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EFFECTS OF IRRADIATION ON THE MECHANICAL PROPERTIES OF TANTALUM

by

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Tensile, bend ductility, and hardness tests were performed at room temperature on irradiated tantalum sheet to determine the effect of irradiation on the strength and ductility of tantalum. Sheet tensile specimens were irradiated in the MTR in an attempt to produce conversions of tantalum to tungsten of approximately 1.5 and 3.0 w/o. Unirradiated tantalum and arc-melted alloys of tantalum-1.5 and –3.0 w/o tungsten were tested for comparison with the irradiated material.

The tensile and yield strengths of tantalum were found to increase appreciably as a result of irradiation, whereas the tensile properties of unirradiated tantalum-tungsten alloys prepared by arc melting showed that small additions of tungsten do not significantly increase the strength of tantalum. These results indicate that the major part of the increase in strength resulting from irradiation of tantalum can be attributed to fast-neutron damage and that any contribution produced by the conversion of tantalum to tungsten is a minor one.

INTRODUCTION

The plutonium-bearing fuel of the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE) is to be contained in tantalum tubes. Tantalum is attractive as a container material because of its easy formability and because of its relatively high strength at the reactor operating temperature. However, little is known about the effect of irradiation on its mechanical properties.

Tantalum undergoes two modifications when exposed to a neutron flux of the type anticipated in the LAMPRE. The first of these is the conversion of tantalum to tungsten by the thermal-neutron reaction, $Ta^{181}(n,\gamma)Ta^{182}(-\beta)W^{182}$. Under the anticipated operating conditions of the molten-plutonium reactor up to 3 w/o tungsten could be formed in this manner. The second modification is the lattice disordering caused by high-energy neutrons. This damage consists of the formation of vacancy interstitial combinations associated with high-energy atom displacements. Both of these modifications in the tantalum structure should result in increases in hardness and tensile and yield strength, and decreases in ductility.

Since the properties of the container material, particularly its ductility, are very important in this application, it was desirable that data be obtained which would provide a means for evaluating the effects of neutron bombardment. The most direct manner for accomplishing this was to produce and test in the unirradiated state unalloyed tantalum and tantalum alloys containing tungsten in the amounts which would be produced by irradiation. In addition, unalloyed tantalum was exposed to neutron dosages sufficient to produce the reference conversions of tungsten and then tested after the irradiation. By a comparison of hardness, tensile properties, and ductility of these various test materials, the effect of tungsten formation and of fast-neutron damage was assessable.

A research program based on the approach outlined above was initiated at Battelle Memorial Institute at the request of Los Alamos Scientific Laboratory. The results of the program are reported in the following sections.

EXPERIMENTAL WORK

Specimen Preparation

The tantalum used in the program was supplied by LASL. The material was received in the form of 0.030-in. sheet which had been rolled from Temescal electron-beam-melted tantalum stock and fully annealed at 2500 F. This sheet stock was also used in the preparation of high-purity tantalum-1.5 and -3.0 w/o tungsten alloys. These alloys were prepared by arc melting under a partial pressure of helium in a copper hearth using a tungsten electrode. A tantalum rod near the edge of the hearth was used to strike the arc and to gather the residual gases in the furnace. The alloys in the form of 250-g buttons were melted six times to insure homogenity.

The buttons were cold pressed with total reductions ranging from 20 to 40 per cent to break up the cast structure and then machined to have top and bottom surfaces parallel. After cleaning in aqua regia, the buttons were cold rolled to a thickness of 0.150 in., vacuum annealed for 1 hr at 2600 F, and cold rolled to a final thickness of about 0.030 in. Tensile specimens were machined from the 30-mil sheet and again annealed in vacuum for 2 hr at 2600 F.

The final dimensions of the tensile specimens were:

Length	4.50 ± 1/64 in.
Width	11/32 ± 1/64 in.
Gage length	1.0 ± 0.002 in.
Gage width	0.250 ± 0.005 in.
Radius at gage length	1.0 in.

A chemical analysis of the unalloyed tantalum and the tantalum-tungsten specimens is given in Table 1.

Capsule Design

The capsules prepared for the irradiation were designed to provide the following conditions: (1) 1.5 and 3.0 w/o tantalum-to-tungsten conversion; (2) high, uniform thermal flux; (3) fast flux; and (4) low specimen temperature.

TABLE 1. CHEMICAL ANALYSIS OF THE TANTALUM AND TANTALUM-TUNGSTEN ALLOYS[a]

Impurity	Analyses for Indicated Specimen, ppm		
	Unalloyed Tantalum	Tantalum-1.5 w/o Tungsten	Tantalum-3.0 w/o Tungsten
Aluminum	<5	15	20
Chromium	10	4	5
Copper	10	20	15
Iron	3	6	15
Molybdenum	--	20	15
Niobium	--	300	100
Nickel	--	6	3
Silicon	--	30	60
Zirconium	--	15	10
Nitrogen	<10	20	35
Carbon	10	20	35
Hydrogen	0.3	1	1
Oxygen	40	53	22

(a) Average of two analyses taken of the alloys after they had been cold rolled to 0.030-in. strip and vacuum annealed.

To incorporate the above specifications into the capsule design, the nuclear properties of tantalum along with the physical and thermal characteristics of the capsule proper were evaluated. The neutron perturbation of the specimen and engineering considerations needed for design of the capsules are given in the following paragraphs.

Examination of the nuclear reactions showed that in the presence of thermal neutrons tantalum will be converted to tungsten by the following schemes:

$$Ta^{181} + n \longrightarrow Ta^{182} \xrightarrow{-\beta} W^{182}$$

$$Ta^{182} + n \longrightarrow Ta^{183} \xrightarrow{-\beta} W^{183}$$

The former is the more important reaction and, as outlined in detail in the appendix, the fraction of tungsten formed as a function of time can be expressed as follows:

$$\text{Fraction of tungsten formed} = 1 - e^{\sigma_1 \phi t},$$

where

σ_1 = tantalum-181 cross section, cm^2 per atom

ϕ = thermal-neutron density, $n/(cm^2)(sec)$

t = time, sec.

Since the design conversions are 1.5 and 3.0 w/o, the necessary dosages are found from this equation to be 7×10^{20} and 1.4×10^{21} nvt, respectively.

Calculations to determine capsule thermal-neutron-flux perturbation characteristics and, therefore, the unperturbed flux levels required to provide these dosages in

in-pile exposures of reasonable length took into account the macroscopic cross section of both tantalum-^{181} and tantalum-^{182}. The perturbation factor was obtained by use of the IBM 650 computer with the P_3 simplification of the spherical harmonics transport equation. The average thermal-neutron flux in the tantalum region was found to be about 0.37 of the unperturbed flux level in the reactor facility.

Engineering Design and Loading

It was desirable to irradiate the tensile specimens at as low a temperature as possible and still have all six specimens at about the same thermal-neutron-flux level. To meet these requirements, the tensile specimens were held vertically around a stainless steel basket to form a "ring" as shown in Figure 1. A sodium-potassium alloy (NaK containing 56 w/o potassium) which has a high thermal conductivity was used as the heat-transfer medium. A total heat-generation rate of 1340 Btu per hr was found for six specimens using a value for gamma heating of 6 w per g. This amount of heat dissipated by radial heat transfer into a reactor water sink of 120 F should produce a specimen temperature of about 200 F, which is acceptable.

To reduce specimen contamination during irradiation, the tantalum specimens were scrubbed with Alconox, washed with aqua regia, and thoroughly rinsed with distilled water to remove surface grit; they were then baked in a vacuum furnace to remove moisture. The stainless steel capsule parts were subjected to a similar cleaning operation. The NaK and specimens were loaded into the capsules in a dry box containing a helium atmosphere. The helium permitted leak checking of the capsule welds with a helium mass spectrometer.

Nickel-cobalt dosimeter wires were placed around the tensile specimens in order to verify the thermal- and fast-neutron-flux density in the region of the specimens. Aluminum-cobalt wires were also attached to the surfaces of the capsules.

Irradiation History

The two stainless steel capsules, designated 25-1 and 25-2, were inserted into MTR core positions L-53 and L-57, respectively, during the shutdown for Cycle 122.

Capsule BMI-25-1, which was scheduled for a tantalum-to-tungsten conversion of 1.5 w/o, was irradiated for four cycles (66 days) in an estimated unperturbed thermal-neutron flux of 4.0×10^{14} nv. Capsule BMI-25-2, scheduled for a conversion of 3.0 w/o, was irradiated for seven cycles (109 days) in an estimated unperturbed flux of 5.0×10^{14} nv.

The total estimated specimen thermal-neutron doses based on the reactor-quoted fluxes and the perturbation factor of 0.37 are 8.6×10^{20} and 1.8×10^{21} nvt for Capsules BMI-25-1 and BMI-25-2, respectively, which are about 25 per cent higher than the required doses.

Capsule BMI-25-1 contained Specimens 1 through 6 and Capsule BMI-25-2 contained Specimens 7 through 12.

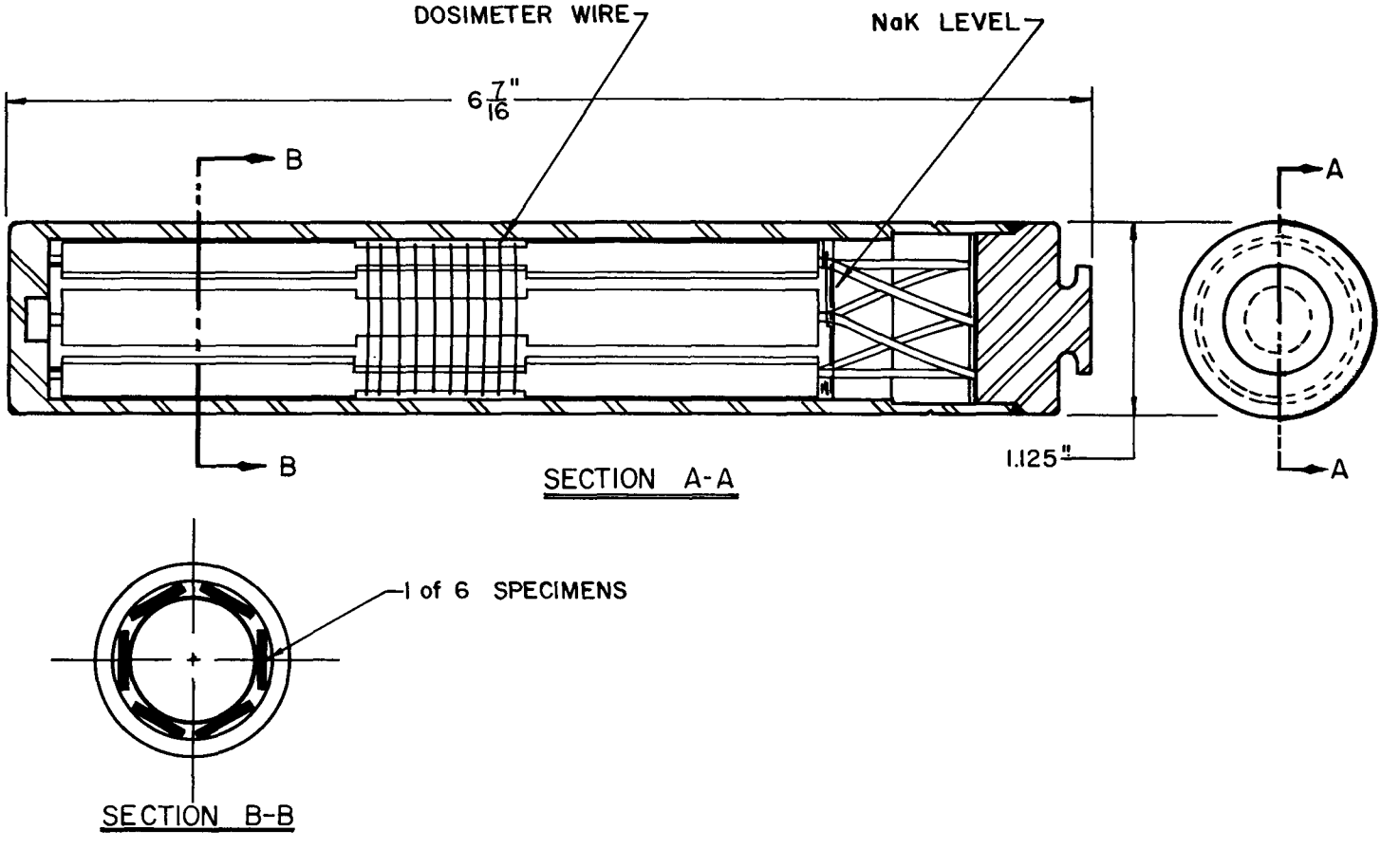


FIGURE 1. CAPSULE ASSEMBLY

POSTIRRADIATION EXAMINATION

The examination of the irradiated specimens was performed in the Battelle Hot-Cell Facility. Where possible, the pre- and postirradiation measurements were taken with the same equipment and equipment standardization tests were made to validate the pre- and postirradiation measurement comparisons.

The capsules were opened upon their arrival at the Hot-Cell Facility. Particular care was exercised in removing the specimens so as not to bend or stress them in any way. As soon as the specimens were removed from the stainless steel capsules, they were cleaned with butyl alcohol and photographed. Photographs of the specimens are shown in Figure 2. Visually, the specimens appeared undamaged as a result of the irradiation. A white coating was seen at various areas on the surfaces of the specimens and was thought to be the result of the NaK-butyl reaction. The residue was found to be quite adherent and had to be removed with a wire brush.

Mechanical-Properties Tests

Tensile Tests

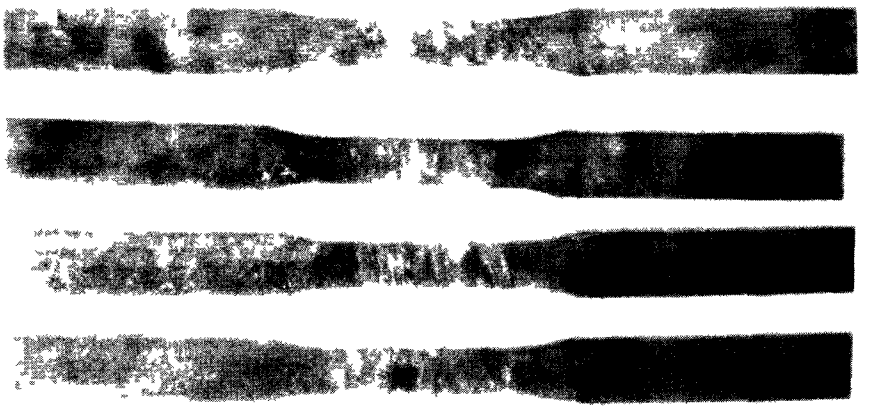
The tensile properties of the unirradiated and irradiated tantalum specimens were determined at room temperature with a Riehle tensile machine (maximum load range 3000 lb) using a strain rate of 0.005 in. per in. in the plastic region. A clip-on linear variable differential transformer-type extensometer was used with an autographic recorder to obtain load-deformation curves. Figure 3 shows an irradiated specimen and attached extensometer mounted in the Riehle tensile machine. The arc-melted alloy specimens were tested with a Baldwin-Southwork-Universal testing machine (maximum load range 2400 lb) using a strain rate of 0.005 in. per in.

The results of the tensile tests of the unirradiated and irradiated tantalum and the tantalum-1.5 and 3.0 w/o alloy specimens are shown in Table 2. Comparison of the tensile properties of unirradiated and irradiated tantalum shows increases in strength with increasing irradiation exposures. The ultimate strength of tantalum which had been irradiated for approximately 66 days increased from about 42,000 to about 70,000 psi. Exposures to 109 days further increased the ultimate strength of the material to about 86,000 psi. Comparison of these data with those for the unirradiated arc-melted alloys shows that the increases in tensile strength are far greater than would be expected from alloying alone. Ultimate strengths of 45,000 and 52,000 psi were observed for the arc-melted alloys containing 1.5 and 3.0 w/o tungsten, respectively.

The irradiated specimens also show a marked loss in tensile ductility as indicated by the total elongation measurements. The ductility of tantalum was reduced by about 83 per cent after 109 days of radiation.

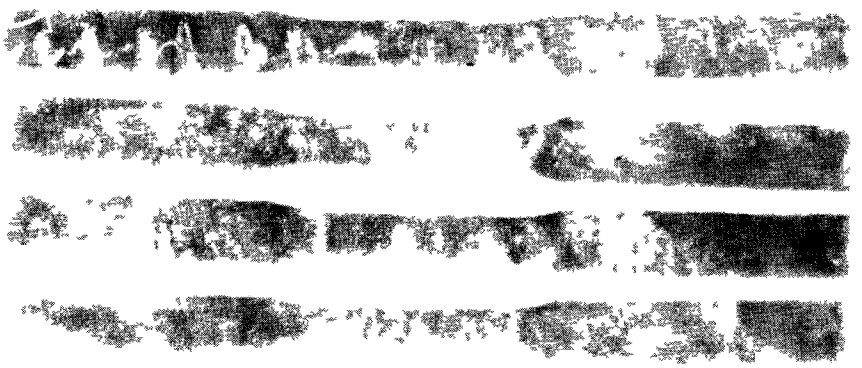
Bend Tests

A qualitative measure of the ductility of the irradiated tantalum was made by comparing bend characteristics of the unirradiated and irradiated specimens. In this



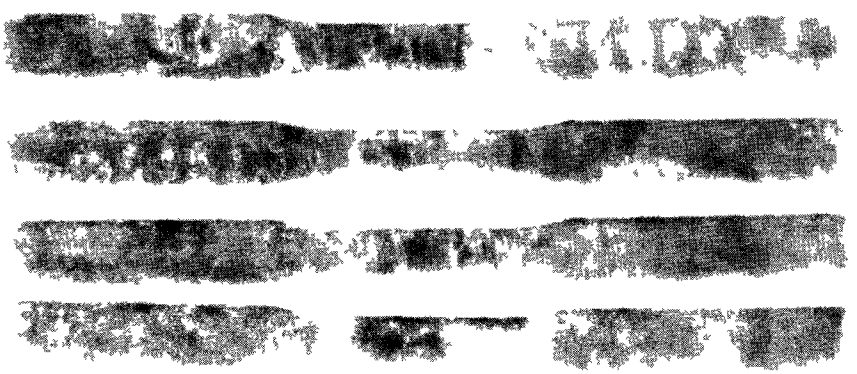
1X HC4258

a. Specimens 1, 2, 3, and 4



1X HC4259

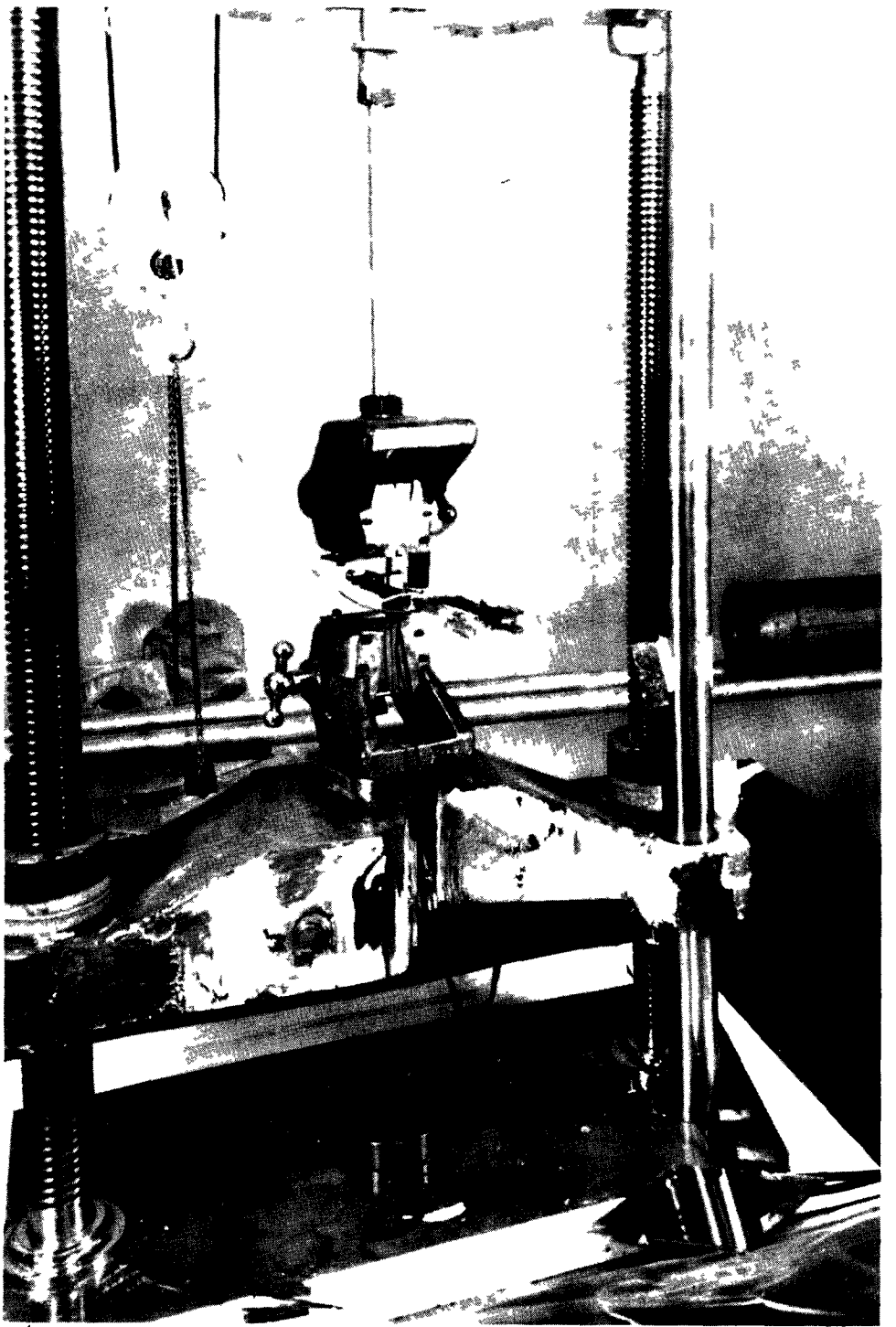
b. Specimens 5, 6, 7, and 8



1X HC4260

c. Specimens 9, 10, 11, and 12

FIGURE 2. IRRADIATED TANTALUM SPECIMENS



1/2X N64905

FIGURE 3. IRRADIATED TENSILE SPECIMEN MOUNTED IN THE RIEHLE TENSILE MACHINE WITH THE EXTENSOMETER ATTACHED

test, the grip ends of broken tensile samples were fastened securely in a vise and bent 90 deg from vertical. After bending 90 deg, the specimens were examined visually at 20 magnifications under a 3-D viewer. No evidence of cracking in or near the bending zone was found in either the unirradiated or irradiated specimens.

TABLE 2. ROOM-TEMPERATURE MECHANICAL PROPERTIES OF UNIRRADIATED AND IRRADIATED TANTALUM AND UNIRRADIATED TANTALUM-1.5 AND -3.0 w/o TUNGSTEN ALLOYS

Specimen Description	Number of Specimen Tests	Total Integrated Thermal Flux Based on Dosimetry Analysis, nvt	Average of Properties Tests				
			Ultimate Tensile Strength, psi	0.2 Per Cent Offset Yield Strength, psi	Elongation at Maximum Load, per cent	Elongation in 1 In., per cent	Hardness, KHN
Unirradiated tantalum	4	--	42,000	30,000	--	40	103
Unirradiated tantalum-1.5 w/o tungsten	3	--	44,900	31,000	--	39	151
Unirradiated tantalum-3.0 w/o tungsten	3	--	52,400	38,500	--	35	170
Irradiated tantalum (66-day irradiation)	4	7.8×10^{20}	69,500	65,800	--	16	274
Irradiated tantalum (109-day irradiation)	4	1.57×10^{21}	86,300	81,400	--	7	309

Hardness Determinations

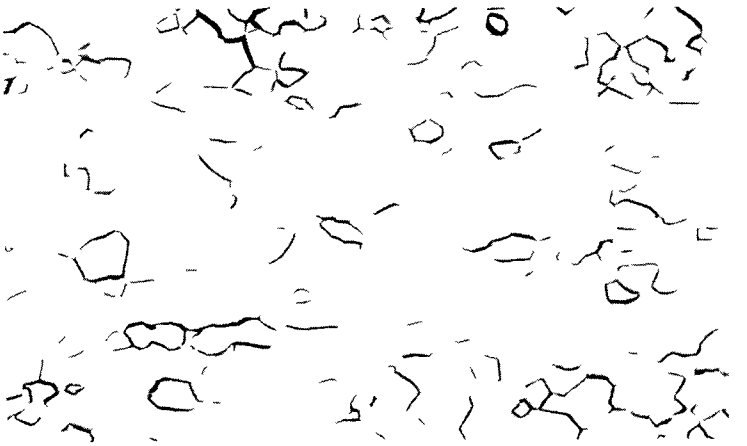
A series of hardness impressions was made on both unirradiated and irradiated specimens with a Tukon hardness tester using a Knoop diamond indenter and 100-g load. The samples used in determining the microhardness were taken from near the end of the tensile specimens so as to avoid testing worked material. Four readings were taken on each specimen. Table 2 gives the average hardness of the materials.

Analysis of the hardness data indicates that the increase in hardness of irradiated tantalum is much greater than that expected from alloying alone. The arc-melted alloys show increases in hardness of about 50 to 70 KHN depending on the amount of alloying addition; but in the case of the irradiated tantalum, increases of as much as 205 KHN were found.

Variations of hardness were found for some of the irradiated specimens. Within a single sample, variations of 30 to 50 KHN for different areas were noticed. These variations in hardness were associated with fine nonmetallic dispersions found in localized areas of the material.

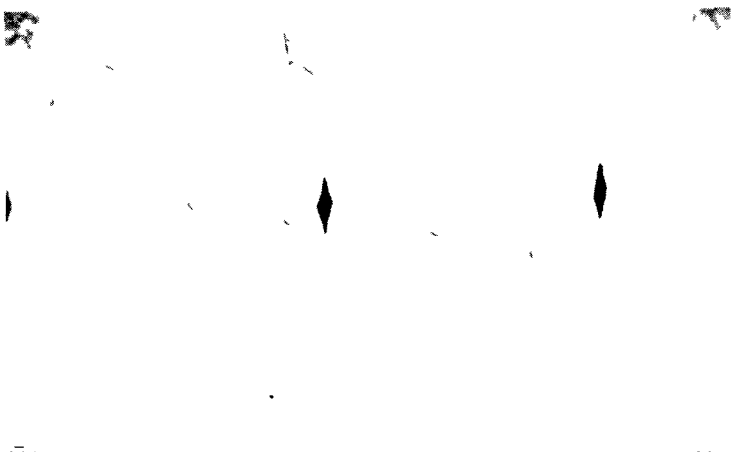
Metallography

A metallographic examination of the tantalum was made before and after irradiation. Specimens were mounted in bakelite and rough polished through 600-grit silicon



100X RM4332

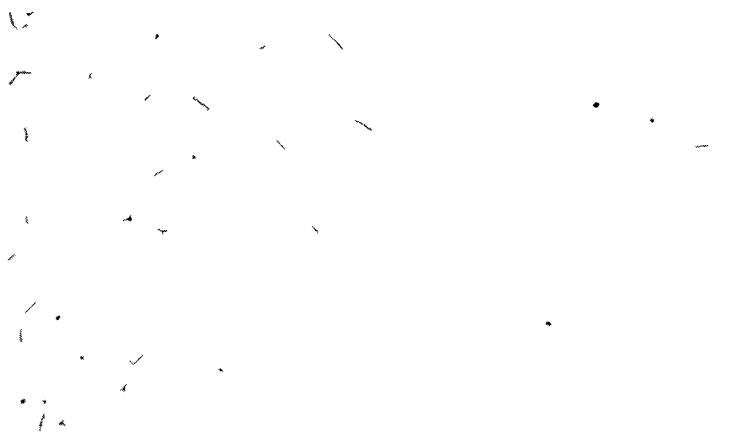
a. Unirradiated Specimen



100X HC4334

b. Specimen 3, Irradiated 66 Days

Hardness impressions may be seen.



100X HC4333

c. Specimen 12, Irradiated 109 Days

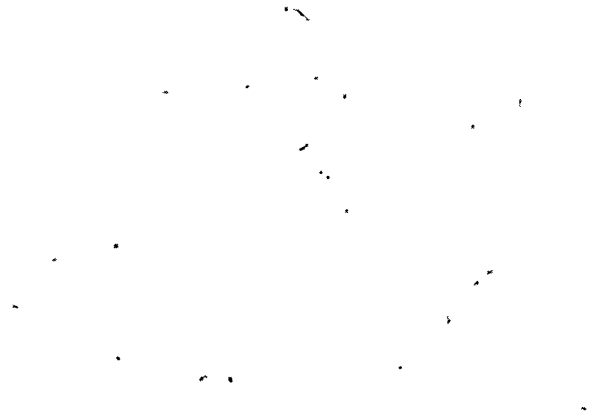
FIGURE 4. MICROSTRUCTURES OF IRRADIATED AND UNIRRADIATED TANTALUM SPECIMENS

All specimens were etched with $HF:HNO_3:H_2O$.

100X HC4335

d. Specimen 10, Irradiated 109 Days

Note small amount of dispersed particles.



100X HC4336

e. Another Area of Specimen 10

This region of the specimen contained large amounts of fine dispersion.

FIGURE 4. (CONTINUED)

carbide papers. Final polishing was accomplished on a Model "L" vibratory polisher with slurries of Linde A and Linde B abrasive using water as a lubricant. The specimens were then swab-etched with an etchant consisting of equal parts of hydrofluoric acid, nitric acid, and water.

Figure 4a shows the microstructure of unirradiated tantalum. Figure 4b shows the microstructure of tantalum (Specimen 3) after 66 days of irradiation. The microstructures of tantalum (Specimens 10 and 12) after 109 days of irradiation are shown in Figures 4c, 4d, and 4e.

No significant changes in appearance of the microstructure were observed as a result of irradiation. Localized areas of fine dispersions were found in some of the irradiated specimens. Examples of these areas are shown in Figure 4d and 4e. The identity of these dispersions in the specimens was not established but they appear to be an oxide or possibly a carbide. There was no observable change in the grain size.

Hardness measurements taken within these areas containing precipitate generally gave higher hardness readings than those taken in areas void of the dispersions.

Dosimeter Analysis

Neutron fluxes were determined by radiochemical analysis of the aluminum-cobalt and nickel-cobalt wires from Capsules BMI-25-1 and BMI-25-2. The dosimetry results are given in Table 3. The total integrated fast flux is reported as based on the determination of cobalt-58 produced by the $Ni^{58}(n,p)Co^{58}$ reaction. It is a threshold detector which has a threshold energy of approximately 5.0 Mev. By assuming a fission spectrum, extrapolation will give the total neutrons having energies >1 Mev. Fast-flux calculations based on long-time exposures of nickel, greater than 2500 megawatt-days result in error related to the neutron cross section of cobalt-58, i.e., the measured fast flux is less than experienced. This is evident from Table 3 where the instantaneous flux measured for 65.9 days is about twice that measured for 109 days.

TABLE 3. RESULTS OF DOSIMETRY ANALYSIS OF ALUMINUM-COBALT AND NICKEL-COBALT WIRES FROM CAPSULES BMI-25-1 AND BMI-25-2

Capsule	Wire Composition	Wire Location	Irradiation Time, days	Decay Time, days	Dosimetry Analysis[a], nv: Thermal-Neutron Flux	Dosimetry Analysis[a], nv: Fast-Neutron Flux
BMI-25-1	Aluminum-cobalt	Outside	65.9	160	1.51×10^{14}	--
	Nickel-cobalt	Inside	65.9	172	1.31×10^{14}	3.38×10^{13}
BMI-25-2	Aluminum-cobalt	Outside	109.0	99	1.73×10^{14}	--
	Nickel-cobalt	Inside	109.0	106	1.65×10^{14}	1.61×10^{13}

(a) In the positions which these capsules were irradiated, the fast flux is lower than the thermal flux by a factor of 10.

Calculation of the integrated thermal flux using the values obtained from the aluminum-cobalt dosimeter analysis shows that the level of radiation was sufficient to give the desired conversion of tantalum to tungsten.

Chemical Analysis

In order to determine the amount of tungsten present after irradiation, samples from three specimens from each capsule were analyzed. These samples, weighing approximately 0.10 g each, were taken immediately adjacent to the fracture zone. They were dissolved in a solution of HF and HNO_3 and diluted to 50 ml in the hot cell. A spectrographic analysis was originally contemplated in which about 100 mg of tantalum was deposited on specially prepared electrodes. Checking the radiation level of the electrodes after the tantalum had been deposited gave readings of approximately 250 mr per hr at contact, higher by a factor of 10 than can be tolerated in the present facilities for spectrographic samples. However, a recently developed ion-exchange separation method for analysis for niobium-tantalum-tungsten-titanium already in use at Battelle appeared suitable and was employed with an appropriate scale-down for the small-size samples used.

The separation is based on the absorption of tantalum-tungsten complexes of the materials on Dowex 1 from an aqueous HF-HCl solution followed by selective solution with appropriate solvents. Experimental testing of unirradiated tantalum samples showed that tantalum was not soluble in the HF-HCl solution required for ion exchange but that it could be dissolved in HF to which a little HNO_3 was added an an oxidant. The HNO_3 addition was kept at a minimum since it can interfere in the tungsten separation if excessive amounts are used. New 0.10-g samples were taken adjacent to the fracture. The samples were dissolved and the composition of the solution then adjusted by addition of suitable quantities of an aqueous HCl solution. The adjusted solutions were passed through a 1/2 by 7 in. column of Dowex 1 (minus 200 plus 400 mesh), the effluent being discarded. A check on one of the effluents indicated negligible activity (~200 mr per hr) indicating quantitative absorption of the tantalum. The tungsten was then eluted with 30 ml of an HN_4Cl-HCl-Hf mixture. The eluate samples were withdrawn from the hot cell, diluted to 50 ml, and subsequently analyzed.

The tungsten was analyzed by the colorimetric dithiol procedure routinely used in the analytical laboratory. The results of the analyses are given in Table 4. These values are much lower than the values of 1.5 and 3.0 w/o tungsten conversion predicted by the radiochemical analyses of the dosimeter wires. The separation procedure is reportedly quantitative, and no significant loss of tungsten should have occurred. However, there is the possibility that the 30 ml used to elute the tungsten was an insufficient volume to completely remove it.

TABLE 4. TUNGSTEN ANALYSIS OF IRRADIATED SPECIMENS

Capsule and Specimen	Tungsten, w/o ± 10 per cent
BMI-25-1	
1	0.6
3	0.8
6	0.7
BMI-25-2	
8	0.8
10	1.2
12	2.2

DISCUSSION AND CONCLUSIONS

Mechanical Properties

The mechanical properties of tantalum are found to be affected by irradiation. Figure 5 summarizes the changes in tensile properties of tantalum as tungsten was added by: (1) irradiation and (2) arc melting. Yield strength in the case of the irradiated material (109 days of irradiation) increased nearly 3 times over that of the unirradiated tantalum. Comparison of the tensile properties of the unirradiated arc-melted alloys with unirradiated tantalum shows that the addition of relatively small amounts of tungsten does not markedly increase the strength of tantalum. It is felt that the large increase in tensile and yield strength noted after irradiation is primarily the result of damage to the material by high-energy neutrons. There is no evidence that the tantalum-tungsten alloys made by arc melting will be any different than comparable tantalum-tungsten alloys formed by the nuclear transmutation of tantalum to tungsten. The atomic size of the tantalum and tungsten atoms is effectively the same. Alloying occurs by atomic substitution. Therefore, no appreciable atomic-lattice distortion or stress will occur because of the addition of tungsten atoms to a tantalum matrix no matter if the atoms are added by the arc-melting process or by nuclear transmutation.

The tantalum specimens showed a marked loss in ductility after irradiation as indicated by the total elongation measurements. Reductions as high as 83 per cent were observed after 109 days of irradiation. Although the tensile elongation was greatly reduced, bend tests of the specimens after irradiation showed that no extremely severe embrittlement of the material had occurred.

Hardness

Tantalum showed a great increase in hardness after irradiation. Hardness increased from 103 KHN to 309 after seven reactor cycles. The tantalum-tungsten alloys produced by arc melting showed hardnesses of 151 KHN for additions of 1.5 w/o tungsten and 170 KHN for 3.0 w/o tungsten.

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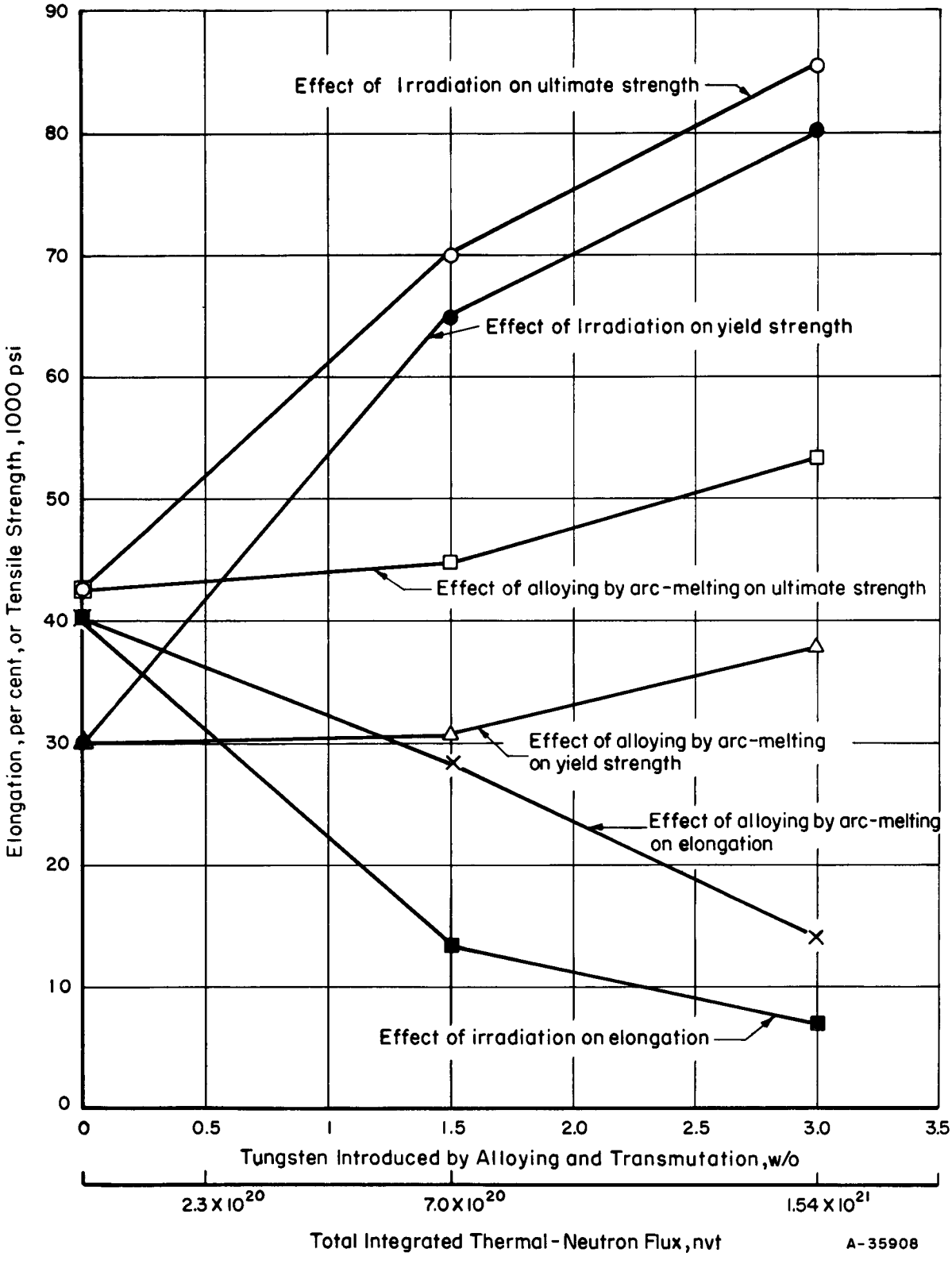

FIGURE 5. THE EFFECTS OF ALLOYING AND OF IRRADIATION ON THE TENSILE PROPERTIES OF TANTALUM

APPENDIX

NUCLEAR CALCULATIONS

APPENDIX

NUCLEAR CALCULATIONS

In the case of tantalum, radioactive nuclei, tantalum-182, are subject to transformation by neutron absorption in addition to decay by beta emission, as seen in Equations (A-1) and (A-2):

$$Ta^{181} + n \longrightarrow Ta^{182} \xrightarrow{-\beta} W^{182} \qquad \text{(A-1)}$$

$$Ta^{182} + n \longrightarrow Ta^{183} \xrightarrow{-\beta} W^{183} \; . \qquad \text{(A-2)}$$

Equations are then set up for the change in the concentration of each nuclide with time:

$$\frac{dN_1}{dt} = \sigma_1 \phi N_1 \; , \qquad \text{(A-3)}$$

$$\frac{dN_2}{dt} = \sigma_1 \phi N_1 - \sigma_2 \phi N_2 - \lambda_2 N_2 \; , \qquad \text{(A-4)}$$

$$\frac{dN_3}{dt} = \sigma_2 \phi N_2 - \sigma_3 \phi N_3 - \lambda_3 N_3 \; , \qquad \text{(A-5)}$$

$$\frac{dN_4}{dt} = \lambda_3 N_3 - \sigma_4 \phi N_4 \; , \qquad \text{(A-6)}$$

where

N_1 = number of tantalum-181 atoms at any time, t

N_2 = number of tantalum-182 atoms at any time, t

N_3 = number of tantalum-183 atoms at any time, t

N_4 = number of tungsten-183 atoms at any time, t

σ = thermal-neutron-absorption cross section, barns

ϕ = effective thermal-neutron-flux density, n/(cm^2)(sec)

λ = disintegration constant, 1/sec.

Because of the very high thermal-neutron-absorption cross section of tantalum-182 (20,000 barns) and its long half-life (112 days), very little of the tantalum-182 is depleted by decay and substantially all of the tantalum-182 is depleted by thermal-neutron capture and transformed to tantalum-183. Equation (A-4) can then be simplified as follows by neglecting the $\lambda_2 N_2$ term:

$$\frac{dN_2}{dt} = \sigma_1 \phi N_1 - \lambda_2 \phi N_2 \quad . \tag{A-4a}$$

Tantalum-183, on the other hand, has a small thermal-neutron-absorption cross section (11 barns) and a short half-life (5.2 days) so that substantially all of the tantalum-183 is depleted by beta decay to form tungsten-183. Neglecting the $\sigma_3 \phi N_5$ term in Equation (A-5) yields:

$$\frac{dN_3}{dt} = \sigma_2 \phi N_2 - \lambda_3 N_3 \quad . \tag{A-5a}$$

The tungsten-183 formed as the result of beta decay as shown in Equation (A-5a) is not readily depleted because its macroscopic capture cross section is small.

Equations (A-3), (A-4a), (A-5a), and (A-6) are then integrated in order between the limits of time, zero, and time, t, substituting the results of Equation (A-3) into (A-4a), and so on. Equations are obtained for the amount of each nuclide as a function of time. Of these four equations, the two expressing the amounts of Ta^{182} (N_2) and W^{183} (N_4) are most important:

$$N_2 = \frac{\sigma_1 N_{1(o)}}{\sigma_2 - \sigma_1} \left[- e^{-\sigma_1 \phi t} - e^{-\sigma_2 \phi t} \right] \tag{A-7}$$

$$N_4 = N_{1(o)} \left[1 - e^{-\sigma_1 \phi t} + \frac{\sigma_1 \lambda_3}{\sigma_2 (\lambda_3 - \sigma_2 \phi)} e^{-\sigma_2 \phi t} - \frac{\sigma_1 \sigma_2 \phi \phi}{\lambda_3 (\lambda_3 - \sigma_2 \phi)} e^{-\lambda_3 t} \right] \quad . \tag{A-8}$$

Since the last two terms in Equation (A-8) are very small for large values of time (4×10^6 sec), the following simplified equation results:

$$\frac{N_4}{N_{1(o)}} = 1 - e^{-\sigma_1 \phi t} \quad , \tag{A-8a}$$

where

$N_{1(o)}$ = number of tantalum-181 atoms initially present.

From Equation (A-8a) and the tantalum-to-tungsten design conversion of 1.5 and 3.0 w/o, the necessary doses are found to be 7.0×10^{20} and 1.4×10^{21} nvt, respectively. Since the nvt doses are now known, a knowledge of the capsule perturbation factor* will yield the in-pile time for any unperturbed thermal-neutron-flux density. The P_3 simplification of the spherical harmonics transport equation was used to determine this factor. It obtains its name from the fact that the angular distribution of the flux is approximated by a series of Legendre polynomials which is terminated with the third (or P_3) term. The calculations were made on the IBM-650 computer using the actual capsule geometry, and the perturbation factor was found to be 0.37.

*It should be noted in the calculation of perturbation that both tantalum-181 and tantalum-182 are competing for the thermal neutrons since the macroscopic cross section (the product of the atom density, atoms per cm^3, and the microscopic cross section, barns) of tantalum-182 approaches that of tantalum-181. The fact that this equilibrium exists can be verified with the use of Equation (A-7) which gives the number of tantalum-182 atoms at any time and the thermal cross sections of both nuclides, assuming that the number of tantalum-181 atoms does not change appreciably.

In-pile times of 73 and 149 days (five and ten cycles) for the 1.5 and 3.0 w/o conversions, respectively, were obtained with an unperturbed thermal-neutron-flux density of 3×10^{14} nv. Increasing the unperturbed thermal flux will, of course, decrease the cycle time as seen in Table A-1 below:

TABLE A-1. CYCLES REQUIRED AT VARIOUS UNPERTURBED THERMAL-NEUTRON-FLUX DENSITIES FOR 40-MEGAWATT OPERATION IN THE MTR

Thermal-Neutron Flux, nv	Exposure for Indicated Conversion			
	1.5 w/o Conversion		3.0 w/o Conversion	
	Days	MTR Cycles	Days	MTR Cycles
3.0×10^{14}	73	5	149	10
3.5×10^{14}	63	4	128	9
4.0×10^{14}	55	4	112	8
4.5×10^{14}	49	3	100	7
5.0×10^{14}	44	3	90	6