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NINTH HIGH TEMPERATURE FUEL MEETING

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METALLIC URANIUM IRRADIATION STUDIES (G. A. Last)

I. SUMMARY OF CLUSTER IRRADIATIONS

A. The following rod cluster fuel element irradiations have been conducted:

Number of tests	- 9
Number of elements irradiated	- 44
Number of rods in each element	- 3 to 7
Total number of rods irradiated	- 277
Rod diameters	- 0.63 and 0.593 inch
Zircaloy-2 cladding thickness	- 20 to 30 mils
Exposure	- 250 to 2520 MWD/T
Bulk water outlet temperature	- 185 to 260 C
pH	- 4.5 to 10.0
Maximum uranium temperature (calculated)	- 510(1)

(1) Film formation from low pH water caused the maximum uranium temperature to exceed this value by an unknown amount.

B. A brief summary of the conditions and results of several of these tests are presented in Table I.

Table I

Test	214A	226A	226ASA	226ASB	172A	146A
Maximum uranium temperature, °C	430	430	432	453	625-700	410
U-Zr interface temperature	295	300	297	310	417-480	310
Cladding surface temperature	225	250	254	259	325-360	270
Exposure MWD/T	1500	1200	2240	2520	2200	1250
a/o	0.18	0.15	0.26	0.31	0.28	0.16
fissions/cm^3	0.9×10^{20}	0.72×10^{20}	1.3×10^{20}	1.5×10^{20}	1.35×10^{20}	0.75×10^{20}
Cladding Thickness	0.030"	0.030 0.020	0.020 0.030	0.030	0.030	0.030
External pressure psi	1500	1500	1500	1500	1200	1500
R values clad 0.020") (clad 0.030")	1.7	No dim. Change	0 1.3	1.6	8.6-18.9	2-3
Water pH	10.0	10.0	10.0	10	4.5	9.5
Remarks	No film on clad.	No film on clad.	No film on clad.	No film or clad.	Heavy iron oxide film.	Thin film of iron oxide.

C. 226-A, Supplement B

A charge of six Zircaloy-2 clad seven-rod clusters, two with 30 mil and four with 20 mil cladding, sustained a rupture after an average total exposure of

[redacted]

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2520 MWD/T. The 1.6 percent enriched uranium rods for all clusters were fabricated from rod stock coextruded at Nuclear Metals Incorporated. The failure was in a peripheral rod of one of the 20-mil clad clusters. The cladding of this rod was split longitudinally for approximately 7/8 inches and was similar in appearance to the failure of 226ASA which occurred after 2240 MWD/T exposure. The uranium in the region of the split in the cladding is badly cracked, and may have been the cause of the failure. Figure 1 shows the cluster "as received" and Figure 2 is an "as cut" section through the lower end of the split.

Irradiation and temperature conditions were as follows:

Loop outlet temperature	232 C
Loop pressure	1500 psi
pH	10.0 (LiOH added)
Cladding surface temperature (calculated)	259 C
Uranium core temperature (calculated)	453 C

Dimensional changes on the 0.030" clad rods.

Average diameter increase	0.0029"
Standard deviation	0.0018"

D. 146 A

Examination is complete on the two 7 rod clusters of 0.630" diameter 0.030 inch Zircaloy-2 clad uranium rods. Irradiation conditions and results are in Table I. Extrusion of the uranium into the end cap voids occurred but no fission gas pores could be seen in electron micrographs of the uranium core.

A very thin film deposit could be seen on the cladding after irradiation, but no corrosion attack was evident.

II. SUMMARY OF TUBULAR IRRADIATIONS

A. The following irradiations of tubular elements have been conducted:

Number of tests -	4
Number of elements -	11
Number of tubular components -	19
Number of rod components -	3
Cladding thicknesses-Zr-2	0.030 and 0.020"
Exposure	250 to 2000 MWD/T
	0.15 - 1.21×10^{20} f/cm^3

[redacted]

Bulk water outlet temperature - 245 C
pH - 4.5 to 10.0
Calculated maximum uranium temperature

tubes - 385 C (1)
rods - 650 C (1)

(1) Due to the formation of a film at low pH, this temperature may have been exceeded by as much as 150 C.

B. Tubular Irradiations BF-3 and DoI-2

These two rod-in-tube elements are the only two of tubular geometry that have been irradiated to high exposure. These elements were examined at BMI.

Irradiation conditions:

Bulk water temperature - 245 C

pH - 4.5

Maximum uranium temperatures

Tube	350
Rod	650-670 C

Exposure:

DoT-2 Tube	0.23 a/o	1.1×10^{20} fissions/cm^3
DoT-2 Rod	0.22 a/o	1.06×10^{20}
BF-3 Tube	0.20 a/o	0.96×10^{20}
BF-3 Rod	0.17 a/o	0.82×10^{20}

Dimensional Changes

Length - No change within accuracy of measurements.

	Diameter	Volume
DoT-2 Rod	+ 1.0%	+2.0%
BF-3 Rod	+ 0.73	+1.46%

Density Changes

DoT-2 Tube	-	1.17%
Dot-2 Rod	-	1.43%
BF 3 Tube	-	1.01%
BF 3 Rod	-	0.53%

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R Values	Calculated from Volume Changes	Calculated from Density Changes
DoT 2 Tube		5.1
DoT 2 Rod	9.1	6.5
BF 3 Tube		5.0
BF 3 Rod	8.6	3.1

An iron oxide film was observed on the cladding surface of the components of both elements.

Metallographic examination showed hydride formation in some regions of the cladding surface. Pits were also observed in the cladding, some of which were associated with the hydrides.

C. Low Exposure Irradiation of Tube-in-Tube Elements

Two tests of tube-in-tube elements were discharged at low exposures, 500 MWD/T, because of rupture indications in the coolant systems. One test was in low and the others in high pH water.

1. Low pH RM-553

Three elements were irradiated under the following conditions:

Coolant Water Temperature	280 C
Calculated Maximum Fuel Temperature	385 C
pH	4.5

Nominal dimensions were:

Inner Tube		Outer Tube		Length
ID	OD	ID	OD	
0.500	1.050	1.450	1.820	36.0

Metallographic examination showed the elements to be heavily filmed with iron oxide and a more or less complete destruction of the autoclave film had occurred. Failure of the clad on the inner bore of one of the inner tubes resulted from accelerated pitting corrosion of the clad. Metallographic examination near the failure showed extensive, uniform hydriding of the clad. The hydrogen content of the inner clad is estimated to be 500 ppm and of the outer to be 200 ppm.

2. High pH RM-550

The second test discharged consisted of two unalloyed and two U-2 w/o Zircaloy alloy tube-in-tube elements. They were irradiated in water at: 240 C
pH 10.0

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The point of rupture has not been found. The fuel element surfaces were free of deposited film with no indication of corrosion of the clad.

Bumping was found on the unalloyed components but none on the alloy element. Large grains resulting from a vacuum beta heat treatment is believed to be the primary cause of the bumping and severe warping of the unalloyed elements.

Radial cracks about 0.001 inch wide were found in the outer uranium tube.

III. SUMMARY OF MTR-ETR IRRADIATIONS ON NaK CAPSULES

Five fuel rods in the Hanford ETR-MTR NaK capsule irradiation program have been irradiated and the examination completed. The fuel rods were natural-unalloyed uranium coextrusion clad with 0.030 inch Zircaloy-2. Outside diameter of the cladding was 0.630 inches. Dimensional changes and exposures are presented in Table II. Exposures were calculated from the center uranium temperatures and the power generation in the uranium.

A volume average temperature range was calculated based on the range of core temperatures and the range of the uranium-clad interface temperatures. The range of center core temperatures are shown in Table III.

TABLE II

Irradiation Number	Average ΔD inches	% ΔD	% ΔA	Exposure a/o B.U. f/cm^3	R
3-31	0.0026	0.412	0.82	0.239 1.25×10^{20}	3.45
3-32	0.0045	0.714	1.43	0.415 2.0×10^{20}	3.44
3-57	0.0087	1.38	2.76	0.19 0.91×10^{20}	14.5
3-58	0.0301	4.78	9.56	0.228 1.1×10^{20}	41.9
3-59	0.039	6.19	12.76	0.26 1.25×10	49.1

[redacted]

TABLE III

Range of Irradiation Temperatures

Irradiation Number	Center Core Temperature C°	Volume Average Temperature C°
GEH 3-31	400 - 430	342 - 365
3-32	400 - 525	342 - 444
3-57	475 - 565	394 - 478
3-58	550 - 770	465 - 649
3-59	750-950	632 - 800

The data in Tables II and III are plotted in Figure 3. GEH 3-32 had a split failure of the cladding during the irradiation and GEH 3-59 had an end cap failure during the irradiation. It is felt therefore that diameter changes from these two irradiations hsould not be weighted equally, if at all, with the data from the other irradiations. In the temperature range of 400-450 C the uranium volume begins to increase rapidly with increasing metal temperature.

IV. EFFECTS OF pH ON FILM FORMATION

Irradiation testing in high pH water (pH = 10.0) has been generally successful from the standpoint of cladding and fuel performance. No evidence of general corrosion or film formation has been observed.

Heavy film formation has occurred during the irradiation of both rod cluster and tubular elements in low pH water (pH = 4.5). Although no cladding failure occurred in cluster elements, excessive uranium temperatures resulted from film formation (172A). Accelerated pitting corrosion, almost complete destruction of the autoclave film, and failure of the cladding resulted from the film formation in tubular test RM-553.

V. FUEL PERFORMANCE

The data from the high pH cluster tests show that satisfactory fuel behavior can be expected for fuel temperatures up to 450 C and exposures of 2500 MWD/T. The data presented from the low pH cluster test 172A and tubular tests BF-3 and DoT-2 indicate that poor metal behavior can be expected at somewhat higher central metal temperatures.

VI. EFFECT OF PRESSURE ON SWELLING

The possible importance of externally applied pressure on uranium swelling is indicated in the comparison of the results from pressurized loop irradiations and the swelling capsules. The range of uranium volume average temperatures and R values for cluster element and the rods from the tube and rod element are shown along with the NaK capsule results in Figure 3.

[redacted]

Data from the density decreases of unalloyed uranium irradiated in the SRF by Atomics International indicate that swelling is somewhat greater than clad rods irradiated at comparable temperatures.

This could be an indication of the amount of external restraint offered by the Zircaloy-2 cladding of the rods used in the NaK capsule tests.

VII. ETR RUPTURE TESTS

A series of three planned rupture tests has been completed at the ETR 3X3 pressurized loop facility. In this series of tests, a hydraulically operated chisel was used to shear off a cap which had been welded over a (25 mil) diameter hole drilled through the cladding of one rod of a seven-rod cluster. The chisel was operated from outside the reactor so that the defect in the cladding could be uncovered at the time desired.

Rupture 1

In the first test of the series, a previously unirradiated seven-rod cluster of 0.020 inch Zircaloy-2 clad uranium was used. After the reactor reached full power, the defect cap was sheared off. At this time an increase of 40 mr of gamma activity was observed in the loop water. The reactor was operated for an additional seven hours with no further increase in gamma activity. The reactor was then shut down the defected element removed and returned to Hanford for examination. The defected rod was sectioned through the defect and polished. A small lens-shaped pocket of oxide, approximately 40 mils wide by 12 mils deep had formed under the defect. An estimated 1.5 milligrams of uranium were converted to oxide. The oxide pocket can be seen in both the macro of the section through the defect (Figure 4) and the micro of the defect itself (Figure 5).

In tests performed in high temperature, out-of-reactor, recirculating loops in the same manner as the in-reactor test and using similar material, failure occurred in two hours or less after the defect was opened to the loop water. As can be seen in Figures 4 and 5, failure, in the sense of sufficient oxide formation to split open the cladding, has not occurred.

Irradiation and temperature conditions were:

Loop outlet temperature	258 C
Loop pressure	2000 psia
Cladding surface temperature (calculated)	286 C
Uranium core temperature (calculated)	501 C

Rupture 2

The second test of the series was similar to the first, except that the cladding was 30 mils and the cluster had been irradiated to 2400 MWD/T in 25 C, 200 psi, pH 7 water. During the rupture test the reactor was operated for three days to build up short half-life fission products and then the defect cap was sheared off. After about two hours, a seven mr burst of gamma activity was observed and then no further activity was noted. Fourteen hours after the defect cap was sheared off the reactor was shut down and the defected element removed. During this 14-hour period, the reactor was cycled several times from zero power to full power. The reactor was cycled in the hope that the thermal shock to the fuel would assist in causing the defected rod to fail.

The defected rod was sectioned through the defect and a spherical pocket of oxide approximately 60 mils in diameter was observed under the defect. An estimated 5 milligrams of uranium were converted to oxide. Cracks were seen in the uranium, both the section through the defect and in a section taken approximately midway between the ends of the rod. Figure 6 is a cross section through the defect showing the oxide pocket and macrocracks in the uranium. Figure 7 is a photomicrograph of the area of the defect showing cracks radiating from the oxide pocket.

Temperature and coolant conditions during irradiation at the ETR were:

Outlet temperature	287 C
Cladding surface temperature (calculated)	323 C
Core temperature (calculated)	633 C to 800 C

Rupture 3

The third test was identical to the second in all respects except that 33 minutes after the defect cap was sheared off, the loop water activity was so great that a reactor shutdown was required. Figures 8 and 9 show the extent of the failure. An estimated 53 grams of uranium were converted to oxide in this test. Figure 10 is a cross section through the failure. Extensive cracking of the uranium has occurred and two pieces of completely unbonded uranium remained in the rupture area at the time the test was terminated.

BASIC SWELLING STUDIES - R. D. Leggett

A. Irradiation Program

An instrumented general swelling capsule containing uranium spheres one centimeter in diameter was charged into a front-to-rear test hole in a Hanford Reactor in the latter part of February. This capsule is operating satisfactorily at a temperature of 550 C ± 5 C which is precisely

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maintained when the reactor is shut down. An expected 50 C temperature gradient exists between the center of the sphere and the surface.

B. Pore Site and Distribution in Irradiated Uranium

The statistical study of the pores in irradiated and post-irradiation annealed specimens of uranium is continuing.

Synthetic pores have been prepared by dispersing polystyrene spheres with diameters of 1.17, 0.81, and 0.56 microns respectively in plastic, and after microtoming selectively dissolving the spheres and replicating the surface. Statistical analysis of the results of studying the replicas in the electron microscope revealed that those pores cut at their centers suffered distortion on replication. Instead of a sphere a shape results that is almost cylindrical with a dome on the end.

[redacted]

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Figure 1

A Seven-Rod Cluster Fuel Element Which Failed After 2520 MWD/T Exposure in a High Temperature Recirculating, Water-Cooled Loop. ~0.6X

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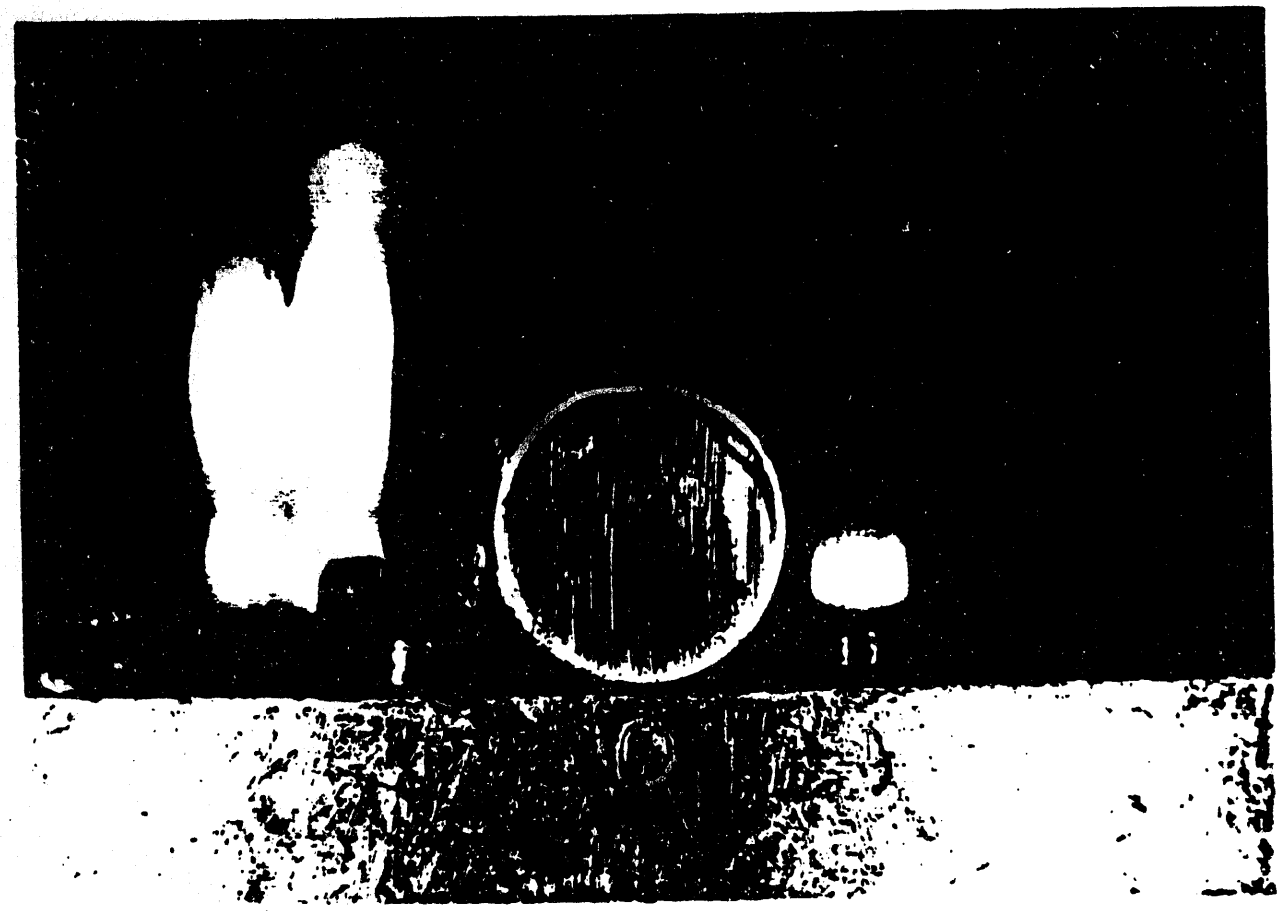

Figure 2

A Section Through the Lower Edge of the Split Visible in Figure 1. Note That the Uranium Appears to Have Corroded More Rapidly on Either Side of the Split Than Directly Under it.

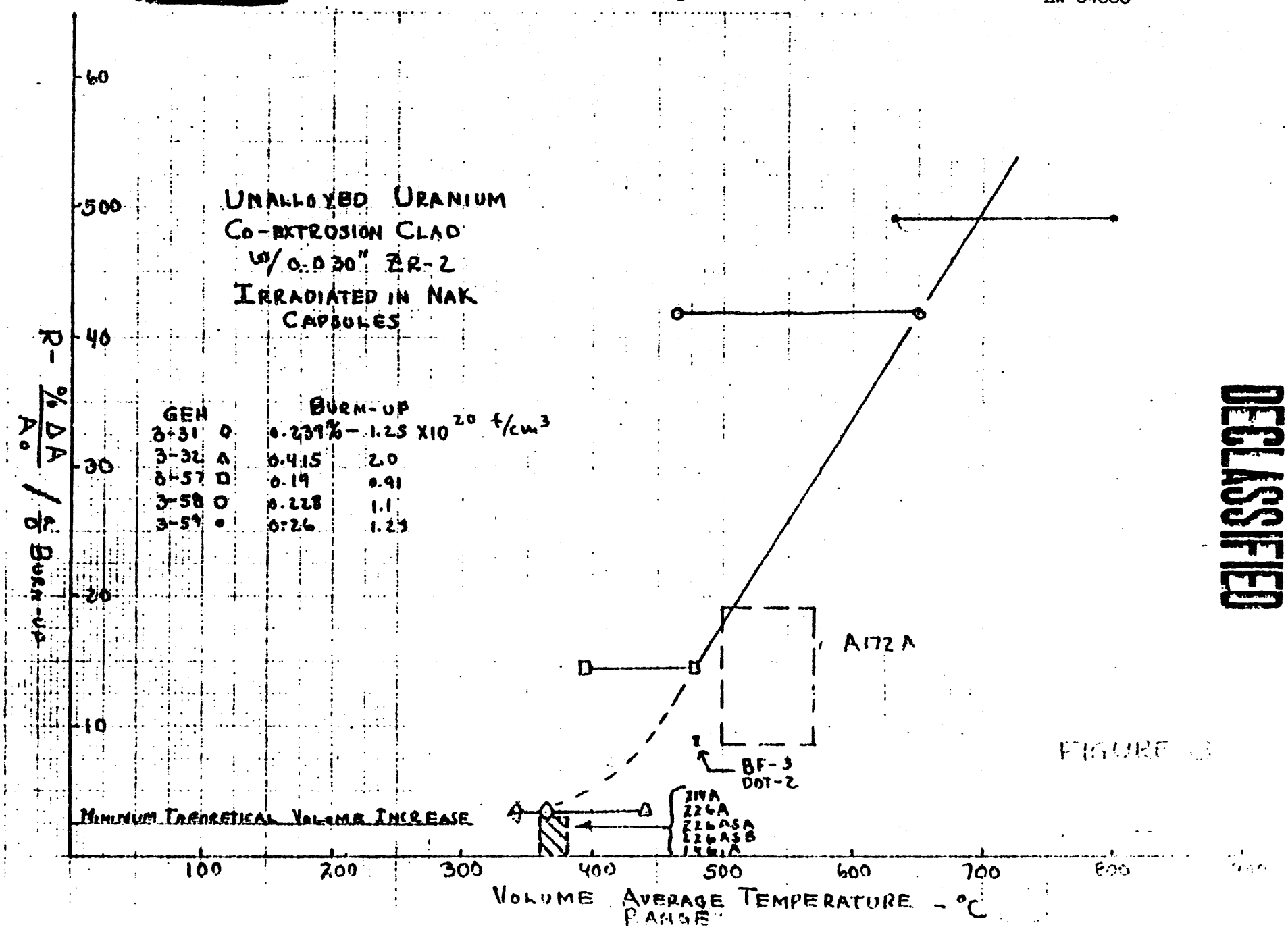

FIGURE 3

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Figure 4

A Section Through the Defect of the Ruptured Rod of the First Rupture Test. The Reactor Operated About Seven Hours After the Defect Was Opened Up. Note the Lens-Shaped Pocket of Oxide Under the Defect. ~1.5X

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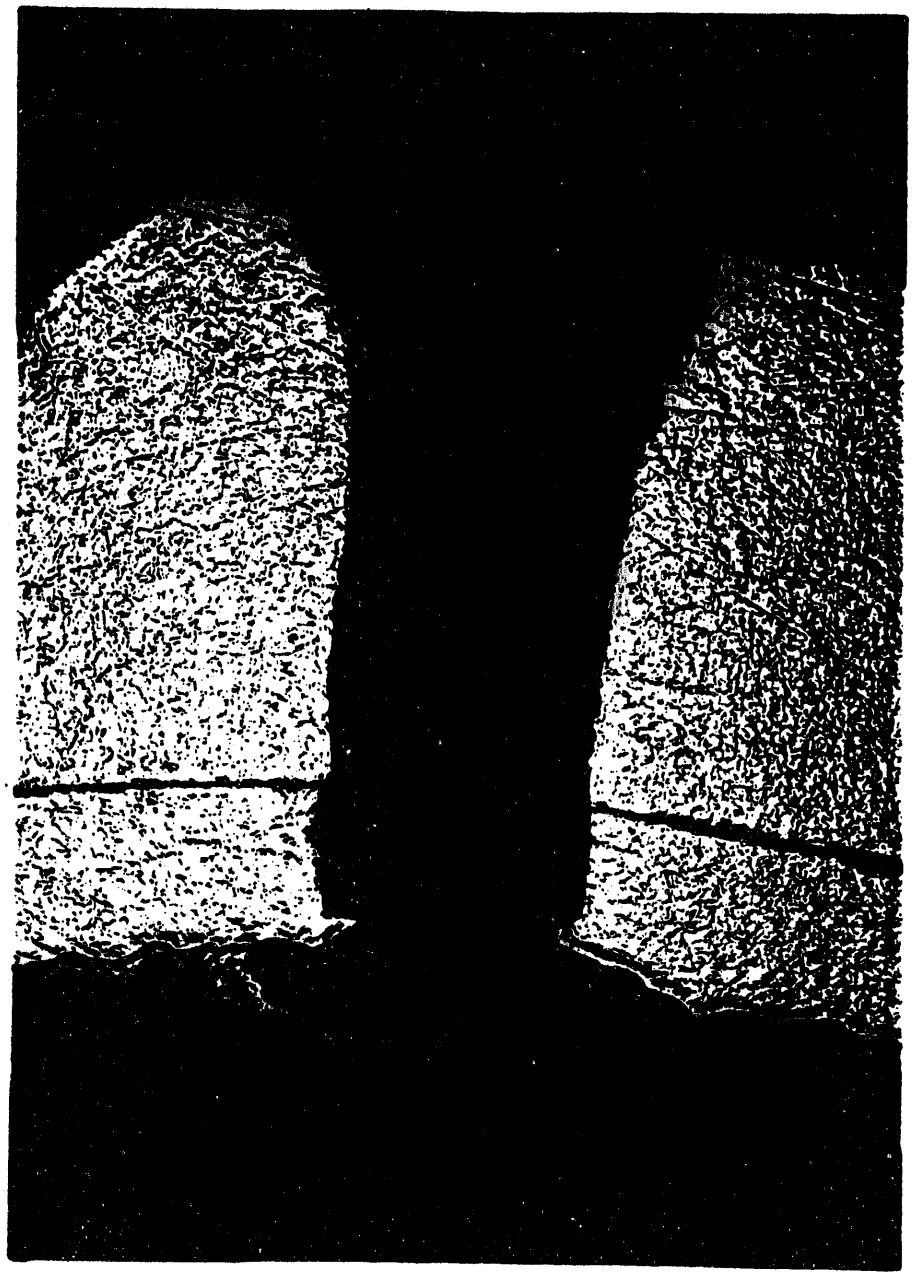

Figure 5

A Cross Section Through the Defect Hole of the First Rupture Test. Cathodic Etch 50X

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Figure 6

A Rough-Polished Cross Section Through the Defect of the Second Rupture Test. ~ 4-1/2X

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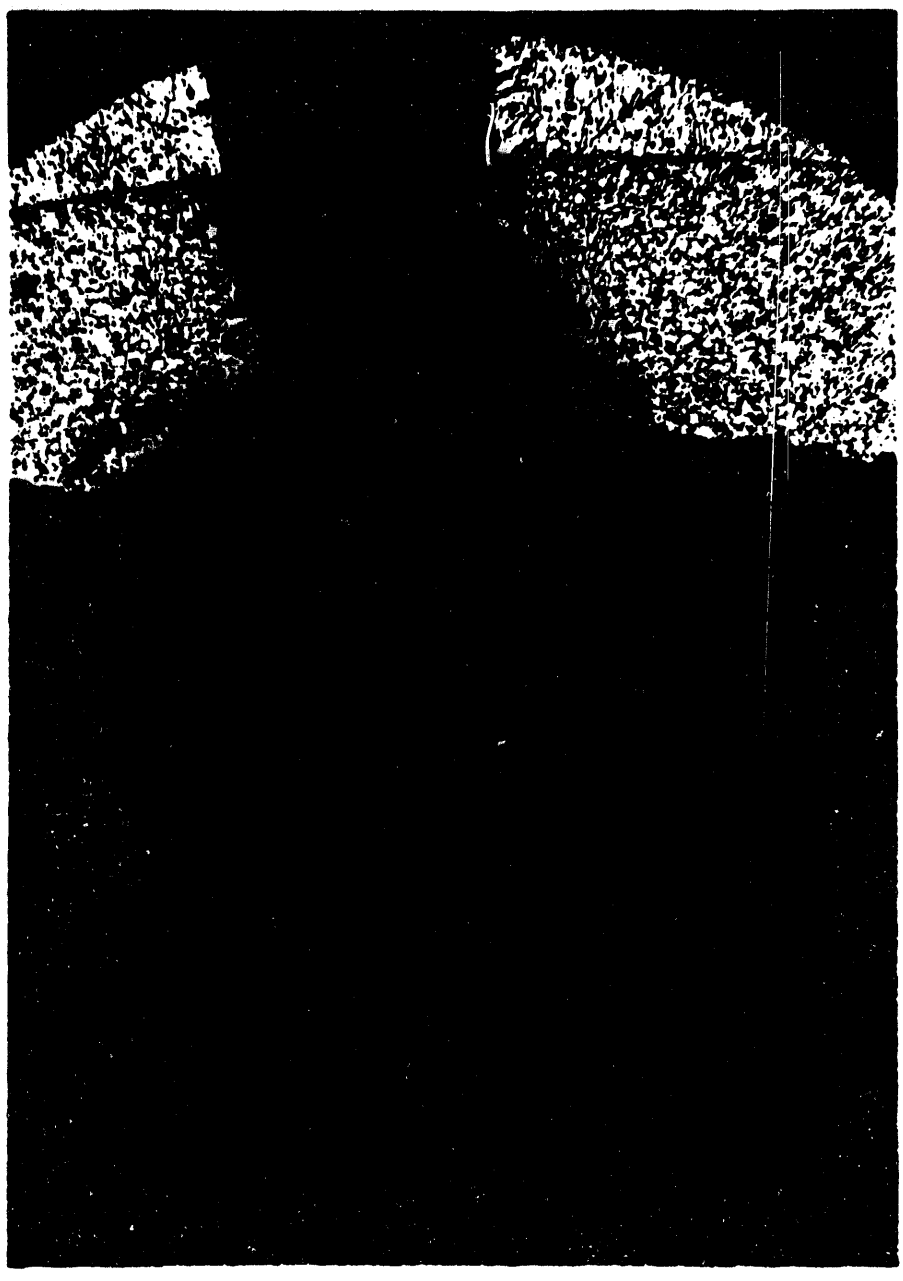

Figure 7

A Photomicrograph of the Area of the Defect Hole in the Cross Section Through the Defect of the Second Rupture. 50 X

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Figure 8

The Cluster Used in the Third Rupture Test (as received). Note the Large Amount of Swelling of the Defected Rod and the Splits Visible in the Cladding. ~1.3X

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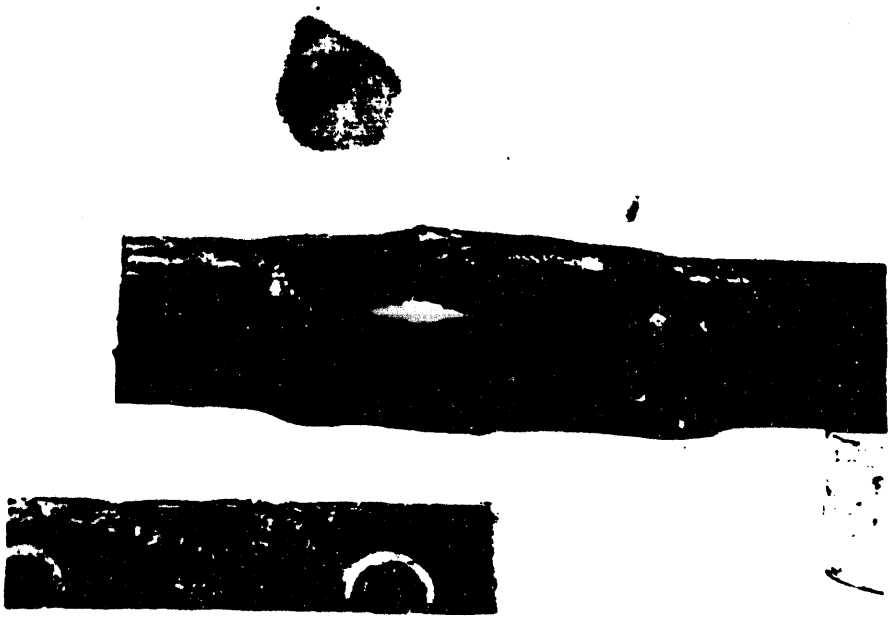

Figure 9

The Defected Rod of the Third Rupture Test After it Was Cut From the Cluster. As can be Seen, the Uranium is Completely Gone From Inside the Cladding in the Swollen Area Except for Some Loose Pieces, One of Which is Visible Through the Split in the Cladding. ~1.5X

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Figure 10

Cross Section Through the Defected Rod of the Third Rupture Test. This Cross Section Was Made Through the Loose Piece of Uranium Which Can be Seen in Figure 9 Through the Split in the Cladding.
~2.5X

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