the radial position of the facility, the flux drops rapidly from the in-reactor end of the fuel section to the outer end.

Loop flow is channeled in through a rectangular tube, approximately 1 in. by 2 in., located in the center of the cylindrical pressure tube and out through the space on the outside of the rectangular tube. The fuel element is located at the inner end of the rectangular tube by means of a holding cartridge (see Figs. 2 and 3).

Fig. 2
Specimen-holding Cartridge

Fig. 3
ANL-2 In-pile Tube

Flow through the loop is maintained at the desired rate by circulation pumps, and bulk water temperature is maintained by the use of a temperature controller and electric heaters. Water quality is maintained by means of a bypass ion-exchange system (Fig. 4). This system consists of three mixed-bed ion-exchange columns, and three sulphite deoxygenating columns. For this experiment the system was valved so that flow was through one sulphite bed and one mixed bed in series.

Fig. 4
Schematic Drawing of ANL-2 Loop in MTR

To distinguish between the effects of irradiation and those of exposure to loop water without irradiation, a second exposure location is provided in the external portion of the loop.

B. Conditions of Irradiation

The irradiation was conducted during MTR cycles 114 through 124. Each MTR cycle is three weeks in duration, including a shutdown period of from one to five days for refueling and manipulation of experiments. The test plate was actually in the loop for 227 days and was subjected to the equivalent of 156 days of full-power (40 Mw) operation of the MTR.

The flux values used in the calculations for this experiment were taken from a flux run made at low power for a previous experiment. A subsequent full-cycle flux run produced somewhat lower, but more reliable, values. The results of this flux run together with the actual perturbed flux as determined from burnup determinations on fuel samples taken from the test plate after completion of the test are plotted in Fig. 5.

Fig. 5
Thermal Neutron Flux Plot for ANL-2 In-pile Tube

The temperature of the loop for this experiment was fixed at 420°F to duplicate that of the ALPR. The pressure was then calculated to allow as much local boiling on the plate as possible and still maintain a reasonable margin of safety from burnout.

The desired water conditions were neutral pH and a resistivity greater than 1 megohm.

During periods when the reactor was not operating, the temperature of the loop water was maintained at 350°F by use of additional line heaters.

A complete tabulation of the conditions which existed in the loop during the irradiation period, with the exception of the last of the eleven cycles, are presented in Tables 1 and 2.

Table 1

SUMMARY OF CONDITIONS OF IRRADIATION

Total Time in Loop	227 days (MTR cycles 114-124)
Total Neutron Exposure	6293 Mwd
Temperature (during irradiation) 415-425°F above 400°F	91.2% of time subject to irradiation 93.7% of time subject to irradiation
Temperature (total time in loop)	See Table 2
Pressure	380 psig
Flow Rate	60 gpm 11 to 12 ft/sec past test specimen
Ratio of Aluminum Surface Area to Loop Water Volume	5.8 cm ² /ℓ
Water Resistivity*	0.8 to 3.4 megohm-cm(1)
pH*	
Average	7.1(2)
Maximum	7.6
Minimum	6.7
Dissolved Oxygen*	Less than 0.08 ppm
Dissolved Hydrogen*	
Average	23 cc/ℓ
Maximum	31 cc/ℓ
Minimum	11 cc/ℓ
Dissolved Nitrogen*	
Maximum	2.3 cc/ℓ(3)
Power Fluctuations	
Full power cycles	42
Power dips (40 Mw to one- half power)	5

(1) During the first cycle the resistivity varied between 0.25 and 0.8 megohm-cm for a total of approximately 190 hr.

(2) During the first cycle the pH varied between 7.9 and 8.3 for approximately 190 hr.

(3) Gas samples taken the fourth day of the first cycle and the seventh day of the sixth cycle indicated 84 and 69 cc nitrogen per liter of water. In each case degassing of the loop reduced this to below the amount indicated above.

*Values listed are exclusive of the MTR cycle during which the failure occurred. Conditions of this period are described in detail in another section of this report.

Table 2

TEMPERATURE OF EXPOSURE FOR W-11 FUEL ELEMENT

Total Exposure Time - 227 Days

Temperature of Loop (°F)	% of Time above Temperature Indicated	Days above Indicated Temperature
180	100	227
200	96	219
220	96	219
240	96	219
260	96	$217\frac{1}{2}$
280	95	216
300	90	$204\frac{1}{2}$
320	89	$203\frac{1}{2}$
340	89	203
360	85	194
380	68	155
400	67	153
420	66	149

V. FISSION BREAK

A. Detection

On July 7, 1959, after the element had been in the loop for 215 days and had had an equivalent of 156 full-power (40 Mw) days of exposure to irradiation, a 200-ml sample of water taken from the loop for the purpose of pH measurement was observed to read 1.0 r/hr beta-gamma. This was the first indication that a fission break had occurred. A closer examination of the loop water activity monitor charts indicated a gradual increase in activity, probably beginning on July 5, but possible as early as July 3. A plot of water activity during this period is presented in Fig. 6. After a Cs^{138} analysis verified the fact that a break had occurred, the element was pulled back in the pressure tube a distance of five feet by means of pull rods which had been provided for this purpose (see Fig. 3).

Results of various analyses of loop water samples taken during the period following the fission break are presented in Tables 3, 4 and 5.

Table 3

GROSS ACTIVITY OF 100 ml OF LOOP WATER

Date	Time	Activity (mr/hr)*
7-3-59	0440	120
7-3-59	2311	5
7-4-59	0530	140
7-5-59	0145	150
7-7-59	0255	500

*Readings taken with hand counter in contact with sample container. Normal readings range from 5 mr/hr to 100 mr/hr.

Table 4

RADIOCHEMICAL ANALYSIS OF LOOP WATER

Sample Time		Nuclide	d/(min)(ml)
Date	Time		
7- 7-59	1002	I^{131}	1.20×10^6
7- 7-59	1002	I^{133}	4.48×10^6
7- 7-59	1002	Ba^{140}	1.10×10^5
7- 7-59	1002	Cs^{138}	7.15×10^6
7- 8-59*	1432	Cs^{138}	1.45×10^6
7-10-59	1002	Cs^{138}	1.00×10^6
7-17-59		Cs^{138}	1.81×10^5

*Fuel plate withdrawn from flux at 1300 on 7-7-59.

Table 5
CHEMICAL ANALYSIS OF LOOP WATER

(After Withdrawal on 7-8-59 of Ruptured
Element from High-flux Zone)

Time	Element	Concentration	Remarks
1000	H ₂	110 cc/kg	Blacet-Leighton method
1000	O ₂	Nil	
1000	N ₂	Nil	
1000	O ₂	Nil	Winkler method
1330	Na	0.1 ppm	
1330	Li	less than 1 ppb	
1330	K	less than 25 ppb	
1330	Al	less than 1 ppm	Flame Spec.

It is of interest to note that the operation of the MTR was in no way affected during this entire series of events. The activity levels encountered in the loop were not high enough to cause a reactor scram and the element was retracted from the high-flux zone without affecting the operation of the loop.

B. Loop Operating Conditions prior to Fission Break

Loop operation during the previous MTR cycle (cycle 123) was satisfactory and all conditions were within the desired limits. Toward the end of this cycle, the pressure drop across the ion-exchanger inlet filter was becoming excessive, and conductivity measurements of the loop water indicated that the loop demineralizer was becoming exhausted. On June 29, during the shutdown period prior to cycle 124, the cycle during which the break occurred, the ion-exchange filter was bypassed and spare sulphite and mixed bed ion-exchange columns were valved into the system.

Subsequently it was found impossible to maintain the desired conditions of water purity and pH. The pH rose from the normal value of approximately 7 to a value of from 9 to 10. The resistivity of the water dropped to approximately 50,000 ohms during the same period.

On June 30, the pressure drop across the in-pile tube increased from 8 to 16 psi over a six-hour period. Differential pressure changes of this magnitude had occurred in the loop several times previously, usually during periods following reactor transients, so this occurrence in itself did not cause any great concern. During the following six-day period, this pressure drop decreased to 5.4 psi, which is within the range of normal operating conditions.

The reactor was brought up to power for cycle 124 on July 2. Within five hours after startup a loop water temperature of 418°F was reached. Approximately nine hours after startup, a high reading on a tube tip temperature thermocouple gave the loop automatic cool down. For the following 60 hr the loop was maintained at below 360°F due to loss of flow control. The maximum flow which could be maintained during this period was 56-58 gpm. On July 5, flow control was re-established and the loop water temperature was increased to 420°F. On July 7, after the fission break had been established, the loop temperature was reduced to 350°F. Within eleven hours the element was withdrawn from the flux zone and the temperature was maintained as low as possible (270 to 290°F) for the rest of cycle 124 (15 days).

On July 1, it was found that no reading could be obtained on the loop water resistivity instrument. The instrument was removed from the loop and checked but no malfunction was detected. A plot of the pH readings taken during this period is presented in Fig. 6.

C Loop Operation Conditions after Removal of Ruptured Element

The ruptured element was transferred on July 19 from the loop to a shipping cask by the established procedure.

A radiation survey of the ANL-2 equipment cubical made on July 13 indicated no gross increase in radiation hazard due to the element failure. Due to corrosion product buildup in the various components in the cubical, the background level of radiation was quite high before the rupture occurred. This high initial level made it impossible to detect any small degree of contamination which might have occurred.

As is indicated in Table 4, the Cs^{138} activity of the water dropped rapidly after removal of the element from the flux zone. Some residual contamination of the loop with fissionable material is indicated by the fact that the loop water contains 1.1×10^4 d/(min)(ml) of Cs^{138} activity even after several cycles of operation subsequent to the removal of the ruptured element. Since no ruptures had occurred in the loop prior to this irradiation, this failure is the only source of fission material in the loop.

At the beginning of cycle 130, fifteen weeks after the removal of the ruptured element from the loop, an increase in the loop activity, as indicated by the activity monitor on the outlet of the in-pile tube was noted. The recorded value was approximately one and one-half times the normal reading. A Cs^{138} analysis indicated 2.8×10^4 d/(min)(ml). Since the fuel element being irradiated at this time was a uranium alloy meat plate clad with Zircaloy-2, a type that would be expected to fail in a much more

Fig. 6
Plot of Loop Operating Conditions for Period Prior to Fission Break

violent manner, it did not seem likely that this element had ruptured. The fission product activity was attributed to a burst of crud containing uranium from the MTR W-11 element. This proved to be the case, since no additional buildup of activity was noted and the loop activity level was back to the normal level of 1.1×10^4 within three days.

VI. POSTIRRADIATION EXAMINATION

A. Procedure

1. Appearance and Physical Measurements

After removal from the loop, the irradiated specimen was shipped to Argonne, where it was placed in a hot cell for examination. After removal from its holding cartridge, the plate was examined visually through the cave window and through a low-magnification periscope. Pictures were taken before any of the scale was detached by handling. After examination of the plate surface under magnifications of up to 13X and photographing of areas of interest, thickness, width, and length measurements were taken. A similar examination was made on the out-of-pile plate.

2. Removal of Section Containing Rupture

In order to prevent disturbance of two blisters, which were apparently the point of cladding failure, a section, the full width of the plate and approximately one-inch long, containing these defects was sawed from the plate with a power hacksaw (see Section F in Fig. 7). The section was then given to the Metallurgy Division for metallurgical examination.

3. Scale Removal

Oxide scale was removed from the two pieces of the irradiated plate which remained, as well as from the out-of-pile plate. An electrolytic descaling technique with a bath of saturated boric acid as the electrolyte and an alternating current density of approximately 0.02 amp/sq cm was employed to remove the scale. Samples of oxide were retained for analysis.

4. "Clean" Appearance and Physical Measurements

The plate was re-examined in the "clean" condition, both through the cave window and under the microscope. Pictures were again taken of areas of interest, and another set of thickness and width measurements was taken.

5. Sampling for Burnup Analysis

Samples for burnup analysis consisted of $\frac{1}{8}$ -inch punchings taken through the entire thickness of the plate. Nine sets of two punchings each were taken at various intervals along the length of the plate (see Fig. 7). Five of these sets were submitted to the Chemical Engineering Division for burnup analysis.

6. Sectioning of High Burnup Portion of Plate

Thickness measurements and visual examination had revealed the presence of swelling in the portion of the plate which had been subjected to the highest flux. In order to examine this portion of the plate metallurgically, a section extending from one to two inches from the in-reactor end was cut out with a hacksaw (see Section B in Fig. 7). This section was then submitted to the Metallurgy Division for metallurgical examination.

In addition, a portion extending from two to two and one-half inches from the in-reactor end was cut off and sectioned to allow separation through the meat (see Section C in Fig. 7). The section which remained between the cut at two and one-half inches from the in-reactor end and the cut made at four and one-fourth inches from the in-reactor end to remove the blistered portion was then sectioned lengthwise through the center in an effort to determine the extent of the swelling and delamination of the core (see Section D in Fig. 7).

B. Results

1. Appearance

Figure 8 shows both sides of the plate as it appeared when removed from the holding cartridge upon return from irradiation in the MTR loop. Figures 8-A through 8-D are photomicrographs of portions of the plate in this condition.

Two blisters on one side of the plate between $4\frac{1}{2}$ and 5 in. from the in-reactor end are apparent in these photographs. These blisters, together with the scale which had peeled back from the surface of the plate in the immediate vicinity of each blister, are also apparent in the edge view shown in Fig. 9.

Figure 10 is a view of both sides of the plate after removal of the oxide scale. The dark area on the in-reactor end of the plate was the zone subjected to the highest neutron flux. The dark appearance is due to scale which was not removed by the descaling process. The section containing the two blisters was cut from the plate prior to descaling and, although it is included in this picture to indicate position, it was not subjected to the descaling process.

Photomicrographs of typical areas of the plate surface and of all abnormalities noted in a complete microscopic examination of the plate surface are presented as Figs. 10A-L.

The appearance of the out-of-pile fuel plate before and after removal of scale is illustrated in Figs. 11 and 12.

(BOTTOM ARROWS INDICATE LOCATION OF PUNCHINGS FOR BURNUP ANALYSIS)

Fig. 7
View of W-11 Plate Showing Sections Made in Examination

Fig 8
Irradiated W-11 Plate as Removed from Reactor

Fig. 8A

Top View of Ruptured Blister

Fig. 8B

Top View of Unruptured Blister. Although no hole was apparent in the initial examination of this blister, further examination definitely established that penetration of the cladding had occurred at this point

Fig. 8C

Typical view of surface in high-flux area

Fig. 8D

Typical view of plate surface in center of plate

Fig. 9

Edge View of Plate as Removed from Reactor

Irradiated Plate after Removal of Scale

Fig. 10

Fig. 10A

Typical view of plate surface in high-flux region. The descaling process did not remove all of the oxide in this area.

Fig. 10B

Typical view of plate surface in high-flux region. Note the pitting of the oxide which was not removed by the descaling procedure. This attack was noted over an area of approximately 1 sq. in. in the region indicated in Fig. 10.

Fig. 10C

View at edge of plate in high-flux region. The lighter colored region on the left-hand side is the "picture frame" portion of the plate with no fuel beneath. Dark area is fueled portion of plate with unremoved oxide on surface.

Fig. 10D

View at edge of plate showing fuel to picture frame transition. Picture frame is at bottom and fueled portion is at top.

Fig. 10E

View at edge of plate showing fuel to picture frame transition. In both this and the previous photograph the fuel portion is slightly raised above the picture frame portion, indicating swelling of the meat and good bonding of the frame to side plates.

Fig. 10F

Fig. 10G

Views of plate surface over swelled portion in high-flux region. Note cracked appearance of the plate surface in this region.

Fig. 10H

Typical edge view of plate. Some slight preferential corrosion at bond line is apparent but no severe attack is indicated.

Fig. 10I

Edge view at in-reactor end.

Fig. 10J

Small blister in bond line. Blister indicates attack along bond from edge of plate.

Fig. 10K

Blister in bond line at high-flux end of plate. This and the blister in the preceding photograph were the only indication of penetrating attack along the bond line on the edges of the plate. There was no evidence that the attack reached the fueled portion of the plate.

Fig. 10L

Pitting attack on edge of plate. This type of attack was noted in only a few places along the edges of the plate.

Fig. 11

Out-of-Reactor Plate as Removed from Loop

Fig. 12

Out-of-Reactor Plate after Removal of Oxide Scale

Table 6

PHYSICAL DIMENSIONS OF IN AND OUT-OF-PILE SPECIMEN

Thickness Measurements
(All measurements in inches)
In-pile Plate (66-14-1)

In. from In-Reactor End	Original Plate				As Removed from Reactor				With Scale Removed			
	Top	Center	Bottom	Average	Top	Center	Bottom	Average	Top	Center	Bottom	Average
14	0.1198	0.1200	0.1201	0.1200	0.1183	0.1174	0.1170	0.1176	0.1047	0.1072	0.1060	0.1060
12	0.1197	0.1200	0.1200	0.1199	0.1177	0.1168	0.1197	0.1181	0.1066	0.1056	0.1071	0.1064
10	0.1196	0.1200	0.1200	0.1199	0.1144	0.1200	0.1187	0.1177	0.1045	0.1062	0.1067	0.1058
8	0.1196	0.1200	0.1200	0.1199	0.1166	0.1229	0.1172	0.1189	0.1077	0.1112	0.1085	0.1091
6	0.1200	0.1200	0.1200	0.1200	0.1214	0.1314	0.1248	0.1259	0.1075	0.1136	0.1090	0.1100
4	0.1196	0.1200	0.1201	0.1199	0.1245	0.1290	0.1255	0.1263	0.1166	0.1186	0.1235	0.1196
2	0.1198	0.1200	0.1200	0.1199	0.1218	0.1242	0.1267	0.1242	0.1192		0.1233	0.1213
Average	0.1197	0.1200	0.1200	0.1199	0.1192	0.1231	0.1213	0.1212	0.1095	0.1104	0.1120	0.1107
Out-of-Pile												
				Average original					0.1206			
				Average as removed from reactor					0.1185			
				Average with oxide removed					0.1102			

2. Physical Measurements

Table 6 lists the measurements taken on the in-pile plate before irradiation, after irradiation with scale still on, and after removal of scale. All measurements taken on the out-of-pile plate were very close to the average and since no local variations were noted only the averages are presented in Table 6.

Oxide thickness was determined by subtracting the "clean" measurements from the "as removed from reactor" values at the same points. Corrosion loss was determined by the difference between the original measurements and the "clean" measurements at the same points. The results of these determinations are listed in Table 7.

Table 7

OXIDE THICKNESS AND CORROSION LOSS (determined from thickness measurements)

In-pile Plate		
Inches from In-reactor End	Scale (mils/side)	Corrosion (mils/side)
14	6.8	7.0
12	5.8	6.7
10	5.9	7.0
8	4.9	5.4
6	7.9	5.0
4	3.3	0.2
2	1.5	0.7
<div> <div> Scale Not Completely Removed </div> <div> } </div> <div> Area of Swelling </div> </div>		
Average outer 10 in.	6.3	6.2
Out-of-pile Plate		
Inches from out of Reac- tor End	Oxide (mils/side)	Corrosion (mils/side)
Average	4.1	5.2

Due to the swelling which occurred on the part in the zone of highest flux (readings at 2 and 4 in. from the in-reactor end), the values obtained for corrosion loss by difference in thickness measurements in this area were of no value. Likewise, values for oxide thickness in this zone could not be determined in this manner due to the oxide which could not be removed by the descaling treatment.

3. Cross Sections through Fuel

The portions of the section cut from the plate for examination of the blisters which contained the opened blister and the blister which appeared to be unopened were mounted and polished for examination of the interior of the plate at center of these defects. Figures 13, 14 and 15 are photographs of the sections thus prepared.

A portion of the swelled section of the plate (Section B in Fig. 7) was also mounted and polished. Figure 16 shows this section. The unpolished edge view of the cut made at $1\frac{1}{4}$ in. from the in-reactor end (Fig. 17) gives a rough indication of the extent of the swelling at this point. Separation within the meat in this portion of the plate is illustrated by the opened section pictured in Fig. 18. There was no evidence of gross swelling or separation within the meat beyond a point approximately $2\frac{1}{4}$ in. from the in-reactor end of the plate. The surface of the lengthwise cut made on the portion of the plate extending beyond this point was not mounted and polished, so minor defects could not be detected.

Fig. 13

Polished Cross Section through Ruptured Blister

Fig. 14

Polished Cross Section through Ruptured Blister

Fig. 15

Polished Cross Section through Unruptured Blister

Fig. 16

Polished Cross Section through Swelled Portion of Plate

Fig 17

Rough Cross Section through Swelled Portion of Plate (Cut between sections A and B in Fig. 7).

Fig. 18

Open Section of High Burnup Portion of Plate Showing Internal Surface where Separation Occurred. (Section C in Fig. 7).

4. Scale Analysis

Table 8 presents the results of analysis of oxide samples from the in and out-of-pile plates. Table 9 lists the results of x-ray diffraction analysis of these samples.

Table 8

SPECTROCHEMICAL ANALYSIS OF SCALE (% of Metal Present)

Sample	Al	Fe	Ni	Ci	Others
From high-flux area of plate	vs	s	6	1	<2
Loose, black scale from around blisters	vs	s	4	0.5	<2
From out-of-pile plate	vs	1.5	1	0.06	<2

vs - very strong, 90-100%

s - strong, 10-100%

Table 9

X-RAY DIFFRACTION ANALYSIS OF SCALE

Sample	Composition	Remarks
From high-flux zone	Corundum, Bohmite and Spinel	~ 50% Corundum and 40% Bohmite
Loose, black oxide from around blister	Bohmite, Corundum and Spinel	> 90% Bohmite
From out-of-pile plate	Bohmite and Corundum	> 90% Bohmite

Measurement of the thermal conductivity of a piece of scale from the in-pile element with an apparatus developed at Argonne by R. L. Leyse⁽⁵⁾ yielded a value of 0.56 Btu/(ft²)(°F)/ft.

5. Burnup Analysis

Table 10 includes the results of burnup analysis of the $\frac{1}{8}$ -inch punchings taken from the irradiated plate. The number of fissions was determined by a Cs¹³⁷ analysis, and the amount of uranium remaining was determined chemically. Values of flux required to produce this burnup were calculated and are plotted in Fig. 5.

Table 10

BURNUP ANALYSES OF FUEL

Inches from In-reactor End of Element	% U ²³⁵ Fissioned
$\frac{7}{16}$	58.3
$1\frac{1}{16}$	48.2
$2\frac{1}{8}$	45.3
$4\frac{1}{16}$	30.0
$12\frac{1}{8}$	7.55

VII. DISCUSSION OF RESULTS

A Cladding Failure

From an examination of the curve of water activity in Fig. 6 it appears that the initial penetration of the element cladding probably occurred at about noon on July 5. There is a steady rise in loop water activity from this time until the element was withdrawn from the high-flux zone on the afternoon of July 7.

At the time of the failure it is estimated that the heat flux at the point of failure was approximately $160,000 \text{ Btu}/(\text{hr})(\text{ft}^2)$ and that the fuel surface temperature was between 500 and 600°F , values depending on the thickness and thermal conductivity of the oxide scale layer.

It is apparent from the cross sections through the plate at the points of failure (Figs. 13, 14, and 15) that the attack which penetrated the cladding was local and not general in nature. The operation of the loop with water of high pH for seven days prior to the detection of the failure could have been the cause of this attack. If this condition was not completely responsible, it is certain that the attack was accelerated by it.

Hanford has reported corrosion rates of 9 mils per month with extensive erosion and pitting attack for M388 aluminum alloy in water at pH 8.5-9.5 in a dynamic corrosion loop at a temperature of 290°C (554°F) (6). This test was made at a small ratio of aluminum surface area to water volume, similar to the conditions which existed in the ANL-2 loop during the W-11 irradiation. Although the bulk water temperature of 420°F was considerably lower than that of the Hanford test, the temperature of the corroding aluminum surface was undoubtedly substantially higher as a result of the temperature rise through the scale.

On the other hand, if the temperature of the corroding surface beneath the oxide layer was above the saturation temperature of the water at the pressure existing at that point, the surface would be blanketed with steam formed in the pores of the oxide layer. This would in effect protect the surface from the high pH water of the loop. If this is the case, the maximum temperature at which the aluminum surface was in contact with the water of high pH would be approximately 447°F , which is the saturation temperature for water at a pressure of 409 psia. This pressure was determined by adding to the loop outlet pressure, 380 psig, the atmospheric pressure 13 psi, and the maximum pressure drop across the loop just prior to the occurrence of the cladding penetration 16 psi.

This discussion suggests another possible corrosion mechanism which could have played a part in the failure of the cladding. In a series of experiments in superheated steam at 450°C (842°F), Ruther and

Draley⁽⁷⁾ found that M-388 aluminum alloy samples completely disintegrated in three days. No experiments were run at lower temperatures so the temperature where this type of attack by superheated steam begins is unknown. While the maximum surface temperature calculated in the region of failure was 500°F, it is possible that chips of oxide adhered to the surface in local areas and caused somewhat higher temperatures in these areas.

The fact that the failure of the cladding of the element was detected and the element removed from the flux zone without affecting the operation of the MTR in any way is considered to be indicative of the safety of using similar elements in an actual reactor core. The element operated at a heat flux of 160,000 Btu/(hr)(ft²) for at least two days after the initial penetration occurred. From Figs. 13, 14, and 15 it is evident that considerable surface of the meat was exposed. In spite of this, the amount of uranium and fission products which were released to the loop water was not sufficient to raise the loop activity above the alarm point. The lack of permanent high-level contamination in the loop is also a point in favor of using elements of this type.

B Corrosion Rate

Table 7 indicates a loss in thickness due to corrosion of 6.2 mils per side on the in-pile plate and 5.2 mils per side on the out-of-pile plate. These figures are based on thickness measurements taken before exposure and after removal of oxide scale from the irradiated element. Measurements of the cladding thickness on the photomicrographs of the polished cross sections (Figs. 13, 14, and 15) indicated a loss of approximately 7 mils per side at the location of the defects. These losses represent a corrosion rate for the in-pile plate of from 10 to 15 mils per year at the conditions of this test.

This high corrosion rate might be explained by two factors. The factor which probably accounts for the major portion of the corrosion loss is the small ratio of aluminum surface area to loop water volume which existed during this irradiation. The value calculated for this experiment is 5.8 cm²/liter of water. A typical reactor fueled with aluminum-clad elements would be expected to have a ratio of over 300 cm² of aluminum surface per liter of water in the primary system. The fact that M388 alloy corrodes much more rapidly in a system with a small ratio of aluminum surface area to water volume has been demonstrated in several out-of-pile experiments.^(8,9)

The second and less important factor in the high overall corrosion of the test specimen is the operation of the loop with water at pH 10 for a period of one week prior to the failure of the cladding. A corrosion rate of the magnitude noted in the Hanford test mentioned above could be responsible for, at the most, one to two mils of the total corrosion observed

In a previous experimental run in the ANL-2 loop, a fuel plate of the BORAX-IV type, which was also clad with M388 alloy, was exposed to water conditions similar to those of this experiment but at a somewhat higher temperature (465°F).⁽¹⁰⁾ The corrosion rate determined for the BORAX-IV plate was very close to that determined for the W-11 plate. In the test of the BORAX-IV plate, water conditions were maintained within the desired range with no period of sustained operation at high pH as in the W-11 irradiation. From this comparison it may be concluded that the portion of the corrosion of the W-11 plate attributable to the high pH operation is not a large percentage of the whole.

In view of the above discussion, the corrosion rate of the element irradiated in this experiment is not considered to be indicative of the corrosion rate to be anticipated in a reactor fueled with elements clad with this alloy.

C. Bond Performance

There is no evidence of gross failure in the bond between any of the component parts of the plate. The small blisters on the edge views in Figs. 11J and 10K indicate some preferential corrosion at these points. These are the only points along the edges that showed any evidence of accelerated corrosion along the bond line.

The cross sections of the plate through the points of cladding failure (Figs. 13, 14 and 15) indicate preferential attack along the bond between the clad and the meat after penetration of the cladding. This attack is not considered serious inasmuch as the element was operated at a high heat flux for from two to four days after the initial penetrations of the cladding. During this time the failure was easily detectable and had not progressed to a point where any difficulty would be experienced in removing a defective assembly from a core composed of similar elements.

D. Scale and Fuel Temperature

Table 7 indicates an average scale thickness of 6.3 mils on the in-pile plate. The maximum thickness indicated is 7.9 mils at six inches from the in-reactor end of the plate. It should be noted that no determinations were made on the inner four inches of the test specimen due to the fact that complete removal of scale in this region was not accomplished by the method used.

One chip of scale recovered from the stripping bath used for stripping the "hot" end of the plate was measured with a micrometer and found to be 10 mils thick. It was impossible to determine the exact location from which this chip was removed except that it came from the surface of the four inches of the plate which were on the in-reactor end.

From the photograph of the plate in the "dirty" condition (Fig. 8), it appears that, since this chip was almost $\frac{1}{2}$ in. across and had a relatively smooth surface, it probably came from the region between two and four inches from the in-reactor end of the plate. The oxide on the first two inches of the plate seemed to be broken up into smaller pieces than this and also has a different surface appearance.

This piece of scale was used for the determination of thermal conductivity; a value of $0.56 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F})/\text{ft}$ was obtained. While the method used for this determination has not been developed to a point where the values obtained can be relied upon as absolutely accurate, it is considered that the value obtained for this scale is entirely reasonable and definitely the best available at this time.

Considering the range of thickness values and the uncertainty of the conductivity of the scale, it is impossible to calculate fuel temperatures with any degree of certainty. The lack of knowledge as to the rate of scale buildup and the falling heat flux due to burnup makes it difficult to determine even a maximum temperature drop through the scale.

The worst condition possible would be with the initial heat flux and the final scale thickness. It is fairly certain that the scale did not build up to this thickness immediately, but, on the other hand, there is no reason to assume that the buildup is linear with time. Mechanisms could be postulated for a high initial rate with a lowering rate with time or just the opposite, a low initial rate and a higher rate as the scale builds up and the surface temperature increases. Likewise, there is no reason to assume that the thickness measured on the plate as removed from the reactor is the maximum thickness which existed during the irradiation. It is possible that when the scale built up to a certain thickness it sloughed off either entirely or to some intermediate thickness.

A reasonable estimate of the maximum temperature which was reached in the fuel portion of the plate was obtained by using a heat flux of $500,000 \text{ Btu}/(\text{hr})(\text{ft}^2)$, a scale thickness of 7 mils, and a scale conductivity of $0.56 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F})/\text{ft}$. The temperature rise through the scale determined with these values was approximately 500°F . At this heat flux, the temperature rise between the water and the element surface has been calculated to be approximately 70°F . Adding these two values to the 420°F bulk water temperature, we may conclude that the maximum temperature of the fuel could easily have been in the neighborhood of 1000°F .

The X-ray diffraction analysis of a sample of scale taken from the "hot" end of the plate indicated that this oxide was composed of 50% corundum (Table 9). This anhydrous phase of aluminum oxide is formed from the hydrated Bohmite only at temperatures above 720°F . This would indicate that a considerable portion of the scale was subjected to

temperatures at least this high. In order to form such a high percentage of the anhydrous phase, the inner surface would probably have to be considerably above this value.

E. Swelling of Core in High-flux Zone

Figures 16, 17, and 18 illustrate the condition of the fuel portion of the plate at a point approximately one inch from the in-reactor end of the plate. No gross swelling was detected beyond a point approximately two and one-quarter inches from the "hot" end. At the point where this cross section was made, the burnup of U^{235} was approximately 50% of the original 2.2 at-% of total core.

The above discussion of temperatures would indicate that the fuel had been subjected to a temperature as high as 1000°F. It is considered that the combination of these two factors caused the swelling noted.

The burnup at the point beyond which swelling and delamination were not detected, i.e., $2\frac{1}{4}$ in. from the in-reactor end, was approximately 40% of the U^{235} originally present. Unfortunately, due to the uncertainty in calculating fuel temperatures noted above, it is impossible to state with confidence a temperature in this region which could be used as a limit below which swelling would not be expected in elements of this type at this burnup.

VIII. CONCLUSIONS

1. The cladding failed approximately $4\frac{1}{2}$ in. from the in-reactor end of the test plate at two distinct points.
2. The cause of the failure was local corrosion of the cladding alloy. This local pitting was probably the result of attack by loop water of high pH during the ten-day period prior to the detection of the rupture.
3. The failure was not catastrophic, and detection was possible before any large amount of fission products were released.
4. Operation of the loop with the defective element in the high-flux zone for several days after the first penetration of the cladding took place did not cause any permanent high-level contamination of the loop components.
5. Irradiation of this fuel to 58% burnup of the U^{235} content with surface temperatures of from 700 to 900°F and fuel temperatures near 1000°F produced swelling of the fuel alloy.
6. The swelling did not cause failure of the cladding.
7. A heavy oxide scale caused a high fuel temperature, which is considered to be an important factor in producing the swelling which occurred.

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