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THE EFFECTS OF REACTOR IRRADIATION ON THORIUM-URANIUM ALLOY FUEL PLATES

R. M. Carroll

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THE EFFECTS OF REACTOR IRRADIATION ON THORIUM-URANIUM ALLOY FUEL PLATES

By

R. M. Carroll

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Abstract

Several plates of 98.7% Th - 1.2% U 235 (clad in aluminum) were irradiated in the MTR for an integrated flux of 2.6×10^{21} neutrons/cm^2. Although these samples represent an early development in bonding of aluminum to thorium and there are better methods at present, the bond proved to be quite strong and both clad and core were dimensionally stable under irradiation. The production of uranium 233 was as much as theory would indicate and the total amount of fissionable material after irradiation and after decay of the protactinium 233 was greater than before irradiation. A fuel element of this nature appears to offer excellent potentialities from the standpoint of radiation stability.

Introduction

In October 1952, an experiment was initiated by W. M. Breazeale[1] to determine the dimensional stability and change in physical properties of a thorium-uranium fuel plate when approximately 1% of the total number of atoms in the alloy are fissioned.

A fuel element composed of an alloy of thorium and enriched uranium in the right proportions would have a longer fissionable life than an element containing only enriched uranium. The increased life would be due to the formation of uranium 233 from thorium 232, thus maintaining the concentration of fissionable material at a high level. The fuel element life would be limited to either the physical stability of the element or the poisoning due to fission products.

Thorium-uranium plates, clad with 52S aluminum, were rolled at 500°C using a 1.3% Si-Al barrier material. At this temperature and with this barrier material there was no intermetallic compound formation between the thorium-uranium and the aluminum cladding. Four 8 x 1 x 0.072 inch and one 28 x 1 x 0.072 inch plates of 98.7% Th-1.2% U 235, 0.1% other uranium isotopes were fabricated.[2] The fuel elements were made by the ORNL Metallurgy Division with the best available means at that time.

The irradiation experiment was designed by F. C. Zapp.[3] The 28-inch plate and three of the 8-inch plates were placed in the Materials Testing Reactor in April 1953 and removed in July 1953. The remaining 8 inch plate was retained as a control. The total irradiation of these plates were 4153 Mw-days and an estimated 2.6×10^{21} neutrons/cm^2 (approximately 1% burn-up).[4] During irradiation the sample plates were cooled by water flowing past the plates at a velocity of approximately 30 ft/sec.

At the end of the experiment the samples were discharged from the reactor and allowed to cool for forty-five days. The samples were then returned to the Solid State Division of ORNL for a determination of the extent of radiation damage.

Physical Measurements

Sample Appearance

The only visible change in the samples after irradiation was that the outline of the core was clearly visible, (possibly because of selective corrosion) and there was evidence of water erosion in the longitudinal surface defects. There did not appear to be any blistering. (See fig. #1)

Self Heating

At the time the samples were removed from the carrier and placed in the hot cell there was some concern about the amount of heating due to self absorption of gamma and beta radiation. In view of the possibility of the clad or core being damaged by self heating, the samples were placed under water until measurements could be made of the amount of self heating.

The first test of self heating was made by placing the samples in an insulated container and measuring the equilibrium temperatures. The sample in the highest flux, #1, reached an equilibrium at 64°C. The second most active sample, #2, attained 59°C, and the least active sample, #4, 51°C. Crude tests with an improvised calorimeter indicated the samples were emitting approximately 18 cal/min. Since the samples had a large proportion of surface area to volume, this amount of heating was not harmful.

Dimensional Measurements

The dimensions of these specimens were measured with a remotely controlled measuring apparatus. (See Fig. #2). The specimens were compared with a standard by means of a dial gauge, accurate to 0.0005 inch. One of the limitations of the

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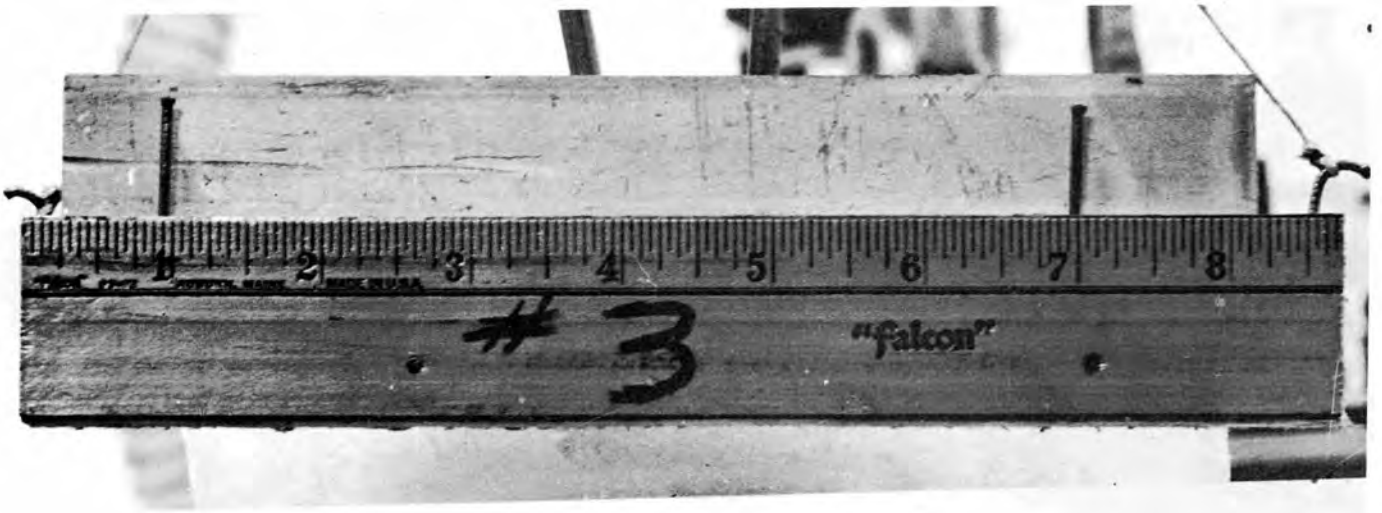

Fig. 1. Typical Appearance of Irradiated Samples. Note the visible outline of the core and the longitudinal errosion marks.

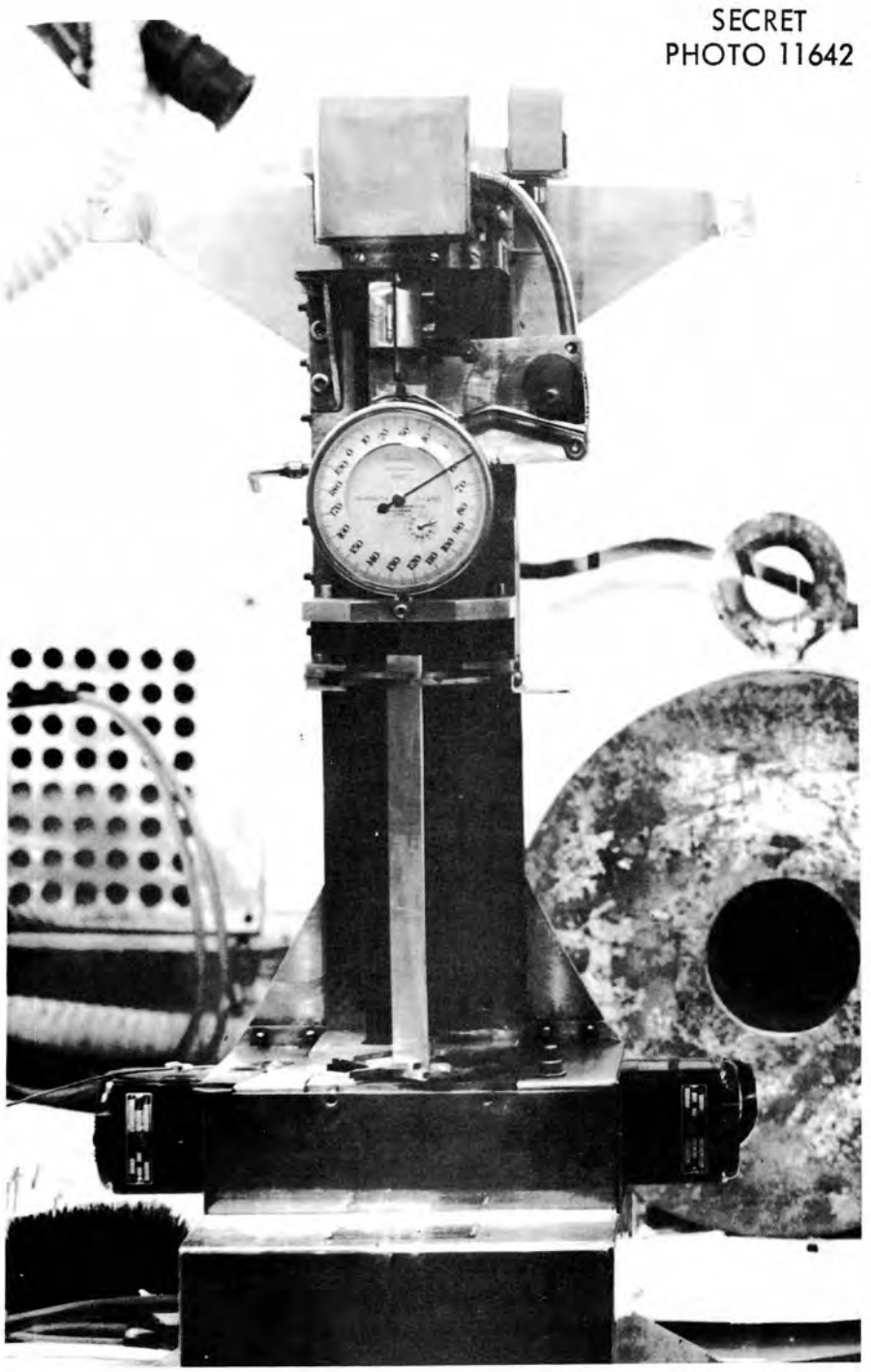

Fig. 11. Remotely Operated Length Measuring Apparatus.

remote dimensional apparatus is that the length, width, and thickness have to be measured on the longitudinal center line of the rectangular samples. Width and thickness measurements were made at numerous positions along the length of the sample and the results were averaged and the maximum deviation from the average noted. The measurements are presented in Table I.

TABLE I

	Width (in.)		Maximum	Length (in.)	
	Preirrad.**	Postirrad.	Deviation	Preirrad.**	Postirrad.
#1	1.000	1.0015	0.0074	8.001	7.999
#2	1.000	1.0003	0.0019	8.002	* 7.992
#3	1.000	1.0003	0.0025	8.002	8.003
#4	1.000	1.0017	0.0081	8.006	8.003

	Thickness (in.)		Maximum
	Preirrad.**	Postirrad.	Deviation
#1	0.0725	0.0760	0.0072
#2	0.0725	* 0.0706	0.0011
#3	0.0725	0.0759	0.0063
#4	0.0725	0.0774	0.0022

** See Text
* Sample #2 was not irradiated and is used as a control

The preirradiation measurements were not made by the group that made the post-irradiation measurements, and unfortunately the precision of these data is not known. The pre-irradiation dimensional measurements are of such uniformity as to suggest they were obtained from the rolls and shears used in the fabrication of the fuel elements rather than individual sample measurments.

One complication of the post irradiation dimensional measurements was the self heating of the specimens. When the samples were placed in the dimensional apparatus the dial gauges indicated a visible creep from the initial position as the samples expanded due to the increased temperatures of the specimens. This was particularly noticeable in the length measurements. The measurements were finally taken at the equilibrium temperature. While this temperature was unknown, it was less than 65°C. The 28 inch specimen was too large for the equipment and no dimensional measurements were made.

It must be emphasized that the deviations in the measurements are primarily due to irregularities of the samples rather than lack of accuracy of the remote measuring apparatus. The unirradiated sample #2 was measured both in the hot cell and on a sensitive micrometer. The irradiated plates appear thicker than the control sample although lack of pre-irradiation measurements under the conditions of remote measuring prevent any definite conclusions. The length and width measurements have indicated no significant changes.

An X-ray examination had been made of the cores of the clad samples before irradiation. (See Fig. #3) After irradiation the samples were so radioactive that autoradiographs on conventional films was impractical. The intensity of the radiation from the samples would darken glass and plastic after several hours exposure near the samples and so autoradiographs of the samples were obtained by laying the samples on a sheet of lucite. The resolution of the resulting autoradiograph was somewhat better than indicated from the photograph (Fig. 4) of the Lucite sheet (the photograph has some reflection from the back of the Lucite). No core dimensional changes could be detected within 0.01 inch, the limits of resolution of the x-rays.

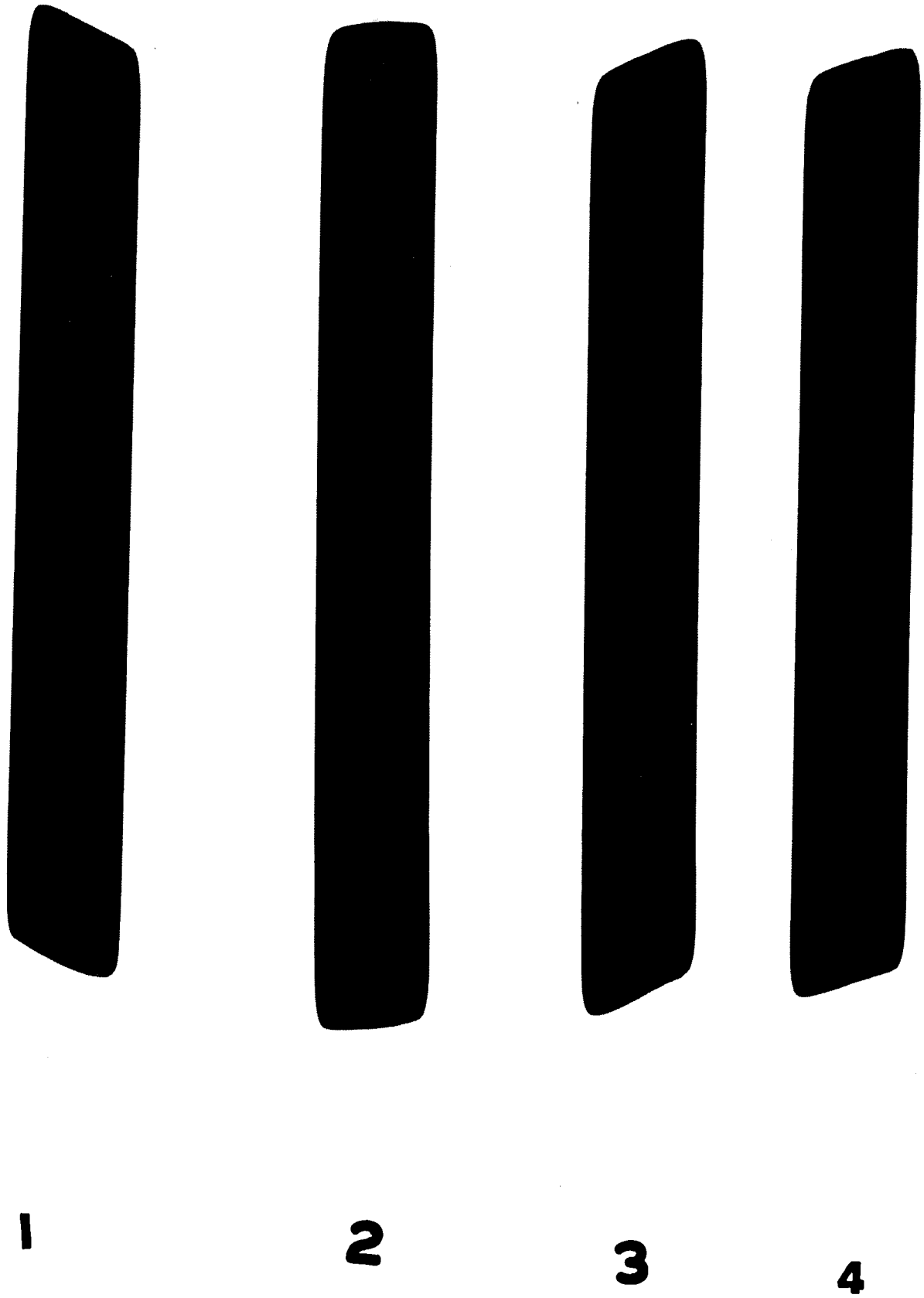

Fig. III. Radiograph of Clad Samples Before Irradiation. Sample #2 was not irradiated.

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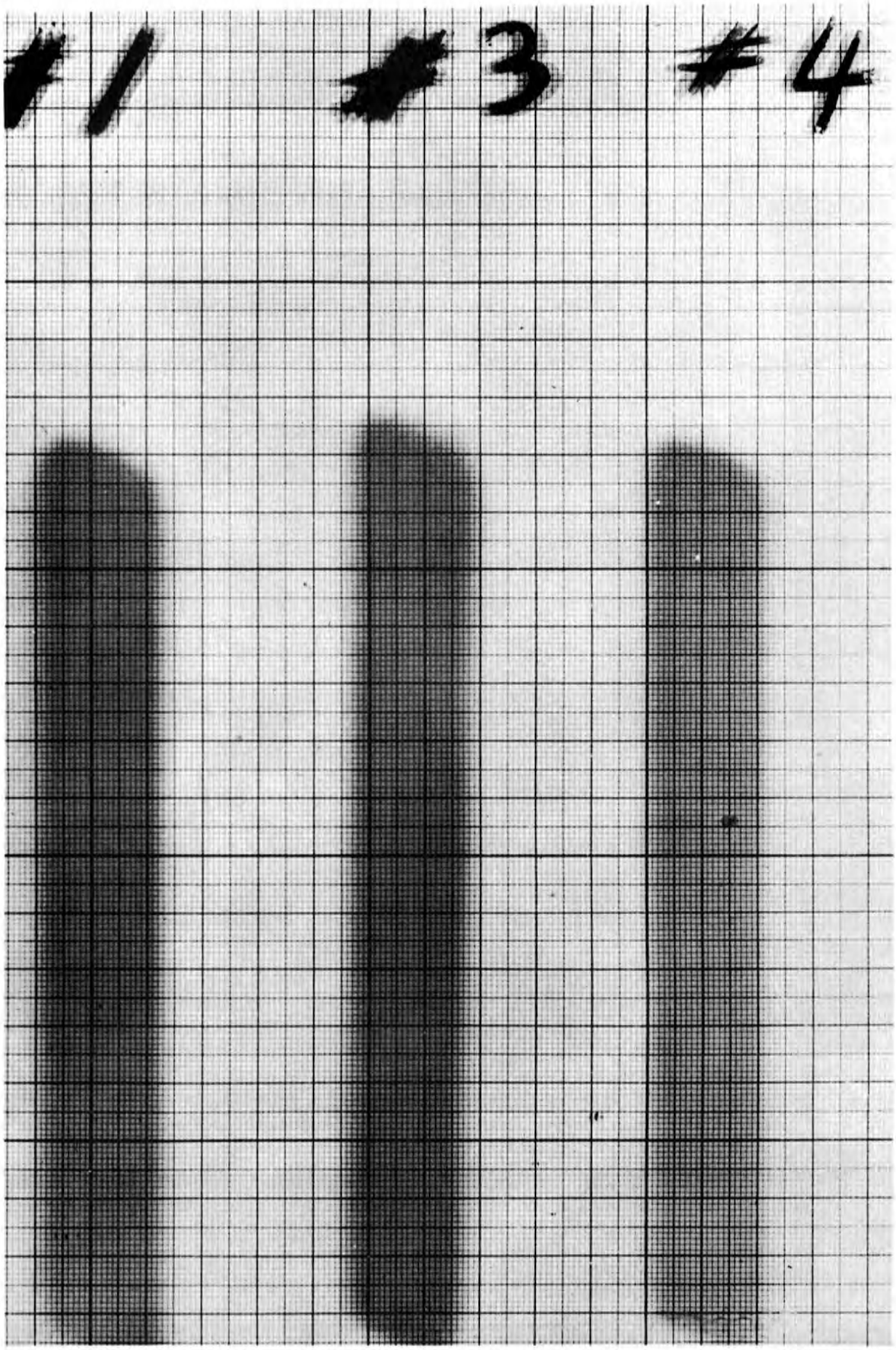

Figure IV. Autoradiograph Samples on Lucite

Density and Weight

The densities of the samples were determined by hydrostatic weighing in a hot cell. The balance was operated with master slave manipulators, and a remotely operated platform built into the case of the balance. The platform was raised and lowered with a small electric motor so that a container of liquid could be raised to immerse the sample. A special hanger was constructed to hold the samples in a horizontal edgewise position so as to minimize the possibility of trapping bubbles. The hanger was connected to the balance with 0.002 inch diameter wires to minimize the effect of surface tension and the need for accurate positioning. The liquid used for the density determinations was carbon-tetrachloride.

The density of the unirradiated sample (#2) was determined outside the hot cell and this sample was used as a standard in the measurements on the others. The density of the carbon-tetrachloride was determined both by the measured temperature of the liquid and by comparison with the known density standard. A typical measurement would involve weighing the standard, the sample, and then again the standard. This method not only determines the varying liquid density due to temperature changes, but also detects zero shifts of the balance. These effects may be important in a non-temperature controlled cell, where the balance may be handled comparatively roughly.

After the densities of the sample were determined the aluminum clad was chemically stripped from the thorium-uranium core with a solution of NaOH and $NaNO_3$. After stripping, (see Fig. 5), the core was thoroughly washed to remove the solution and clinging particles**. The densities of the stripped cores were then determined in the same manner as before, with the exception that a nickel standard was used instead of the unirradiated sample (#2). The densities given are the average of at least four measurements and are presented below in Table II.

** See appendix for contamination problems.

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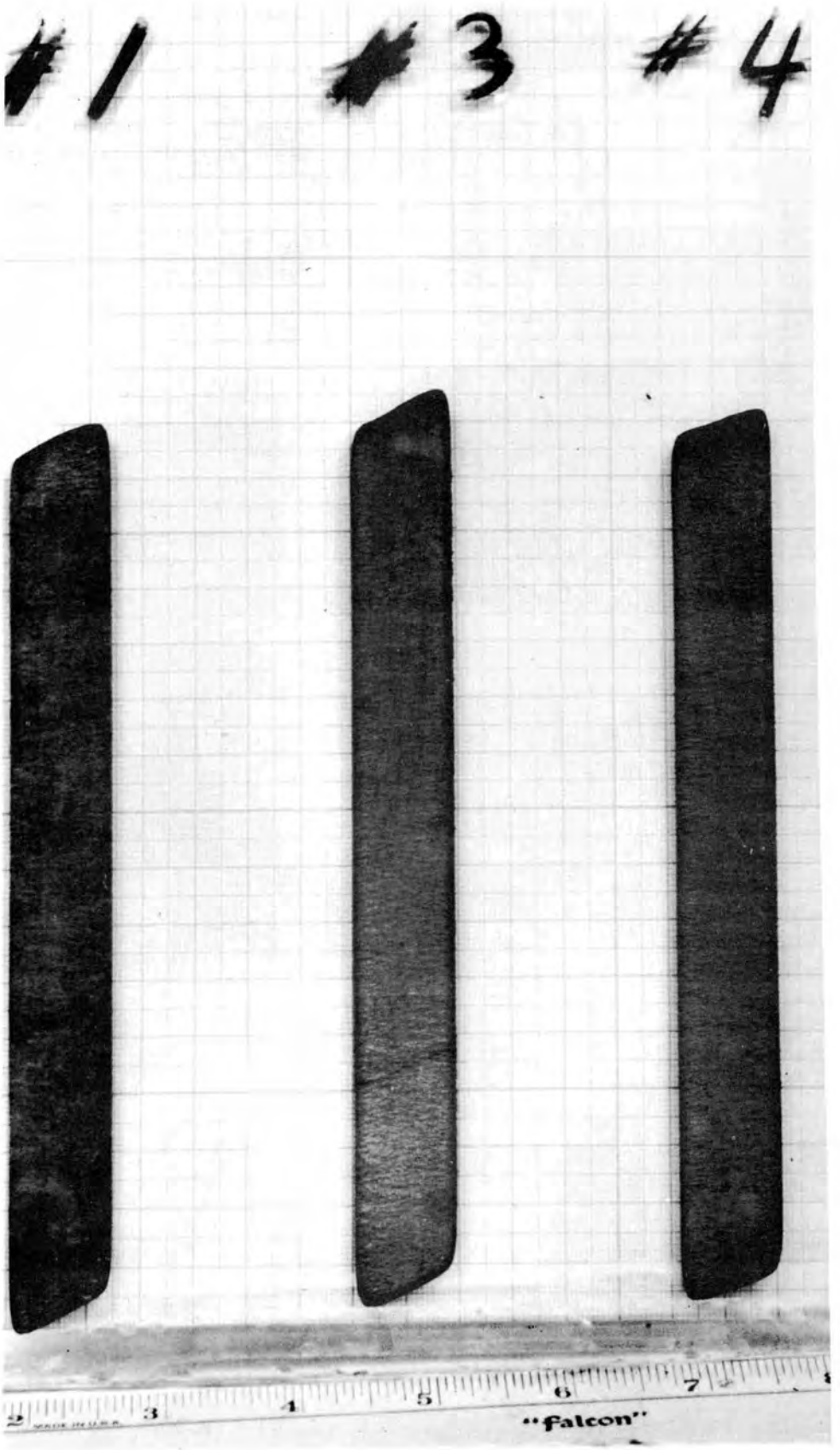

Figure V. Cores of Fuel Elements After Stripping.

TABLE II

Density of Clad Specimens at 30°C		Max. Deviation	Density of Core at 30°C	Max. Deviation
#1	4.4231	0.0001	11.315	0.009
* #2	4.5368	* 0.0005	* 11.538	0.007
#3	4.4330	0.0002	11.336	0.008
#4	4.4233	0.0005	11.314	0.010

* Not Irradiated

The core weight before rolling is compared with the irradiated stripped core in Table III.

TABLE III

Preirradiation Core Weight (1)		Postirradiation Stripped Core Weight
#1	22.2229	22.2555
#2	22.2895 *	Sample sheared before stripping
#3	22.1020	22.1325
#4	22.1578	22.2053

* Not Irradiated

The heavier postirradiated core may be due to an incomplete stripping of the Si-Al barrier. If this is assumed to be the case, then the volume of Si-Al necessary for the additional postirradiated core weight will account for the approximate 2% change in core density. Standard sample #2 was sheared for metallographic examination before stripping so no data is available for the stripped core weight. Sample

#2 was stripped outside the hot cell, so the higher density may be because of more complete stripping. The densities in the clad condition, however, indicated that the irradiated samples were approximately 2% less dense than the unirradiated control. The heavier core of nonirradiated sample #2 will account for the higher average density in the clad condition.

Bond Strength and Hardness

In order to test the bond strength of the clad, a method was developed for twisting samples that were sheared from the 28-inch plate. (See Fig. 6). One end of the sample is fixed while the other end is rotated alternately 45° in either direction about the major axis of the sample. The distance between the fixed and rotated ends of the sample was 4 inches so that this was the equivalent of a 90° twist for the standard 8-inch specimen. After 12 - 16 twists there was evidence of damage which reached a maximum at 25 - 30 twists so that further bends had no apparent effect. The twist were made at a rate of 11 seconds per cycle so that local heating would be dissipated. In no case did the clad break loose or fail in any gross manner. A second test of bond strength was a bend test in which a sample of the 28-inch plate was bent, one time only, 180° about a 0.1-inch radius. The clad of the sample fractured at the point of greatest tensile stress (See Fig. 10). This sample along with samples of unbent irradiated and unirradiated plates were given metallographic and hardness analysis. The summary[5] of the analysis indicated that (1) no definite evidence of an intermetallic phase near the clad-core interface was observed, (Fig. VII and VIII), (2) the bent sample showed separation of the core from the barrier layer for a distance of approximately 1/2 inch and fracture of the core and one side of the clad at the point of maximum bending, (3) comparison of the irradiated sample with a nonirradiated sample showed a drastic increase in hardness of the core and a slight increase in hardness of the clad after irradiation as shown in Table IV.

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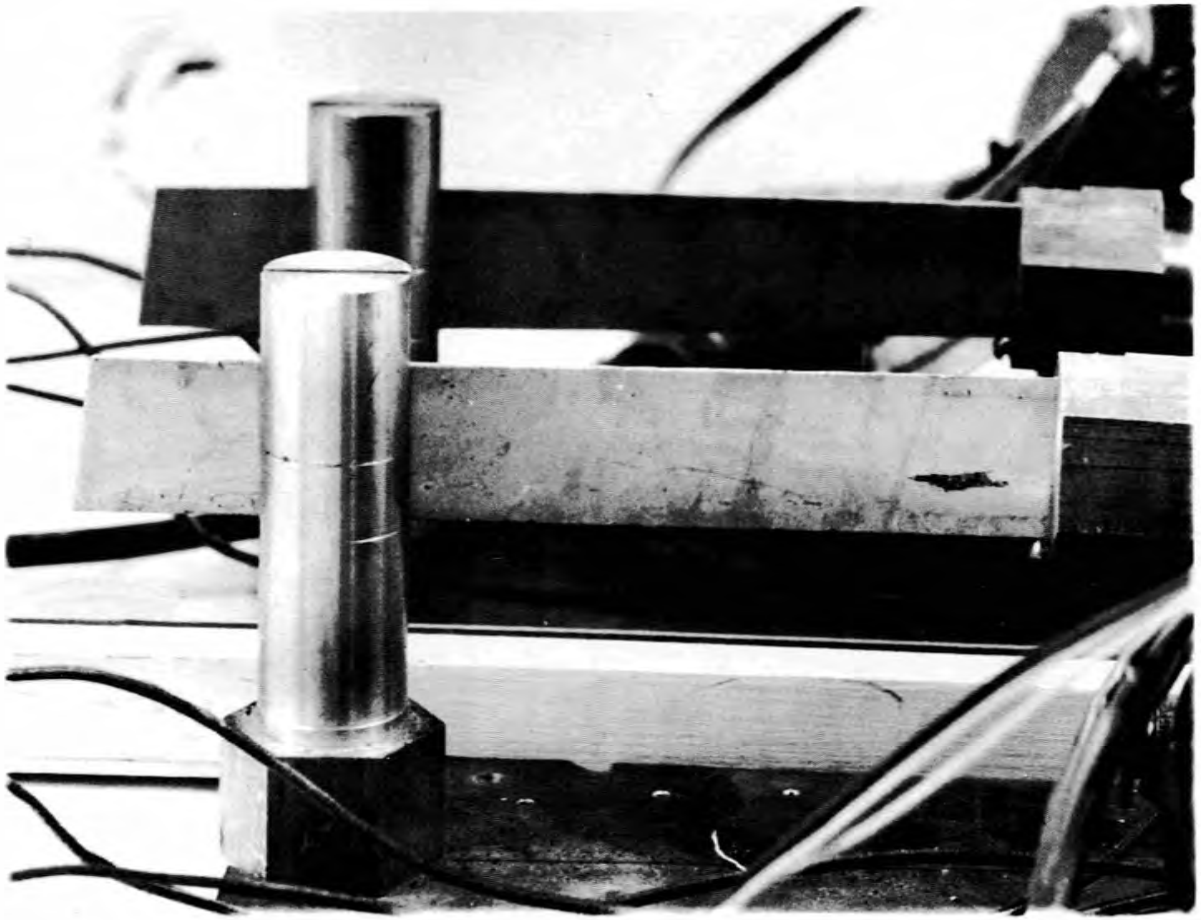

Figure VI. Bond Strength Testing Apparatus. Mirror in background to show reverse side of section of fuel element under test.

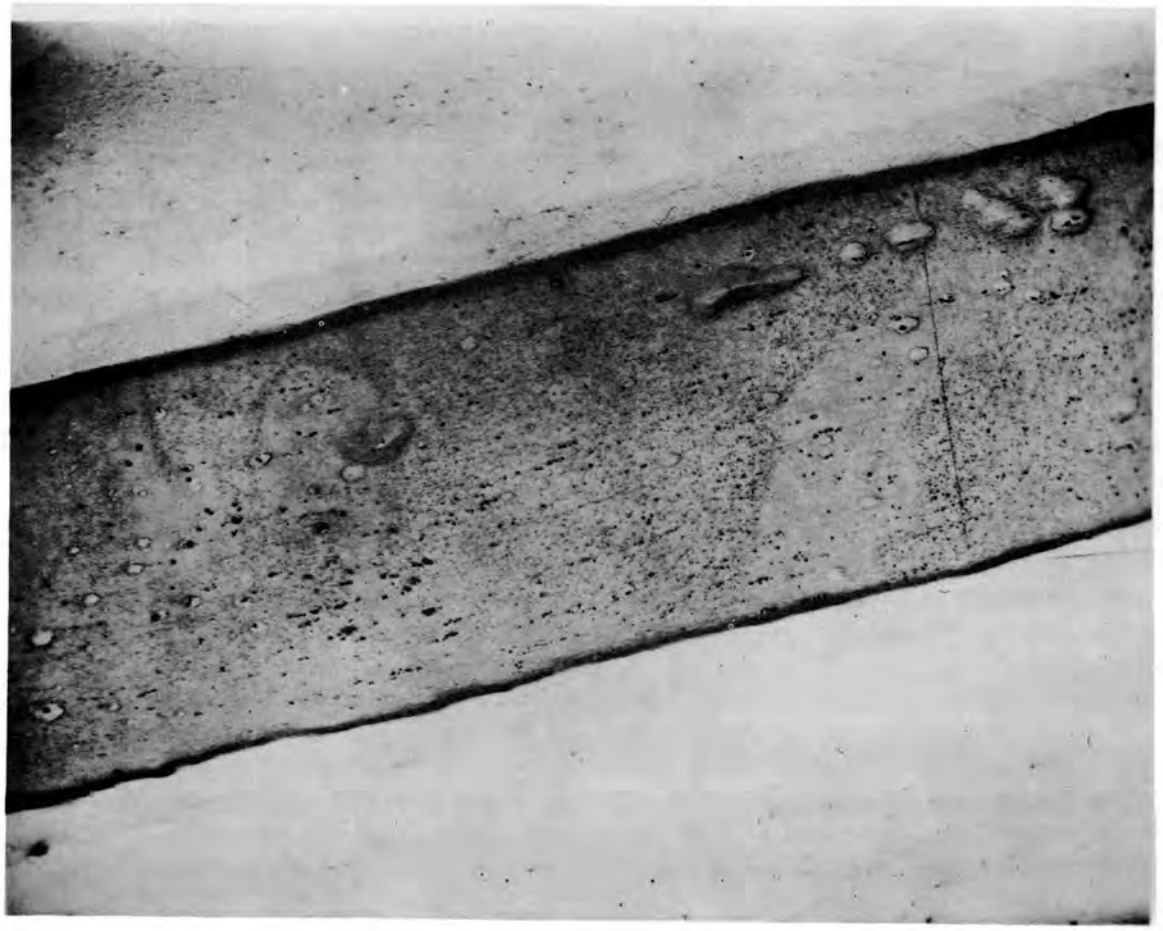

Figure VII. Photomicrograph of Transverse Section of Irradiated Fuel Element. 100X Magnification. This illustrates the typical appearance of an area near the center of the transverse section. The darkening of the core near the core-barrier interface is due to rounding of the edges during the polishing operation.(4)

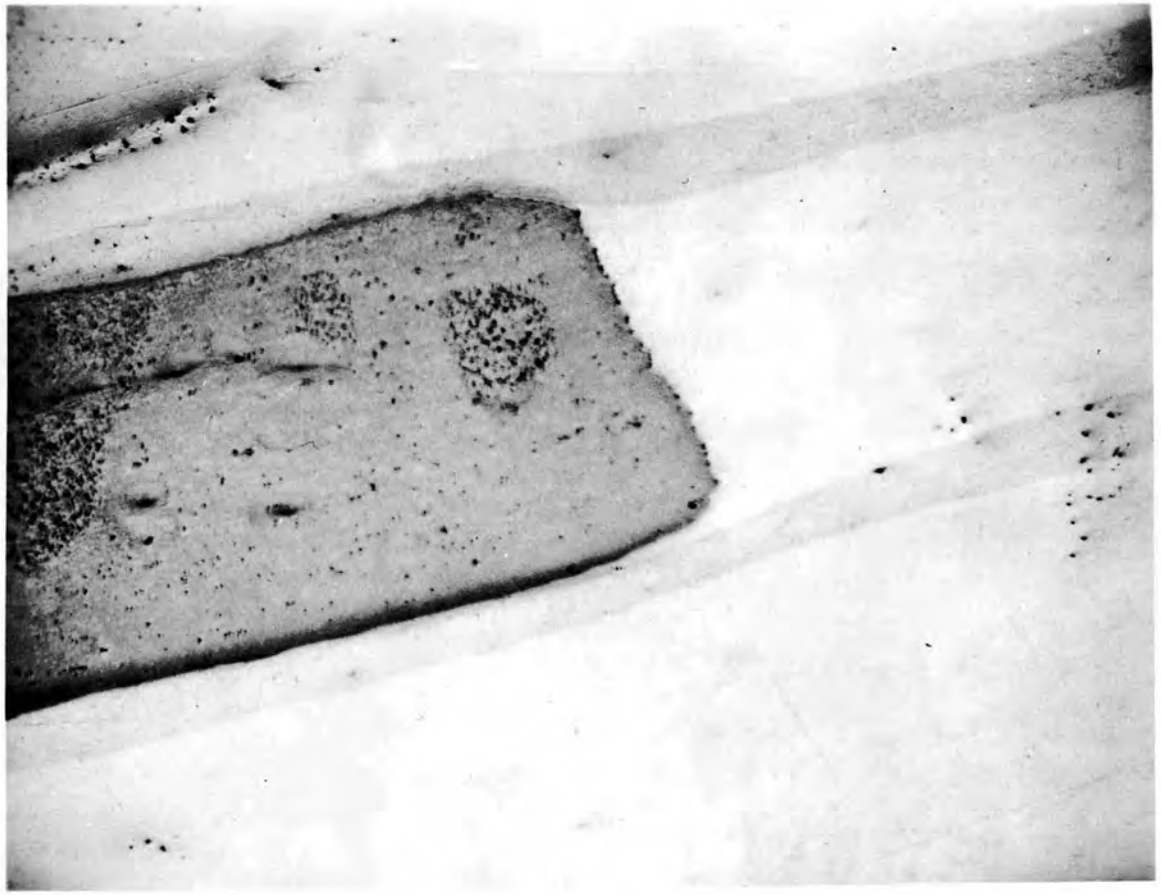

Figure VIII. Photomicrograph of Edge Seal of Irradiated Fuel Element. 100X Magnification.[4]

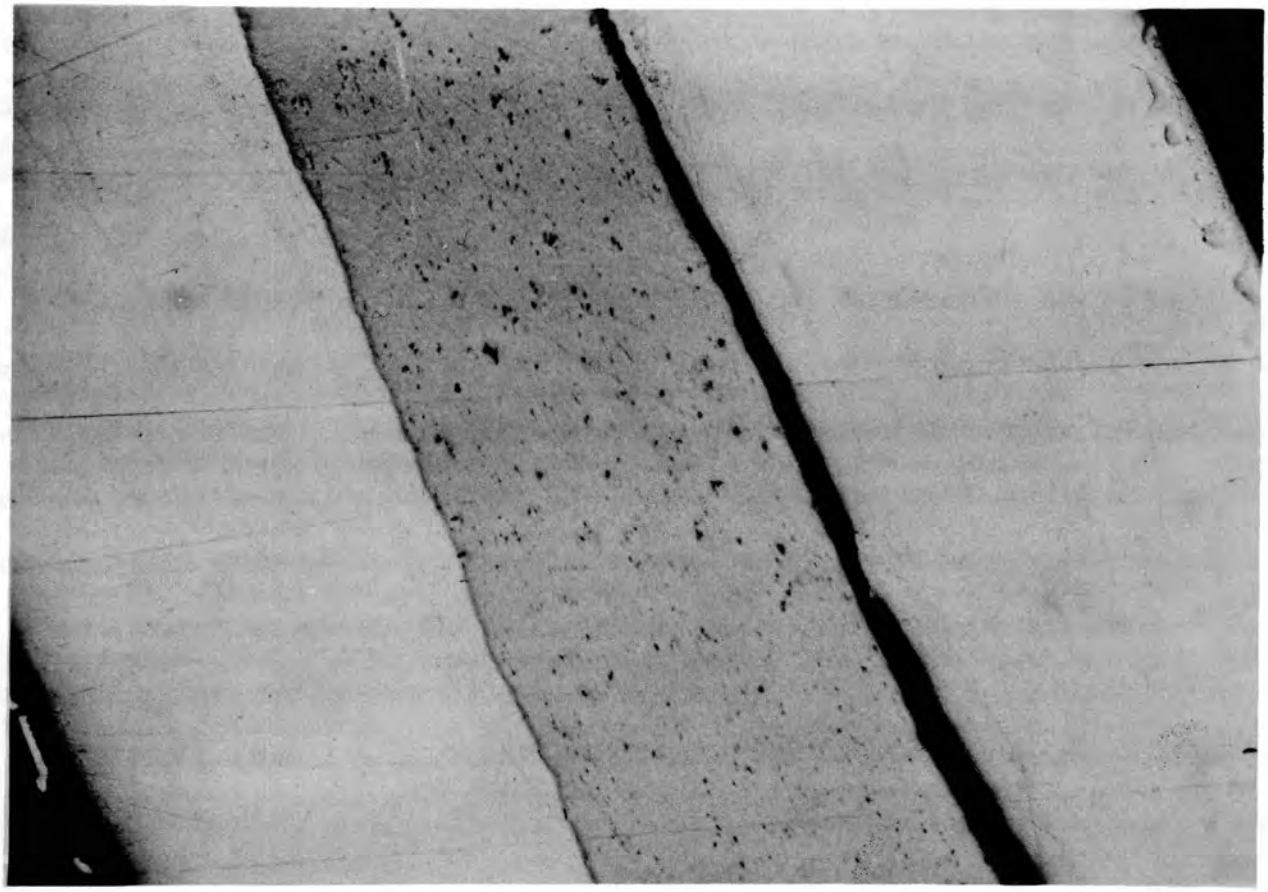

Figure IX. Photomicrograph of Loosened Barrier and Clad from Core of Irradiated Fuel Element as Result of 180° Bend. 100X Magnification. This is an area approximately 1/2 inch from the 180° bend.[4]

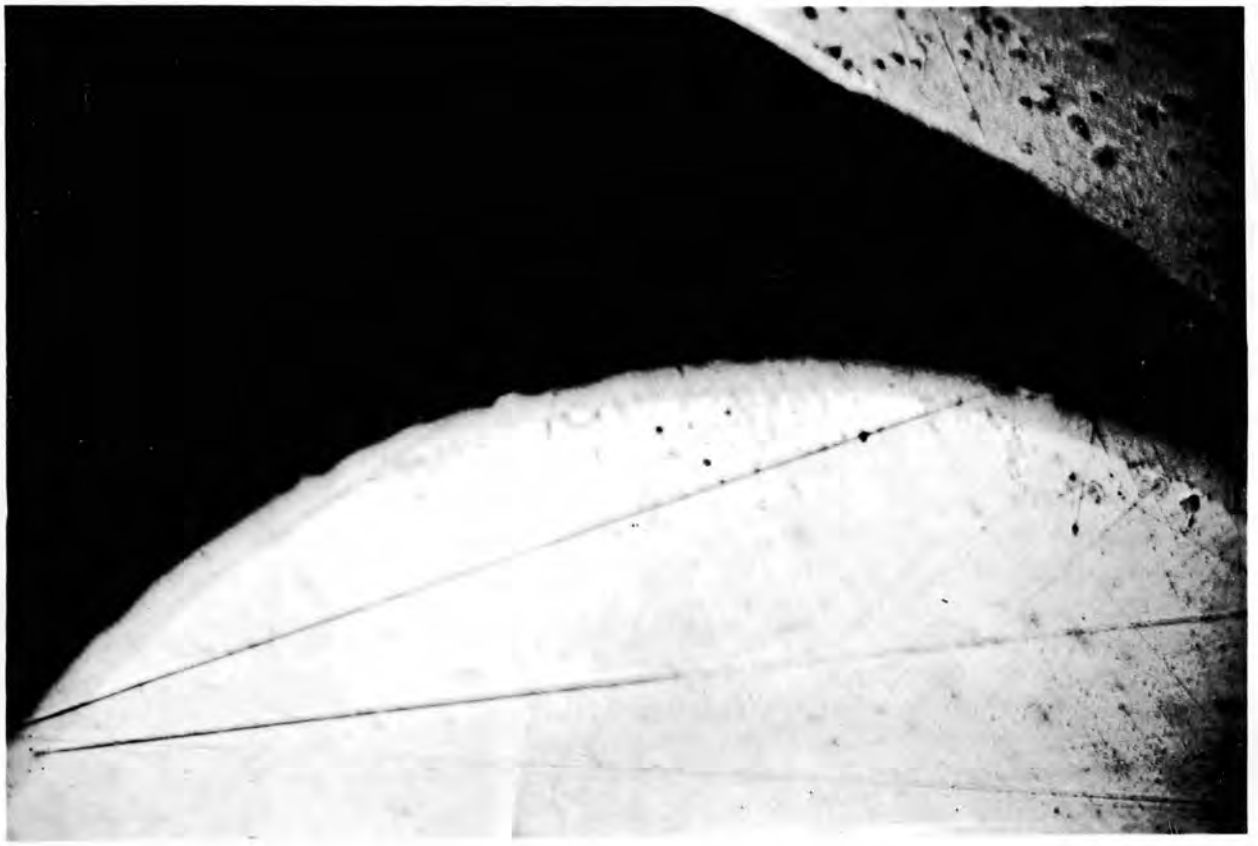

Figure X. Photomicrograph of Clad and Barrier at Point of 180° Bend. 100X Magnification. Although the core and one side of the clad was fractured in this 0.1 inch radius bend, note that the Al-Si barrier is still sound.[4]

TABLE IV

Location	Irradiated Samples	Nonirradiated Samples
Core-Transv.	202/193 DPH*	120/113 DPH*
Core-Long.	198/180 DPH*	114/111 DPH*
Clad-Transv.	88/81 DPH**	80/74 DPH**
Clad-Long.	90/85 DPH**	80/72 DPH**

* 10.25 mm objective, 2 kg load

** 10.25 mm objective, 1 kg load

A method of remotely measuring the thermal conductivity transversely through the samples was desired so that the bond between the clad and core could be non-destructively evaluated. Several methods were attempted, but all failed to produce the desired sensitivity and the thermal conductivity tests were abandoned. Measurements of the electrical conductivity of the cores were also desired, but due to the uneven and irregular cross section of the stripped samples, the experimental error would have been greater than the anticipated changes (See Fig. 5).

Chemical Analysis

The solution containing the dissolved cladding was analyzed and it was determined that approximately 99.99% of the metal removed was aluminum with 0.0011% uranium and less than 0.009% thorium. This suggests that very little diffusion has occurred during rolling and irradiation.

After the completion of the non-destructive tests, samples were obtained for chemical analysis. Since the specimens were very radioactive (10^4 r/hr per gram at contact for plate #1) it was necessary to obtain small samples to be within the limit of activity acceptable for chemical analysis. For this purpose,

a hydraulic press was adapted to operate a small punch within a hot cell. Punchings were obtained from samples #1, #3 and #4. These punchings were not weighed, but punchings from the non-irradiated sample #2 were found to average 0.0277 grams. As nearly as possible, the punchings for the chemical analysis were taken from the center of each sample.

Two punchings, one on each side of the chemical specimen, were taken from sample #1 for a mass analysis of the uranium content. All of the punchings in sample #1 were taken on the longitudinal center line of the sample. The activity of sample #1 indicated that it had received more irradiation than the other samples. The autoradiographs of the cores (Fig. 4) of the samples clearly indicated not only that sample #1 was more active, but also that there is a distinct gradation of activity from one end of sample #1 to the other. In order to minimize the effect of position, the two punchings for mass analysis were taken approximately 1/2 inch from either side of the position of the chemical specimen.

The chemical analysis of the cores indicates the following weight percentages or uranium in the cores.

TABLE V

Sample	% Uranium
#1	2.10 ± 0.2
#3	2.0
#4	1.8
#2*	1.3*
#2*	1.2*

* Not irradiated

The percentage total uranium composition of the alloy from which the cores were formed was determined by three tests prior to rolling.[3] The results of these tests are given as

1.33%
1.45% or an average of 1.3 ± 0.1%
1.23%

This is in agreement with the independently determined analysis of the unirradiated sample #2 (shown in Table V).

The results from the chemical specimens show a constant trend. The irradiated specimens show more uranium content than the unirradiated standard and the percentage composition is in proportion to the degree of irradiation; sample #1 in the highest flux position, #3 in the middle, and #4 in the position of the least flux.

Mass Analysis

A punching from half inch on either side of the center of sample #1 was analysed for the isotopic composition of the uranium. The results for the two punchings are shown below.

TABLE VI

Atom %	1st Sample	2nd Sample
U 233	68.1 ± 0.5	70.9 ± 0.4
U 234	5.6 ± 0.2	5.8 ± 0.1
U 235	15.1 ± 0.2	12.3 ± 0.2
U 236	7.2 ± 0.2	7.3 ± 0.2
U 238	4.0 ± 0.3	3.7 ± 0.2

The difference in isotopic composition is apparently real and due to the variation in neutron flux along the length of the sample.

Since the original composition of the thorium-uranium alloy core was 1.3% uranium and 93.25% of the uranium was U^{235}, it is immediately apparent that the burnup was quite extensive. A simple calculation compares the chemical analysis of sample #1 with the isotopic analysis to show that after irradiation (and decay of the Pa) the core was 1.45 (± 0.15) % U 233 and 0.32 (± 0.05) % U 235.

Theoretical Production of Fissionable Material

It is interesting to compare the measured increase of U 233 with the theoretical increase under the conditions of the experiment.

The process by which U 233 can be produced from thorium is

$$_{90}Th^{232} + {}_{0}n^{1} \longrightarrow {}_{90}Th^{233} \xrightarrow{-\beta} {}_{91}Pa^{233} \xrightarrow{-\beta} {}_{92}U^{233} + {}_{0}n^{1} \begin{cases} \nearrow {}_{92}U^{234} \\ \searrow \text{Fission Products} \end{cases}$$

-23.5 min $\qquad -27.4$ day

When thorium is exposed to a neutron flux the thorium 233 builds up as a function of the flux and reaches an equilibrium concentration in about two hours. The protactinium 233 reaches an equilibrium in about 160 days. The U 233 formed from decay of the protactinium continues to increase until it reaches an equilibrium at the time that the U 233 is fissioned by the neutron flux at a rate equal to that at which it is being produced. If the thorium-uranium sample is removed from the reactor, the concentration of U 233 will again begin to increase due to the decay of the protactinium 233 into uranium 233. Although the equilibrium concentration of uranium 233 decreases with increasing neutron flux, the amount of uranium 233 which is eventually obtainable increases. This is a result of the increasing amounts of protactinium 233 produced during the higher flux.[6]

The samples were irradiated 4153 megawatt days for a total of 2.6×10^{21} neutrons/cm^2 based on an estimated average neutron flux of 2.2×10^{14} neutrons $\cdot$ cm^{-2} $\cdot$ sec^{-1} at reactor midplane.[4] These samples were only about 0.03 inches thick,

excluding the clad, and calculations indicate that slightly greater than 50% of the incident neutrons should penetrate the sample. These samples were in location L-56 of the MTR and at this position the thermal neutron flux should be non-directional. Because of these conditions, the production of U 233 will be considered homogeneous with regard to sample thickness.

The nuclear processes leading to the production of U 233 by exposure of thorium to thermal neutrons may be represented mathematically by a system of linear differential equations of the first order.

(1) $\frac{dN_1}{dt} = \mathcal{a}_0 N_0 - \mathcal{a}_1 N_1$ $\qquad (t = 0,\ N_1 = 0)$

(2) $\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$ $\qquad (t = 0,\ N_2 = 0)$

(3) $\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3$ $\qquad (t = 0,\ N_3 = 0)$

N_0, N_1, N_2, N_3 = number of Th^{232}, Th^{233}, Pa^{233} and U^{233} atoms per unit volume of thorium, respectively, at time, t.

$\lambda_0, \lambda_1, \lambda_2, \lambda_3$ = respective radioactive decay constant.

$\mathcal{a}$ = radioactive decay constant + pseudodecay constant

(i.e. $\mathcal{a}_i = \lambda_i + \sigma_i \phi$)

The analytical solutions of the system of equations have been obtained through the use of an electronic analog computer.[5] The solutions are

(1) $N_1(t) = \frac{k}{\mathcal{a}_1} (1 - e^{-\mathcal{a}_1 t})$

(2) $N_2(t) = \frac{\lambda_1 k}{\mathcal{a}_1 \mathcal{a}_2} \left[1 - e^{-\mathcal{a}_2 t}\right] + \frac{\lambda_1 k}{\mathcal{a}_1(\mathcal{a}_1 - \mathcal{a}_2)} \left[e^{-\mathcal{a}_1 t} - e^{-\mathcal{a}_2 t}\right]$

$$(3)\quad N_3(t) = \frac{\lambda_2 \beta_2}{a_1 - a_3}\left[e^{-a_3 t} - e^{-a_1 t}\right] + \frac{\lambda_2 \beta_1}{a_2 - a_3}\left[e^{-a_2 t} - e^{-a_3 t}\right]$$

$$+ \frac{\lambda_2 \beta_2}{a_2 - a_3}\left[e^{-a_2 t} - e^{-a_3 t}\right] + \frac{\lambda_2 \beta_1}{a_3}\left[1 - e^{-a_3 t}\right]$$

$$k = \sigma_0 N_0 \phi \qquad \beta_1 = \frac{\lambda_1 k}{a_1 a_2} \qquad \beta_2 = \frac{\lambda_1 k}{a_1(a_1 - a_2)} \qquad a_i = \lambda_i + \phi\sigma_i$$

TABLE VII

Isotope	Half Life	Barns
Th^{232}	1.39×10^{10} y	7.0 ± 0.4 (abs.)
Th^{233}	23.3 min	1400 ± 200 (act.)
Pa^{233}	27.4 d	150 (act.)
U^{233}	1.62×10^{5} y	564 ± 10 (abs.)

The values of the constants were calculated using the cross sections and half lives in Table VII. The value of the flux, $\bar{\phi}$, was taken as the estimated average of 2.2×10^{14} neutrons $\cdot$ cm^{-2} $\cdot$ sec^{-1}.(4) The time, t, was assumed as the time required for the product $\bar{\phi}t = 2.6 \times 10^{21}$ neutrons/cm^2, or $t = 1.182 \times 10^{7}$ sec. This calculation will then assume a uniform flux rate for a given time. Since thorium 233 reaches an equilibrium concentration in about two hours it is apparent that any shutdown or decrease in flux will result in an increased rate of change of thorium 233 to protactinium 233 (since at shutdown all of the thorium 233 will decay into protactinium 233 rather than being lost to the process because of the 1400 barn cross section of thorium 233). This would result in an eventual increase in uranium 233.

Thus these "ideal" conditions for purposes of calculation will represent the minimum actual production of uranium 233.

Since there is considerable latitude in the determined values of the cross sections, three calculations were made, one each for the maximum, average, and minimum possible values. The results are

	2.309×10^{20}			(max)
$N_3(t)$	2.068	"	(atoms of U 233/cm^3 of natural thorium)	(avg)
	1.738	"		(min)
	1.417×10^{20}			(max)
$N_2(t)$	1.342	"	(atoms of Pa 233/cm^3 of natural thorium)	(avg)
	1.267	"		(min)

$N_1(t)$ around the order of 10^{17}, neglected.

The time elapsed between removal from reactor and analysis of the sample is sufficiently long to assume that all of the protactinium 233 has decayed into uranium 233. The sum of the two results is then

3.726×10^{20}			(max)
3.410	"	(atoms of U 233/cm^3 of natural thorium)	(avg)
3.005	"		(min)

Assuming a 11.6 gm/cm^3 (7) density for natural thorium, there are 30.084×10^{21} atoms/cm^3 of thorium. Therefore, the calculated conversion of thorium to uranium 233 is

1.24% (max)

1.13% (avg)

1.00% (min)

Since the isotopic and chemical analysis has determined the percentage of uranium 233 to be 1.45 ± 0.15% of the total mass, this is in good comparison to the calculated value. As previously mentioned, the calculated values are for the minimum possible growth under the irradiation conditions.

A determination of flux was made at the point of isotopic analysis by the comparisons of the ratio of the amounts of uranium 236 to uranium 235. Capture of a thermal neutron by uranium 235 leads to fission in about 84% of the captures and to radiative capture the remaining times. Since uranium 236 does not occur in nature it is assumed that all of the uranium 236 was formed from the original uranium 235. The flux calculated by this method was 2.35×10^{21} neutrons/cm^2. This method which is dependent upon the accuracy of the isotopic analysis, provides a check of the estimated flux of 2.6×10^{21} (obtained by cobalt foil measurements) but is not accurate enough to replace that estimate.

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ACKNOWLEDGMENTS

Fabrication of the fuel elements and the preirradiation measurements were performed by R. J. Beaver of the Metallurgy Division. The chemical analysis of the fuel element clad was made by G. R. Wilson of the Chemical Technology Division and the proportion of uranium in the core was determined by W. R. Laing of the Analytical Chemical Division. The isotopic analysis of the uranium in the fuel element core was obtained by L. O. Gilpatrick of the Stable Isotope Research Division. Metallographic analysis of the specimens was made by M. J. Feldman of the Solid State Division. The cooperation of these people and others is gratefully acknowledged.

APPENDIX

The thorium alloys and compounds which have been irradiated present a special problem in handling. The stripped cores of these samples contaminated everything that they contacted. This contamination adhered firmly and required stringent cleaning measures to reduce the contamination to acceptable levels.

When the final clean-up of the hot cell was made, a few flakes of the stripped core fell upon the floor of the hot cell. These flakes were removed by the use of a standard commercial vacuum cleaner with a disposable paper bag filter. Almost immediately after removal of the flakes, the radiation monitors indicated an air-borne radiation contamination. Investigation revealed that the small flakes of thorium-uranium (oxide) had apparently disintegrated when buffeted by the air streams of the vacuum cleaner and so passed through the paper filter. The resulting dust contaminated the entire area of the building in which the hot cell was located. The contamination was uniform, with no especially hot spots. Repeated scrubbings were necessary to reduce the contamination to an acceptable level.

Subsequent experience has indicated that all irradiated thorium compounds may be suspected of presenting a severe contamination hazard.