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The following report no new developments:

General Electric, ANP

Oak Ridge National Laboratory

The following have not been heard from:

Ames Laboratory

Brookhaven National Laboratory

Bureau of Mines, Albany

Du Pont Company

Knolls Atomic Power Laboratory

Los Alamos Scientific Laboratory

Mallinckrodt Chemical Works

National Lead Company

825 002

ARGONNE NATIONAL LABORATORY - Reported by L. T. Lloyd

Uranium-Niobium Alloys

Phase Diagram: The investigation of the uranium-niobium equilibrium phase diagram has been extended to 10 w/o Nb. The eutectoid transformation occurs at 634 ± 2°C, and its composition is estimated at 8 ± 1 w/o Nb; characteristic lamellar structures are produced by this reaction. (Dwight)

Uranium-Plutonium Alloys

Irradiation: Extrusion has been found by Kelman to be a successful means of fabricating the higher Pu alloys of uranium. A series of 10, 15 and 20 w/o extruded specimens has been irradiated and examined. The 10 and 15 w/o specimens showed radical changes, apparently due to extremely coarse grain. The 20 w/o as-extruded specimen, however, remained smooth and relatively unchanged at burnups up to .60%. Gross length change was 5.7%, and there was some overall warp. A heat treated specimen of the same material (645°C, with one hour arrest at 500°C), however, developed roughening in irradiation which characteristically accompanies a coarse grain structure. (Paine)

Uranium-Silicon Alloys

Irradiation: Two additional specimens of uranium-3.8 w/o Si alloy, homogenized at 800°C for 2-3/4 days by the supplier, Nuclear Metals, Inc, have been examined after burnups of 0.12% and 0.33% of uranium atoms. The length changes amounted to 2.1% and -1.4%, respectively. The specimens appear to be quite stable under irradiation, though on the specimen having the higher burnup there are some traces of surface cracks. (Paine)

Corrosion: As part of the program of testing material fabricated by Nuclear Metals, Inc., open-ended sections of two Zircaloy II clad co-extruded uranium-3.8 w/o Si alloy rods were tested at 290°C in degassed,

distilled water. The alloy had been heated at 800°C for seven days prior to extrusion. After about 10 days of testing, both core and bond appear good. The corrosion rates are comparable with as cast material.

One 10% enriched uranium-3.8 w/o Si alloy cast at Argonne and epsilonized at 825°C for two hours was tested bare after irradiation; indicated uranium burnup was 0.71%. Results of corrosion tests at 260°C and 290°C are listed in Table I. Although corrosion behavior was only fair, it was approximately the same as shown by the alloy (apparently not too well made) before irradiation. It appears that irradiation is not harmful to the corrosion resistance of this alloy. (Greenberg and Draley)

Table I

Corrosion of Irradiated U-3.8 w/o Si Alloy (BB-3)

Test Temp. (°C)	Time (Hrs.)	Corr. Rate* (mcd)	Appearance After Test
260+	24	0.13≠	No apparent change except for small silver colored spot near one end.
290	87	31.9**	Loose particles in water and on sample. Relatively large piece cracked off one end. Other end roughtned and has hole in cylindrical surface.

* Pre-irradiation corrosion rate at 290°C was 15.7 mcd after 88 hours.
\+ Result at 260°C previously reported.
≠ Wt. change small enough to have been balance error.
** Based on major piece.

Uranium-Niobium-Tin Alloys

Corrosion: Two 10% enriched samples of uranium-3 w/o Nb-0.48 w/o Sn alloy cast at Argonne were tested in degassed, distilled water at 260°C for 70 hours. The samples were water quenched after 4 minutes at 800°C and aged at 350°C for 6 hours. Indicated uranium burnup were 0.58 a/o and 0.19 a/o respectively. Pre-irradiation corrosion rates at

290°C were 28.3 mg/cm^2/day (mcd) and 31.3 mcd, respectively, for 88 hour tests. After irradiation the samples were roughened and warped. Growth rates were 15 and 38 μin./in./ppm burnup, respectively. The samples were completely disintegrated in the corrosion test. (Greenberg and Draley)

Uranium-Zirconium-Niobium Alloys

Irradiation: In investigating the corrosion properties of uranium-5 w/o Zr-1.5 w/o Nb alloy it was observed that these are evidently impaired by irradiation. It is not evident whether the impairment is the result of structural changes in the specimen, or whether it is due to the roughening and surface changes produced by irradiation. Specimens are now available which may resolve this uncertainty. In the irradiated group some were heavily restrained by close-fitting cladding, while others were irradiated under identical conditions without jackets. Upon removal of the jackets the clad specimens appear to have retained their original smoothness at burnups ranging up to 0.33%; the unclad specimens have the characteristic irradiation roughening. The entire group will be tested for corrosion resistance. (Paine)

Corrosion: Work designed to correlate corrosion damage and hydrogen absorption in uranium-5 w/o Zr-1.5 w/o Nb alloy has continued. As previously reported, a relatively high hydrogen content, e.g. 35 ppm as found in a sample quenched from salt at 850°C, is not detrimental to corrosion resistance in the as quenched condition. This is perhaps because of the uniform distribution of the hydrogen.

A group of samples which had been quenched from salt were aged for various times at 400°C and tested for 71 hours in degassed distilled water at 290°C. The aging times bracketed that for optimum corrosion resistance, of a vacuum treated sample, i.e., approximately two hours. The results are shown in Table II.

Table II

Effect of Aging on Corrosion Resistance and Hydrogen Absorption of U-5 w/o Zr - 1-1/2 w/o Nb Alloy; L-170B - as cast - Quenched from Salt at 805°C

Sample	Aging Time at 400°C (Hrs)	Corr. Rate (mcd)	Hydrogen Content of Alloy after Test, (ppm by Weight)	Appearance of Sample After Test
1	0	13.5	60	Dull, pitted.
2	1	337*	832	Sample cracked into irregularly shaped piece and slurry. This piece in good condition.
3	2	450*	1220	Same as above.
4	3	616*	1340	Same as above.
5	4	-	--	Completely disintegrated.

* Based on remaining piece.

The as quenched sample acted in the normal expected fashion; the aged samples corroded in a completely different fashion than do vacuum heat treated and similarly aged metal.

It is thought that the aging process causes the hydrogen to concentrate, perhaps at grain boundaries. This concentration may act as a nucleus for further rapid hydrogen absorption. It is significant that this is the first instance in which very small pieces of metal close to cracked surfaces have been analyzed. Very high hydrogen contents were found, as had been previously postulated.

Unfortunately, the remnants of the corrosion test were not large enough to permit both hydrogen analysis and metallography. The series of heat treatments will be repeated and the samples examined before corrosion testing.

Further study is being made of samples prepared at Sylvania by

powder metallurgy techniques. Preliminary results indicate that during heat treatment, longer time in the gamma region is required than for cast metal. These results also indicate that mild aging improves corrosion resistance as for conventionally prepared metal.

Two completely clad plates (AH-10-2 and AH-12-2) were tested for corrosion after irradiation; indicated burnups were 0.021% and 0.079% respectively. Both samples were from the same standard type clad plate (#64). Exposed core ends and edges were weld clad with Zircaloy II. The samples were water quenched after 15 minutes at 850°C and aged at 425°C for 45 minutes. After irradiation, a 1/8" diameter hole was drilled through the clad to the core of each sample.

Sample AH-10-1 was tested in degassed, distilled water for 58 hours between 240°C and 275°C. At the end of this period there was loose oxide at the hole but no swelling.

Sample AH-10-2 was similarly tested for 66 hours at 290°C. At the end of this period the undrilled cover plate was almost completely severed from the assembly (attached at one corner). The drilled cover plate was not as completely separated. There was no swelling of the cover plate at the drilled hole. All failures were along welds. The core was swelled to at least twice its original thickness and was apparently completely converted to oxide.

A sample duplicate in very respect to AH-10-2 and AH-12-2 except that it was not irradiated was tested at 290°C for 95 hours. There was no change except difference in color between core and clad. (Greenberg and Draley)

ARMOUR RESEARCH FOUNDATION - Reported by R. J. Van Thyne

Isothermal Transformation Characteristics of Uranium-Base Alloys

Previous reports in the Uranium Alloy Newsletters have described transformation kinetics for uranium-rich U-Mo, U-Mo-Nb, U-Mo-Pt, and U-Nb alloys; this work has now been completed. From this information certain generalizations can be drawn. It has been shown (Newsletter No. 11) that for any given alloy and temperature level the observed times for transformation vary depending upon the techniques employed. The reasons for this can be rationalized. However, using any given technique, trends due to alloying are similar.

TTT diagrams have not been presented previously for the 10 and 20 w/o niobium alloys. The effect of niobium content upon stabilizing the gamma phase is clearly demonstrated by Figure 1. Transformation is more sluggish in the U-20 w/o Nb composition. The data points were taken from curves of room temperature electrical resistivity vs. annealing time and represent times required to produce an initial decrease in resistivity. Other techniques generally yielded similar results. However, some difficulties arose in the determination of kinetics for the 10 w/o niobium alloy at temperatures of 400°C and below. For this alloy a good correlation between metallography and resistivity results was not obtained. Ambiguous microstructures were sometimes observed. Additional work is now being done in this region by WAPD.

Resistivity findings for all alloys show that after annealing at 500°C for 1000 hours, transformation was complete only for the U-5.4 w/o Mo, U-8 w/o Mo-1 w/o Pt, and U-10 w/o Nb alloys. At the lower temperatures transformation was not complete within 1000 hours for any of the compositions. TTT diagrams for the U-Mo and U-Mo-X

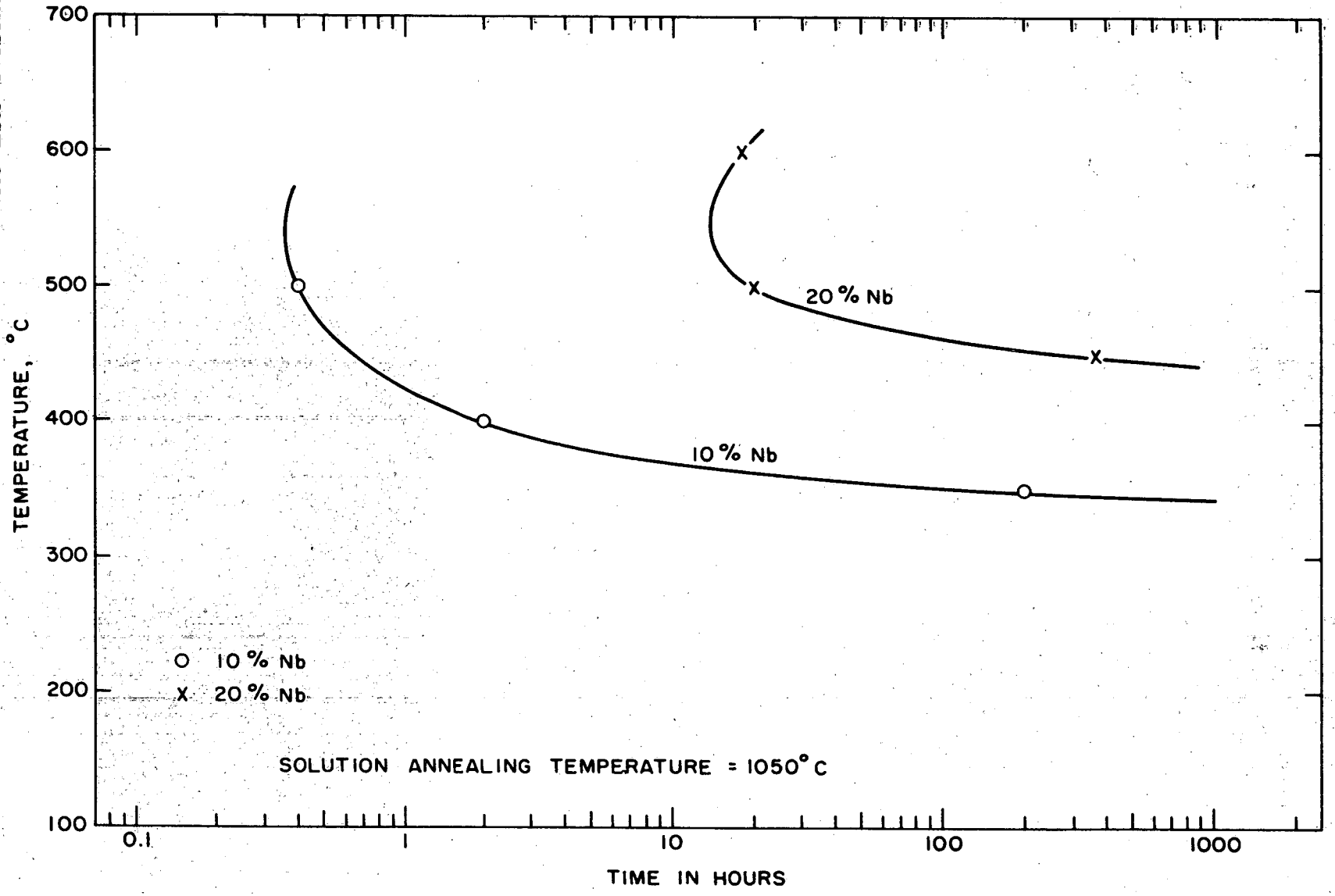

FIG. 1 - TTT DIAGRAMS ILLUSTRATING INITIAL RESISTIVITY DECREASE FOR U-Nb ALLOYS

(Armour Research Foundation)

alloys have been presented previously (Newsletter No. 11). Comparisons of alloy kinetics are easily made. Resistivity results presented in Table I probably represent the most useful data for all alloys.

At temperatures below 400°C the gamma solid solution is metastable for the longest times in the U-8 w/o Mo-3 w/o Nb and U-20 w/o Nb alloys. Using all techniques, no transformation was detected at 350°C in either composition. A trace of decomposition was observed metallographically in ther ternary alloy upon a 1000 hour anneal at 400°C. With a similar treatment no decomposition was noted in the microstructure of the 20 w/o niobium composition. Using X-ray diffraction, only the metastable gamma was detected in either composition at 400°C. Similarly, hardness and resistivity results indicate the superior stability of these alloys.

Table I

Transformation Times Determined by Resistometric Technique to Produce Initial Transformation in Uranium-Base Alloys

Alloy (wt %)	Time (Hours) to Produce Resistivity Decrease			
	500°C	450°C	400°C	350°C
5.4 Mo	0.8		15	150
8 Mo	4		150	500
10 Mo	25		60	500
12 Mo	60		200	2000
8 Mo-1 Pt	80		200	800
8 Mo-1.5 Nb	30		200	1500
8 Mo-3 Nb	6	100	1500	2000
10 Nb	0.4		2	200
20 Nb	20	350	1000	1000

BATTELLE MEMORIAL INSTITUTE - Reported by H. A. Saller

Phase Diagrams

The stability and existence range of the epsilon phase of the Zr-U system are under investigation.

Consumable arc-melted alloys containing 40 to 60 w/o uranium are being prepared. Both crystal-bar and sponge zirconium are to be used

as the base materials.

It has been found possible to form the epsilon phase from zirconium and uranium powders by annealing below 600°C. Uranium hydride and zirconium powders were mixed and compacted at room temperature, dehydrided below 1000°F in a vacuum furnace, and then annealed 17 days at 1000°F. The results of X-ray diffraction examination are shown in Table I. The ability to form epsilon from the elemental powders without first forming the high-temperature beta zirconium-gamma uranium phase shows that epsilon is not a transition phase in the decomposition of beta, but is a stable phase.

Alloys of base 50 w/o uranium composition and with 0 to 20 a/o oxygen additions have been examined metallographically and by X-ray diffraction techniques after having been annealed 100 hr at 550°C. The same trend is shown by both methods of examination. The X-ray diffraction results are given in Table I. It is seen that with increasing oxygen content, the epsilon phase disappears, being replaced by the zirconium and uranium alpha phases. Lattice parameter measurements, particularly of the alpha zirconium, are planned.

Additional data have been obtained on the test 50 w/o uranium alloy in the high-temperature camera. This sample has been examined at 5°C ± 1°C intervals from 585 to 610°C. Phases were either epsilon or beta, and no two-phase region of epsilon-plus-beta was identified.

In order to determine the composition limits of the alpha-plus-beta zirconium phase region, a quantitative study of the phases present in zirconium-rich Zr-U alloys as a function of oxygen and nitrogen content is planned.

A preliminary mathematical analysis of experimental requirements for the study is being made. Upon the basis of this analysis, alloys will be prepared and heat treated for quantitative measurements.

Table I

Results of X-Ray Diffraction Examination
of Zirconium-Uranium Alloys

Composition	Heat Treatment	Intensities[a] of Phase Patterns		
		ϵ	αZr	αU
Powder specimen, Zr-50 w/o U	17 days at 1000°F	S	M	F
Zr-50 w/o U Base	100 hr at 550°C	S	0	0
Base Alloy with 2 a/o O_2	Ditto	S	MF	0
Base Alloy with 5 a/o O_2	Ditto	S	M	F
Base Alloy with 10 a/o O_2	Ditto	M	S	MF
Base Alloy with 15 a/o O_2	Ditto	0 to MF	S	MF to M
Base Alloy with 20 a/o O_2	Ditto	0 to F	MS	MF

(a) S = Strong
F = Faint
M = Medium

Heat Treatment

Zirconium-Uranium Alloys. A series of Zr-U alloys containing from 7 to 70 w/o uranium are being prepared for heat-treatment studies. The effect of oxygen on the heat-treatment behavior of these alloys is also being examined. Oxygen contents will be varied from 300 to 3000 parts per million. Alloys containing 7 to 15 w/o uranium and 900 to 3000 ppm oxygen are now available for study.

Initial attempts to vary the structure of alloys containing 7 to 15 w/o uranium and 900 to 3000 ppm oxygen have been unsuccessful; the familiar Widmanstatten structure being produced in every case. A preliminary examination of end-quench bars shows that these alloys contain equilibrium alpha zirconium at 900 C. This effect is caused by the oxygen present. Hardness traverses on these bars indicate that hardening effects due to quenching are negligible at 7 per cent uranium,

but increase to an appreciable extent in the 15 per cent uranium alloy. A hardness increase of 20 DPH was observed near the quenched end of a 7 w/o uranium alloy; a hardness increase of over 70 DPH was observed near the quenched end of a 15 w/o uranium alloy.

The structure observed in the end-quench bars will be used as a guide to future attempts to vary the structure of these alloys.

Alloys containing 20 to 70 w/o uranium are being prepared for an investigation of the kinetics of the beta zirconium-to-epsilon decomposition. Experimental work, aimed at achieving a description of this decomposition, will begin when the complete series of alloys is available. The alloys are being consumable arc melted from biscuit uranium and crystal-bar zirconium.

A study of the 50 w/o uranium alloy, to correlate resistance and micro-structure, has begun. A resistivity specimen has been machined from a 50 w/o uranium alloy rod after it was annealed at 800°C and water quenched. The isothermal transformation of retained beta to epsilon is to be followed by resistance measurements. Equipment is being set up for this purpose.

An investigation of the ternary system, zirconium-uranium-oxygen, has disclosed that the addition of oxygen to Zr-U alloys causes the formation of a two-phase region where oxygen-rich alpha zirconium and a uranium-rich phase are in equilibrium. Because alpha zirconium has desirable corrosion properties, it is of interest to determine whether favorable properties can be produced by proper treatment of these alloys.

Three 100-g ingots containing uranium, zirconium, and oxygen have been prepared. Each ingot contains zirconium and oxygen in the ratio of four atoms to one, so that the phases present at room temperature are alpha zirconium and alpha uranium. The three ingots were designed

to contain 20, 40, and 60 a/o uranium. X-ray diffraction measurements on the as-cast alloys show that the room temperature constituents are as predicted; namely, alpha zirconium and alpha uranium.

Future work will include heat treatments to vary the hardness and structure of the alloys, X-ray studies to determine the nature of the constituents present, and corrosion tests.

Intermediate or Delta-Phase Alloys. The investigation of the phase relationships between the delta phases of the U-Mo and U-Ti systems has continued.

Alloys prepared for this investigation range in composition from U-31.5 a/o Mo to U-34 a/o Ti. Differential thermal data indicate a transformation in these alloys which increases in temperature with increasing titanium content from about 590 to 650°C. Molybdenum-rich alloys containing less than 2 a/o titanium showed no thermal arrest on heating, the stabilized gamma phase evidently never having transformed during the previous homogenization and furnace-cooling treatment. Molybdenum additions to the U-Ti delta phase, which transforms to gamma at about 900°C, cause a gradual decrease in the gamma transformation temperature.

A series of samples annealed at 850°C are now being prepared for metallographic examination and additional samples are being prepared for both metallographic and X-ray studies.

Radiation Damage Studies

As a part of the over-all program concerned with the study of U-Zr alloys that is being sponsored by the Reactor Development Division of the Atomic Energy Commission, an investigation of their radiation stability has been initiated. The first step in this investigation will consist of post-irradiation studies on specimens of both 22 and 40 w/o Zr-U alloys that have been irradiated by Knolls Atomic Power Laboratory.

It is planned to use these specimens in an attempt to determine the effect of post-irradiation heat treatment on the release of fission products and on other properties such as microstructure, hardness, and density. In addition to these experiments, normal post-irradiation examinations will be performed on the alloys as they are received from KAPL.

Physical Properties

Alloys for High-Temperature Service. The objective of this program is to develop uranium alloys for reactor operation at temperatures above 700°C. At present, interest is centered on alloys containing 3.5 to 15 w/o molybdenum and 3 to 20 w/o zirconium.

Specimens of U-Zr alloys were subjected to heat treatments designed to produce fine-grained random structures. A metallographic examination was made of specimens of each composition, and two of the better heat treatments were selected. At present, dilation and hot-hardness specimens have been prepared and heat treated and are undergoing examination. Specimens have also been prepared for evaluation by thermal stress cycling tests.

U-Mo alloys have also been prepared and subjected to heat treatments designed to yield fully retained and fully transformed gamma structures. These alloys are undergoing similar studies. Ultimately, one or more of the better-behaved alloys of each system will be selected for irradiation-damage tests.

Uranium-Titanium Alloys. A number of alloys of uranium and titanium have been made by consumable-electrode arc melting. Fabrication and testing are underway.

Uranium Alloys for Slug-Type Fuel Elements. A series of alloys are being studied in the hope that a comparison of their irradiation behavior and their physical and mechanical properties will result in a better

understanding of the properties necessary to resist damage. The alloys being studied are induction-melted uranium, arc-melted uranium, uranium plus 0.35 a/o chromium, uranium plus 1.5 a/o silicon, and uranium plus 0.5 a/o titanium. The major differences between these alloys are to be found in their mechanical properties, hardness, grain size, and microstructures. The mechanical properties have been reported previously and show that the silicon alloy is the hardest, strongest, and least ductile; arc-melted uranium has the lowest strength and lowest hardness. The low ductility of the U-Si alloy seems to be significant, but differences between the ductilities of the other alloys may be the result of unknown factors.

Thermal-expansion measurements have indicated no significant differences in this property in these alloys. In the range between 20 and 600°C, the thermal-expansion coefficient in microinches per inch per degree Centigrade was 19 for arc-melted uranium, 16 for the chromium alloy, 19 for the silicon alloy, and 18 for the titanium alloy. The range of values obtained for each alloy was greater than the differences shown above for different alloys.

End-quench tests have been performed on each alloy. Specimens 3 in. long and 1/2 in. in diameter were heated to 800°C for 15 min. and end quenched over a 3/8 in. water jet. Vickers hardness traverses on these bars disclosed no trend in hardness that could be attributed to the different rates of cooling throughout the bars. Local hardness variations of as much as 100 DPH were observed and were attributed to segregation of impurities and alloying elements. Likewise, grain-size differences throughout the bars showed no trends that could be attributed to the quenching operation. Grains up to 1 mm in diameter were observed in the arc-melted uranium, the induction-melted uranium, and

the 1.5 a/o silicon alloy. Grains up to 0.25 mm in diameter were observed in the 0.35 a/o chromium alloy and the 0.5 a/o titanium alloy. Aging the quenched bars for 2 hrs. at 400°C produced no measurable changes in hardness or grain size. No cracking occurred in any bar. These tests show that air cooling and water quenching from 800°C produce identical results as far as hardness and grain size are concerned.

While it may be that these alloys actually exhibit heat-treatment effects, the alloy content is so small that any effects must necessarily be small. The possibility exists that small heat-treatment effects will be masked by and confused with variations in the base metal. Attempts to heat treat these alloys will be abandoned until such time as heat-treatment effects can be clearly distinguished from random variations in the base metal. Attempts will be made to deduce the cause of these variations.

Processing

Preparation and Properties of Low-Melting Alloys. A program aimed at the development of low-melting alloys for possible use as power reactor fuel elements is in progress.

On the basis of recently completed thermal-cycling studies, a number of low-melting alloys were selected for further study. The alloys listed in the following tabulation will be thermally stressed and examined.

Composition	Approximate Melting Point
U-5.97 w/o Cr-2.69 w/o Th	1840°F
U-5.0 w/o Cr-1.0 w/o Mo	1680°F
U-4.82 w/o Cr-0.72 w/o Si	1650°F
U-2.56 w/o Cr-2.26 w/o V	1785°F
U-5.0 w/o Cr-1.0 w/o Ge	1650°F

The objective in thermally stressing the alloys is to reproduce, in model specimens, stress patterns similar to those present in a reactor fuel element. To do this, it is necessary to duplicate the geometry,

temperature pattern, and linear power generation rate in a scale-size model.

Linear power generation can be simulated by the use of surface induction heating effects. The temperature pattern in the fuel can be reproduced by introducing a coolant flow through the interior of the heated cylindrical model. For handling ease and in order to avoid corrosion problems, helium under pressure appears to fulfill the coolant requirements.

The specimens will be in the form of hollow cylinders 3 in. long, 1.50 in. inside diameter, and 0.32 in. thick. Requirements of the testing equipment are that a temperature gradient of approximately 40°F be established between inside and outside cylinder walls at the mean outside wall temperature of 840°F. The gradient is produced by combined surface heating and internal gas cooling of the test specimens.

After screening of alloys by stress cycling, those appearing stable will have physical properties determined. The ultimate goal of the project is to produce one or more promising alloys for irradiation-damage studies.

Bond Behavior of Uranium-Molybdenum Alloys. The nature of the bond interface between a Zircaloy 2 clad and U-12 w/o Mo core and its corrosion behavior in high-temperature water was investigated. The effect of transition layers on the corrosion properties of the interface was also studied.

In addition, an exploratory investigation was made of the effect of barrier layers on the diffusion rate between Zircaloy 2 and epsilonized U-3.8 w/o Si. Reaction rates between core and barrier and clad and barrier were determined for barrier layers of copper, molybdenum, niobium, platinum, and tantalum. Tantalum was the most effective barrier studied, and platinum was the least effective.

Preparation of Aluminum-Uranium Alloys. Al-U alloys in the composition range of 14 to 18 w/o uranium are the subject of a current study. The program of research is concerned with the accumulation of information pertinent to the melting and casting of these alloys. Included in the program are the determination of the cooling rates between the liquidus and solidus temperatures necessary to produce sound homogeneous castings and the confirmation of certain areas of the Al-U phase diagram.

A casting technique has been developed that produces sound homogeneous castings. This consists of pouring the 16 w/o alloy at 1300°F into a steel mold preheated to between 1100 and 1200°F. The steel mold is positioned in a tank and after pouring, water is introduced into the tank. The water rises around the mold freezing the alloy rapidly and allowing the casting to solidify progressively from the bottom to top.

Efforts to confirm the Al-U phase diagram are continuing, and studies of different cooling rates through the liquidus-solidus temperatures and their effects on segregation have been initiated.

Preparation of Uranium-Zirconium Alloys. The development of consumable-electrode arc-melting techniques for the production of U-Zr alloys within specified compositional limits is in progress. These alloys are required for a thorough study of this system in the composition range between 0 and 60 w/o uranium.

A series of alloys to be used in the study of the epsilon region has been prepared. These alloys contain 40 to 60 w/o uranium in 5 w/o intervals with iodide zirconium as the base material. All of this series has been double melted into 2-1/2-in.-diameter ingots and surface conditioning is in progress. Samples for chemical analysis from each ingot have been obtained and the analyses are in progress.

Alloys containing 22, 40, 50, and 60 w/o uranium in Grade A

zirconium sponge have been double melted. Because of a need for a large quantity of both the 50 and 60 w/o uranium alloys, ingots of these compositions were made by melting four 2-1/2-in.-diameter ingots into one ingot having a diameter of 4 in. These ingots were surface conditioned by normal techniques, but, as a result of this treatment, cracking developed near the ends of the ingots. It is felt that the sensitivity of the alloys in this range to heat treatment is responsible for this cracking. In the future, ingots with this uranium content will not be surface welded on the ends, but will be machined to the desired surface finish. Chemical analysis of these alloys is in progress.

An ample supply of sponge zirconium has been treated to raise the oxygen level to 2 w/o. This material has been mixed with enough untreated sponge containing the normal oxygen level of 740 ppm to give a nominal oxygen content in the zirconium of 3000 ppm. This material has been made into electrodes containing 22 and 50 w/o uranium and melting is in progress. Chemical analysis and hardness surveys will be made after double melting is complete.

HANFORD ATOMIC PRODUCTS OPERATION - Reported by S. H. Bush

Structural Stability of Irradiated Uranium Alloys

A study to determine the dimensional stability and mechanical and physical properties of irradiated uranium alloys in essentially isotropic modifications is being pursued. The ϵ phases of U-Si and U-Zr have been selected for the initial work.

In an evaluation of various physical properties as criteria of phase changes, dilatrometry has been found to be of no value. Metallography has confirmed the absence of ϵ (U_3Si) in a U-23 a/o Si (3.7 w/o Si) specimen heat-treated above the peritectoid temperature. This specimen, however, showed the same thermal expansion characteristics in the range 25 to 800°C

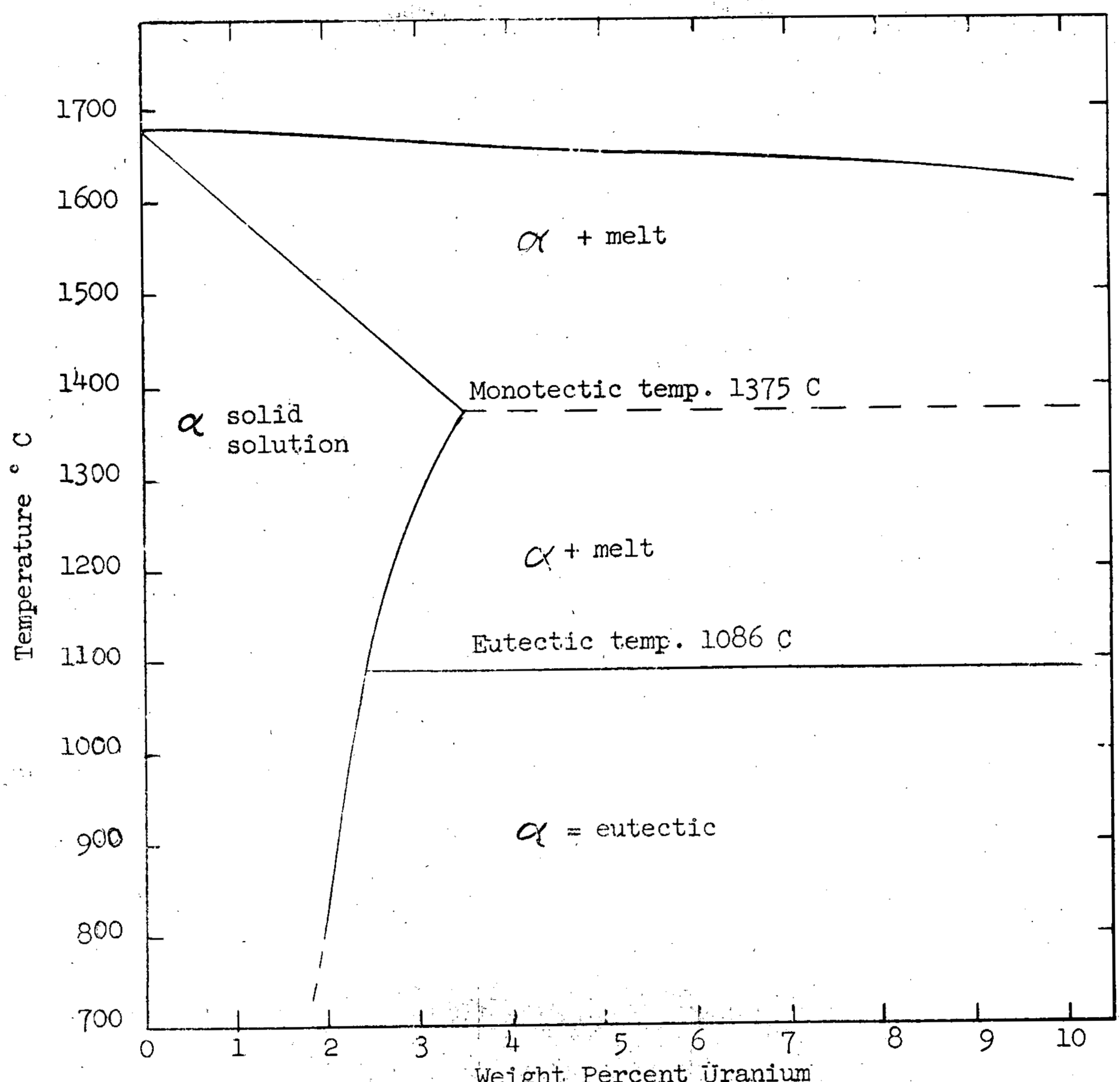

Figure 1. Thorium-rich end of the Thorium-Uranium System as Determined Metallographically.

HANFORD ATOMIC PRODUCTS OPERATION - Fig. 1

as another specimen which contained large amounts of phase. Other properties will be examined. (L.F.Kendall - R.G.Wheeler)

Mechanical Properties of Irradiated Uranium Alloys

The tensile properties of irradiated uranium alloys are to be determined to evaluate the effects of nominal alloying additions to uranium. Fabrication of specimens of U-Ti and U-Cr containing 0.5 to 2.0 a/o additions and of container assemblies is nearly complete. It is hoped that these will be charged into the MTR during the next quarter. Tensile specimens of the epsilonized U-Si alloy are also being fabricated for irradiation in the MTR. (L.F.Kendall - R.G.Wheeler)

Uranium-Thorium Alloys

Th-U alloys are being investigated as possible reactor fuel materials. The thorium-rich end of the thorium-uranium system has been studied metallographically with alloys prepared from normal reactor grade thorium and uranium and containing a nominal two to ten w/o uranium. The metallographic study has been made difficult by impurity inclusions present in the thorium and contamination resulting from the alloy preparation. Heat-tinting of metallographic specimens has been found to delineate the Th-U eutectic phase. The identity of this phase has been further verified by an in-pile autoradiographic technique using nuclear emulsion film sensitive to fission fragments. This technique has worked successfully in identifying the uranium bearing eutectic in a coarse structure of the higher percentage uranium alloys, but has not been successful in lower percentage uranium alloys of a finer structure because of a lack of resolution. However, a solid solution of uranium in thorium, as indicated in Figure 1, has been determined by these metallographic studies. (J.W.Goffard)

Uranium-Thorium-Zirconium Alloys

Ternary alloys of Th-U-Zr have been prepared and examined to determine if zirconium dissolved in the thorium will lower the solubility of uranium in thorium. It does not appear that additions of four to twenty w/o zirconium to alloys of thorium two w/o uranium decrease the solid solubility of uranium in thorium. Samples of these alloys submitted for chemical analysis have proven very difficult to dissolve in hydrochloric, sulfuric, nitric, hydrofluoric, or combinations of these acids. Since this suggests a high corrosion resistance of these alloys, samples have been water autoclaved with the following results.

Sample	Initial Wt.	Final Wt.	% Loss	Test Condition & Time
Thorium				
1. Alloy base, wrought HAPO thorium slugs	32.7 g	0 g	100	Distilled H_2O, 240°C, about 4 hrs
2. Ames as-cast billet	16.8	5.0	70	Distilled H_2O, 240°C, 16 hrs
Ternaries				
1. Th-2 w/o U-6 w/o Zr	58.7	0	100	Distilled H_2O, 240°C about 6 hrs
2. Th-2 w/o U-10 w/o Zr	99.4	92.0	7.5	Distilled H_2O, 240°C, 16 hours
3. Th-2 w/o U-10 w/o Zr	92.0	0	100	Distilled H_2O, 340°C, about 9 hrs.

The binary Th 2 to 10 w/o uranium and ternary Th 2 w/o U, 4 to 20 w/o Zr have been hot forged and rolled with poor results. Forging was not successful until temperatures in excess of 800°C were used. This is believed to result from "poor metal quality" of the alloy samples, i.e., detrimental contamination resulting from the alloy preparation.

A ternary alloy of Th - 2 w/o U - 1 w/o Ti has been prepared and heat treated in an attempt to remove uranium from solution in thorium by

the formation of U_2Ti. This has not been successful. Forging and rolling of this alloy also has been poor because of the suspected "poor metal quality" factor. (J.W.Goffard)

NATIONAL BUREAU OF STANDARDS - Reported by L. L. Wyman

Uranium-Platinum System

Subsequent data based on microscopic, thermal, X-ray, and microhardness analysis has shown that there is no appreciable solid solution at UPt_3. Microhardness determinations substantiate the previous conclusions on the constitution of this system based on thermal, X-ray and microscopic analysis.

Uranium-Ruthenium System

High temperature thermal analysis of the ruthenium-uranium alloys in the composition range 57.1 w/o - 71.0 w/o of ruthenium indicate that a reaction horizontal occurs at 1575°C ± 10°C. In view of this, and the fact that previous microscopic work indicated a two phase region in the composition range between Ru_3U and the terminal solid solution, it appears that Ru_3U is formed peritectically and that the liquidus rises from 1575°C to the melting point of ruthenium.

Uranium-Iridium

Seven iridium-uranium alloys in the composition range 0.25-20 w/o of iridium were prepared by induction melting in beryllia crucibles. A violent exothermic reaction which raised the temperature of the melt some several hundred degrees was observed in the 0.25 w/o of iridium alloy. Subsequent to this reaction, a vigorous evolution of a vapor phase occurred which tended to eject the melt from the crucible. This reaction became increasingly violent with increasing amounts of iridium.

Uranium-Rhodium

Nine uranium-rhodium alloys in the composition range 0.25-40 w/o

of rhodium were prepared by induction melting in beryllia crucibles, and a 50 and 60 w/o of rhodium alloys by arc melting.

The alloys in the composition 0.25-30 w/o of rhodium showed no metal-refractory reaction. However, as there appeared to be some such reaction in the case of the 40 w/o of rhodium alloy, the higher alloys were arc-melted.

Uranium-Palladium System

The thermal, microscopic, and X-ray data show that the two reaction horizontals reported at 990°C and 952°C in the uranium-palladium system extend to at least 20 w/o of palladium. The gamma-beta transformation was lowered from 770°C to 738°C and the beta-alpha transformation was lowered from 660 to 644°C. On both heating and cooling these alloys displayed a hysteresis in the allotropic transformation points similar to that reported in the case of the platinum-uranium system. X-ray and microscopic data showed the alpha and beta solubility of palladium to be less than 0.2 w/o and the indications are that the solubility of palladium in gamma uranium is considerably less than that of platinum, as might well be expected from considerations of the electronic configuration. Microhardness data indicates the existence of a reaction horizontal at 952°C, confirmatory to the thermal data. Microscopic and X-ray data for the "as-cast" alloys in the composition range 0.45-40 w/o of palladium indicate that the uranium-rich phase is present in all the alloys in this composition range.

NORTH AMERICAN AVIATION, INC. -- Reported by B. R. Hayward

The current work at North American Aviation consists of the evaluation of uranium alloys for use as long life and high operating temperature experimental fuel materials in the sodium reactor experiment (SRE). Since none of the alloys have been made on a larger than

laboratory scale, the present phase of the program is to establish fabrication feasibility on a semi-production scale. After satisfactory evaluation on such items as alloy content and homogeneity, uniform microstructure, density, size and surface specifications, etc. the slightly enriched uranium alloy (Th-U uses enriched U) slugs will be fabricated by the same equipment and on the same scale. The alloy slugs currently being fabricated for feasibility include:

(1) U-Mo, 1.2 w/o Mo powder compacted, 3/4" x 1"

(2) U-Si, 0.5 w/o Si powder compacted, 3/4" x 1"

(3) Th-U, 5.4% U extruded, 3/4" x 6"

(4) U-Mo, 1.2 w/o Mo cast, 3/4" x 6"

(5) U-Si, 0.5 w/o Si cast, 3/4" x 6"

(6) U-Zr, 2.0 w/o Zr, cast 3/4" x 6".

Partial shipments of items (1) and (6) have been received.

The choice of alloy element is limited on a nuclear basis to about 5 a/o and to low cross section materials. The powder compacting fabrication technique and small l/d ratio was chosen for dimensional stability to thermal cycling (previously reported) and potential use of mass production methods. It is anticipated that the as-sintered slugs will be satisfactory in size for direct use. The casting technique was chosen on the basis of excellent performance of cast U-Zr alloys reported by Argonne. This process also presents possibilities for high material efficiency, low inventory, remote refabrication, low scrap recovery, and potential low cost fuel slugs. It also allows the production of some alloys which could not be made by working and heat treating techniques. It is planned ultimately to cast the slugs to size except for length.

NUCLEAR METALS, INC. - Reported by S. Isserow

Uranium - 1.5 w/o Niobium - 5 w/o Zirconium Alloy: Dilatometric Studies and Thermal Analysis

The temperature and magnitude of transformation and the thermal expansion of the U-1.5 w/o Nb-5 w/o Zr alloy were determined. Four samples (graphite melted material, extruded at 1150, 1200, 1250, and 1300°F) were cycled in a conventional quartz-tube push-rod type dilatometer. A dilatometer cycle consisted of heating and cooling from room temperature to 800°C in a purified argon atmosphere. The heating rate was approximately 4.4°C/min. and the cooling rate approximately 1.2°C/min

The averaged mean coefficient of thermal expansion of the 4 samples from 25° to 800°C was 24.6 x 10^{-6}/°C on heating and 23.5 x 10^{-6}/°C on cooling and included the effect of the allotropic transformation. From 25°C to 600°C the coefficient was 18.4 x 10^{-6}/°C on heating and 17.2 x 10^{-6}/°C on cooling. For the high temperature phase (700°C to 800°C) the mean coefficient was 17.8 x 10^{-6}/°C on heating and 22.0 x 10^{-6}/°C on cooling.

The transformation is accompanied by an expansion on heating (0.54% change in sample length) and a contraction on cooling (0.44% change in sample length). The exact starting temperature of the transformation on heating is uncertain but appears to be close to 625°C, ending at 678°C. On cooling, the transformation initiated at 650°C and finished at 610°C.

Thermal analysis data indicate the transformation starts at 634°C on hea ing and at 659°C on cooling. (H.F.Sawyer)

Uranium-Silicon Epsilon Alloy

The corrosion of bare U-Si epsilon alloy is now better understood. More information is available on the corrosion of extruded material.

The simultaneous pouring of several castings is now standard except for experimental individual castings of special composition. The effect of impurities such as those in commercial silicon is of particular interest since they may broaden the composition range of the epsilon phase. The establishment of the optimum silicon content has been delayed by difficulties in the usual method of analysis for silicon. It is now established that this method, involving direct solution of the metal in acid, gives low results for the silicon content of non-epsilon material. Oxidation of the sample before dissolution eliminates the effect of the phases on the apparent silicon content. Epsilonization by heat treatment for a week at 800°C is still standard. Essentially the same results are obtained at 825°C. At both temperatures, the inadequacy of shorter times is observed metallographically and confirmed by corrosion data in 650°F water (but not in 500°F water). Heat treatment after extrusion (or a double extrusion scheme involving a second heat treatment before the final coextrusion) cannot be depended upon to eliminate free uranium and U_3Si_2 which are separated by epsilon.

The preparation of material resistant to 500°F water presents little difficulty but material adequate for 650°F service is not being obtained reproducibly yet. Understanding of the metallographic factors determining corrosion resistance was helped by examination of a series of samples of established corrosion resistance. For resistance to 650°F water, concentration of either less resistant phase, i.e. free uranium or U_3Si_2, is disastrous. U_3Si_2 is tolerable if finely subdivided. As expected, the requirements for resistance to 500°F water are somewhat milder. U_3Si_2 still should not be coarse but free uranium can be tolerated to some extent, some of the corrosion products being retained by the metal.

A set of samples selected on the basis of resistance to water at 500°F and 650°F was tested in 750°F steam for 48 hours. Substantial parts of these samples were converted to sludge.

Coextrusion of Zircaloy-2-clad rods is being helped by use of a cold cone, i.e. one that is not preheated at all. Attention is being devoted to prevention of gouging of the Zircaloy and to a barrier between the Zircaloy and the steel can to eliminate the streaks of alloy resistant to pickling after extrusion.

Bare samples obtained by removal of the cladding from coextruded material are withstanding 500°F water but in 650°F water, cracks develop parallel to the axis. These cracks are attributed to uranium stringers generated by agglomerates of free uranium in the billets. The deterioration of the core in the course of extrusion hampers the assignment of any other cause for the failure of rods which were clad, end-plugged, and defected. After insertion of a 40-mil defect, the longest test period before failure is still 36 days. Samples with 7-mil defects have been tested as long as 81 days; none have failed yet.

The behavior of the epsilon alloy in liquid lead was investigated. A two-week test in lead at 350°C showed no attack. At 500°C or above, an as yet unidentified layer built up.

A report was received from Mr. J. H. Kittel of Argonne National Laboratory on the effects of irradiation on two bare, as-cast and epsilonized samples prepared at NMI some time ago. "The sample with 0.33% uranium atom burnup grew in length 2.1%, increased in diameter 1.8%, and developed some surface cracking. The specimen with 0.12% burnup shortened 1.4% and increased in diameter 2.9%. These results are similar to those that have been obtained on castings of the same alloy made at this laboratory (Argonne)." Coextruded rods were shipped

to Argonne for supplementary test. (S.Isserow, J.F.Kuchta, W.B.Tuffin)

Uranium-Rich Alloys

A number of alloys are being studied in an exploratory program to find compositions which might exhibit good irradiation stability to high burnup levels in a fast reactor. To date, twelve binary and ternary uranium-rich alloys containing 5 w/o additions of molybdenum, niobium, titanium, vanadium, and zirconium have been prepared by induction melting and casting in graphite. Fabrication behavior and microstructures in cast, extruded, and heat treated conditions are being examined as initial screening criteria.

Some characteristics as determined to date are listed in Table I. All alloys, with the exception of U-Mo-Ti which was so brittle in the cast condition as to be unmachinable, have been successfully extruded by sheathing in copper. Extrusion constants are compared at 1650°F for most alloys. Vanadium has the smallest effect in raising the extrusion constant of uranium (K for uranium = 3 tons per sq. in. at 1600°F), whereas molybdenum causes the most rapid increase in extrusion constant with alloying content.

Ability of as-extruded rods to withstand cold reduction in swaging has been studied, As indicated in Table I, several of the alloys withstood substantial reductions without annealing before failure occurred. Least success was experienced with alloys containing titanium. The uranium-titanium system is known to form a brittle intermetallic, U_2Ti. Also, in general, difficulty in cold working increases with hardness of the alloy in the extruded condition. Thus, the softer alloys, U-V, U-Mo, and U-Mo-Nb were most easily cold worked. The less readily cold worked alloys require further study. Effect of heat treatment prior to swaging and response to warm swaging should be examined for some of the alloys.

Photomicrographs are being obtained for the alloys in the conditions indicated in Table I. Probable gamma retention is observed in the quenched U-Mo-Zr alloy and the possibility of at least partial retention in the slow cooled condition is suggested by the microstructure. This alloy will be studied further. The U-V alloy shows a two phase dispersion (probably δ vanadium in a uranium matrix) and will also be examined in greater detail, particularly in view of the good fabrication characteristics. Additional attention will also be directed to ternary alloys containing vanadium. (L.R.Aronin)

TABLE I - CHARACTERISTICS OF URANIUM-RICH ALLOYS

Alloy Composition w/o	Extrusion Constant K At 1650°F Tons/sq.in.	Approx. Cold Reduction to Failure by Swaging %	Rockwell A Hardness			
			As Cast	As Extruded	Heated 800°C-15 min. H_2O Quench	Heated 800°C-15 min. Furnace Cool
U-5Mo	16.5	45	57	49	40	69
U-5Zr	*	25	67	70	76	66
U-5Mo-5Zr	22	20	76	67	67	78
U-5Nb	14.5	15	70	69	51	70
U-5Mo-5Nb	21.5	55	61	66	56	68
U-5Zr-5Nb	14	0	75	73	61	72
U-5Ti	14.5	0	67	76	74	69
U-5Mo-5Ti	* Brittle as cast					
U-5Zr-5Ti	14	0	71	75	75	71
U-5V	9	50	61	64	71	60
U-5V-5Mo	13	3	66	70	71	64
U-5V-5Zr	12	0	69	72	78	68

* 25 TSI at 1200°F

Extrusion constant $K = P/\ln \frac{A_o}{A}$ Where P = Running Pressure

A_o, A = Initial and Final Cross Sections

Zircaloy-2-clad Uranium-2 w/o Zirconium: Interdiffusion Treatment

Defected, interdiffusion treated specimen rods of U-2 w/o Zr in a 30 mil thick Zircaloy-2 clad have survived over 2600 hours in 600°F, 1500 psi water or over 1700 hours in 750°F, 1500 psi stream. The defects are 7 mil diameter holes which were drilled through the clad to, or a short distance into, the core at the mid-lengths of the specimens,

usually before the heat treatment. Prior to corrosion testing, the specimens received heat treatments varying from 850°C for 24 hours to 1100°C for one hour. Satisfactory coextruded rods of U-2 w/o Zr core in Zircaloy-2 clad have been obtained. New specimens are being prepared for use in proposed studies of the effects of larger defects, weak clad-to-core bonds, thinner clads, shorter heat treatments, and other factors on their corrosion resistance. Other studies being planned are concerned with the preparation, treatment, and testing of clad rods containing pre-extrusion defects; the effect of the interdiffusion treatment on clad rods with cores of either uranium or alloys such as U-Nb, which do not form brittle compounds with the clad; the effect of irradiation on interdiffusion treated specimen fuel rods. (R.G.Jenkins)

SYLVANIA ELECTRIC PRODUCTS, INC. - Reported by H. S. Kalish

Alloy Preparation

Techniques were improved for the hydrostatic pressing of uranium alloy rod about 0.2" in diameter and approximately 9" long. Rods of 1.2 w/o Mo-U powders were compacted hydrostatically at 30 tsi and vacuum sintered for 5 hours at 1100°C. The samples attained densities of 18.5 gms/cc, which is about 98.5% of theoretical. Straight rods are obtained by utilizing V-grooves in graphite slabs in which the compacts are placed. At the present time, the rods must be centerless ground, but attempts are being made to develop techniques which will permit uniform diameter rods to be obtained.

Mechanical Properties

Work was continued on obtaining the mechanical properties of various powder metallurgy binary uranium alloys. The data in Table I show a comparison between the properties of these alloys with as-cast uranium.

These data, some of which were presented in the previous Newsletter

(No. 12) verified the trend for decreased ductility and increased strength with increased molybdenum content.

The 2 w/o Zr-U alloy is the most ductile of all the alloys made to date.

TABLE I

MECHANICAL PROPERTIES OF SOME POWDER METALLURGY URANIUM ALLOYS IN THE AS-SINTERED CONDITION.

SPECIMENS COMPACTED AT 50 TSI IN A STANDARD POWDER METALLURGY TENSILE BAR DIE WHICH YIELDS A RECTANGULAR CROSS-SECTION TENSILE SPECIMEN.

Nominal Composition, w/o	Vickers Hardness, DPH	Yield Strength psi (0.2% Offset)	Ultimate Tensile Strength psi	Elongation Percent in 1 inch
As-Cast U	--	28,000	56,000	4
0.8 Mo	279	69,000	122,000	6
1.0 Mo	275	63,000	125,000	6
1.4 Mo	296	100,000	136,500	5.5
2.0 Mo	306	87,000	135,000	4
3.0 Mo	335	101,000	145,000	1.5
3.5 Mo	367	--	147,000	--
4.0 Mo	392	136,000	160,000	1
2.0 Zr	278	81,000	122,000	9
0.5 V	260	58,000	111,000	4.5
1.0 V	277	55,000	110,000	4.5
0.75 Si	250	46,000	80,000	3
3.0 Nb	338	100,000	145,000	1.5

Above data is average of 8 samples for each composition.

Thermal Cycling

A series of powder metallurgy Mo-U alloys were thermal cycled in comparison with powder metallurgy Zr-U alloy and wrought ahd heat-treated Zr-U alloy at Argonne National Laboratories. Two pin type samples of each composition were alpha cycled rapidly 200 times between 65° and 620°C. In addition, two samples of each composition were beta cycled rapidly 200 times between 90° and 715°C. The results of this cycling are shown in Table II.

TABLE II

THE EFFECT OF 200 FAST CYCLES ON THE DIMENSIONS OF PIN TYPE (0.145" DIAMETER x 2.3" LONG) URANIUM ALLOY SAMPLES. DATA SHOWN IS THE AVERAGE OF TWO SAMPLES

Nominal Composition, w/o	Fabrication Method	Elongation, %	Weight Change, %	Density Decrease %
	A. ALPHA CYCLES, 65° to 620°C			
2.0 Zr	Cast, wrought and heat treated	*	*	0.83
2.0 Zr	Sintered-Fine Grained	1.02	- 0.49	1.78
2.0 Zr	Sintered-Coarse "	1.24	- 0.26	1.59
1.4 Mo	Sintered	0.9	- 0.32	1.55
2.0 Mo	Sintered	0.98	+ 0.31	1.07
2.5 Mo	Sintered	0.61	+ 0.10	1.12
	B. BETA CYCLES, 90° to 715°C			
2.0 Zr	Cast, wrought and heat treated	**	*	0.7
2.0 Zr	Sintered-Fine Grained	**	- 3.96	21.0
2.0 Zr	Sintered-Coarse "	**	- 3.40	6.3
1.4 Mo	Sintered	1.70	- 4.20	1.0
2.0 Mo	Sintered	1.0	- 2.00	1.3
2.5 Mo	Sintered	0.8	+ 0.03	2.5

* Not determined
** Could not be measured accurately for length change because of distortion.

The above data indicate that the 2 w/o Zr-U made by both methods and the 1.4 w/o Mo-U and 2 w/o Mo-U alloys are all essentially the same when alpha cycled. The 2.5 w/o Mo-U alloy, however, was more stable than any of the others. In beta cycling, both types of 2 w/o Zr-U alloy are far less stable than any of the molybdenum alloys. At the same time, there is definite trend of improved dimensional stability in going from 1.4 to 2.5 w/o Mo-U. It is interesting to note that the 2 w/o Zr-U alloy made by powder metallurgy methods showed a difference in dimensional stability depending upon initial grain size. As one would expect, the finer grain sized material was the more stable of the two.

The density changes which occur seem to be primarily a function of NaK corrosion. This is indicated by the weight change. One exception to this appears to be the 2 w/o Zr-U powder metallurgy alloy which was beta cycled. The large density decrease in these samples appears to be due to the formation of internal porosity during thermal cycling.

The surface appearance of the samples after beta cycling indicated the superiority of the 2.5 w/o Mo-U alloy even more clearly. It had essentially a smooth surface, whereas the 2 and 1.4 w/o Mo-U alloys showed increasing amounts of surface roughness. The 2 w/o Zr-U alloys were extremely rough and distorted after beta cycling.

Alloys for Improved Corrosion Resistance

The investigation of the feasibility of making a completely epsilonized 3.8 w/o Si-U alloy by powder metallurgy was continued. Elemental powder mixtures of 3.7, 3.8, and 3.9 w/o Si-U were hot pressed and subsequently heat treated for 140 hours at 850°C to homogenize and epsilonize the structure.

These samples were corrosion tested in 600°F water with cast epsilonized 3.8 w/o Si-U alloy as a control. The data in Table III show that the 3.8 w/o Si-U alloy is the best of the powder metallurgy samples. It has, however, inferior corrosion resistance to the cast material. This is undoubtedly a function of the low density of the powder metallurgy alloys as indicated in Table IV.

TABLE III

CORROSION RATES OF SOME SILICON-URANIUM ALLOYS IN 600°F WATER			
	Corrosion Rate in $mg/cm^2/hr$		
Sample	24 Hours	48 Hours	72 Hours
3.8 w/o Si Cast and Epsilonized	- 5.5	- 3.2	- 2.5
3.7 w/o Si Hot Pressed and Homogenized	- 25.5	- 23.0	Sample Disintegrated
3.8 w/o Si Hot Pressed and Homogenized	- 6.4	- 3.9	- 3.9
3.9 w/o Si Hot Pressed	- 5.5	- 4.6	- 5.2

TABLE IV

THE DENSITY OF SOME SILICONE-URANIUM ALLOYS		
Sample	Density, As Hot Pressed gms/cc	Density, After Homogenization (Epsilonized) gms/cc
3.8 w/o Si Cast and Epsilonized	--	15.45
3.7 w/o Si Hot Pressed	14.78	14.19
3.8 w/o Si Hot Pressed	14.66	13.87
3.9 w/o Si Hot Pressed	14.53	13.67

The data of Table IV show that the density decreases during homogenizing and epsilonizing, whereas it should increase. This decrease in density is due to the Kirkendall effect, that is, during homogenizing, the diffusion rates of the silicon into the uranium and uranium into silicon are different, such that porosity is developed in the material. The only possible way, then, that the powder metallurgy alloys can be made of high density is by subsequent hot working of the homogenized material.

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WESTINGHOUSE ATOMIC POWER DIVISION - Reported by M. L. Bleiberg

Irradiation Behavior of Uranium-Molybdenum and Uranium-Niobium Alloys

Samples of uranium-molybdenum (9, 10.5, 12, and 13.5% Mo) and uranium-niobium (10% Nb) alloys exposed to pile irradiation in both the gamma-quenched and alpha-transformed conditions reverted to a condition intermediate between the untransformed and transformed states; that is, the gamma-quenched samples showed a slight tendency to transform, while the transformed samples showed a marked decrease in the amount of transformation. All the irradiations were conducted at temperatures far below those at which thermally-induced transformations would be expected. This behavior was also reported in case of the U-9 Mo alloy by Konobeevsky, Pravdyuk, and Kutaitsev (p. 681, Effect of Irradiation on Structure and Properties of Fissionable Materials) at the Geneva Conference. These effects appear to be quite well-established based upon electrical resistivity, temperature coefficient of electrical resistivity and hardness, (the Russian investigators employed, in addition, X-ray diffraction techniques).

U-Mo Alloys

Dimension and Density Changes. The changes in dimensions and density of the U-Mo alloys due to neutron bombardment are shown in Table I. The U-9 w/o Mo alloy specimens which were heat treated to produce the $\delta + \epsilon$ phase showed decreases in density of 1.0 and 1.3 percent after exposures of 920 and 700 MWD/T, respectively. One U-10.5 w/o Mo alloy specimen showed a density decrease of 0.4% after an exposure of 500 MWD/T. No density or diametrical changes were found in the U-9 w/o Mo or U-10.5 w/o Mo alloy specimens irradiated in the gamma phase. The U-12 w/o Mo and U-13.5 w/o Mo alloy specimens showed no changes in diameter or density regardless of their prior heat-treated condition or neutron flux

exposure. Note that the density decreases in the U-9 w/o Mo alloy specimens are such that the transformed specimens after irradiation have density values close to that of gamma-quenched U-9 w/o Mo alloys. The difference in density values of U-10.5, -12, and -13.5 w/o Mo, between the gamma-quenched condition and the transformed condition, is small or negligible and if phase transformation reversal occurred in these alloys no density changes would be noted.

Electrical Resistivity Results. The changes in electrical resistivity in liquid nitrogen of these specimens after irradiation are shown in Table II. The gamma-quenched U-9 w/o Mo alloy specimen showed a decrease in electrical resistivity at -196°C of 1.2% while the transformed specimens of U-9 w/o Mo alloy showed increases of 125% and 114% respectively. The same behavior was found in the U-10.5 w/o Mo alloy specimens with the gamma-quenched specimen decreasing 3% and the transformed specimens increasing 14.6% and 21.7% respectively. In the U-12 and 13.5 w/o Mo alloy specimens, those specimens which were initially in the gamma-quenched condition and those which were slightly transformed by aging at 525°C for 8 hours and 48 hours respectively, showed decreases in electrical resistivity. However, those specimens which were more completely transformed prior to irradiation by aging at 525°C for 312 hours show appreciable increases in electrical resistivity. It is interesting to note that all specimens of the U-12 w/o Mo alloy appear to approach an equilibrium value of electrical resistivity of approximately 74 μΩcm regardless of prior heat treatment, exposure and temperature in-pile. Similarly, the U-13.5 w/o Mo alloy specimens appear to approach equilibrium at about 76 μΩcm, the U-10.5 w/o Mo at 69-73, and the U-9 w/o Mo at 68-71 μΩcm.

The gamma-quenched samples of U-9, -10.5, -12, and -13.5 w/o Mo exhibit a negative temperature coefficient of electrical resistivity

between -196°C to +150°C and the transformed specimens of the same composition exhibit a positive temperature coefficient of electrical resistivity in the same temperature range. Electrical resistivity measurements on the irradiated U-9, -10.5, -12 and -13.5 w/o Mo alloy specimens at -196°C, -75°C, +50°C and +100°C have been obtained and these results show that these alloy specimens irradiated in the gamma phase exhibit a negative temperature coefficient after irradiation similar to that prior to irradiation. However, the positive temperature coefficient of electrical resistivity of the transformed specimens has reversed to a negative coefficient after irradiation.

Hardness Changes. The changes in hardness of these specimens are shown in Table III. It may be seen that the hardness of the transformed U-9, and -10.5 w/o Mo alloy specimens has reverted to that of the gamma phase after irradiation. The differences in hardness of U-12 and -13.5 w/o Mo alloys between the gamma-quenched condition and the transformed condition is small or negligible so that if phase transformation reversal occurred in these alloys no hardness changes would be noted.

U-Nb Alloys

The changes in dimensions and density for the gamma-quenched U-10 w/o Nb specimens shown in Table IV are of the same order of magnitude as the changes found in U-Mo alloys given a similar exposure, while the changes shown for the aged specimens are generally greater than changes observed in transformed U-Mo alloys. Note that the direction of the changes are toward the gamma condition in the aged alloys since on transformation of gamma, density increases. This same effect may be noted for the electrical resistivity data, shown in Table IV. It is to be noted that all the transformed specimens have changed toward the gamma-quenched condition.

Discussion of Above Results

The results above show that the stable $\alpha + \epsilon$ phase of U-9, -10.5, -12 and -13.5 w/o Mo alloys reverts back to the metastable α phase after being subjected to neutron bombardment. The electrical resistivity behavior of these alloys is similar to that shown in order-disorder alloys after neutron bombardment in that the disordered phase (γ) slightly orders while the ordered phase (ϵ) disorders, both conditions approaching the same degree of disorder. It may be surmised that neutron bombardment of the ordered epsilon phase reverts it to the disordered gamma. Subsequently (or perhaps simultaneously) the free alpha uranium present is taken back into solution.

In the case of the U-Mo alloys where one of the products of transformation is the ordered epsilon phase, the results obtained are in harmony with experimental observations on other order-disorder systems, although it is indeed puzzling that alpha precipitated during a 2-week anneal at 525°C is apparently re-absorbed during the 6-week irradiation at a maximum estimated temperature of 300°C. However, in case of the U-Nb system, where no such ordered phase has been reported, this explanation is not available.

Cold Water Irradiation of Prototype Rod Fuel Elements

Prototype fuel rods of uranium and U-2 w/o Zr alloy with enrichments which varied from 0.7 to 5.5 w/o U-235 have been irradiated in the Chalk River NRX reactor to a maximum burnup of 10,500 MWD/T and have been returned to the Bettis Hot Laboratory. At regular intervals during the irradiation, the rods were removed from the reactor and X-rayed so that changes in length of the rods were obtained. These results indicated that the U-2 w/o Zr alloy rods exhibited greater growth (approximately 1 to 3%) than the unalloyed uranium rods (approximately 0.1 to 0.5%).

However, when dimensional measurements were made on the irradiated rods in the Hot Laboratory, it was found that the percent change in volume of the rods ranged from 1 to 3% and appeared to be independent of core composition, and prior heat treatment of the rods. However, there appears to be a trend toward increasing volume change with increasing exposure, amounting to about 3% at 10,000 MWD/T.

Visual examination of these rods showed that the three rods irradiated to 9000 MWD/T or greater exposure exhibited transverse and longitudinal splitting of the cladding in the vicinity of the end cap weld. In addition, smaller longitudinal splits and bulges were observed in the middle of these specimens. No cladding failures have been observed in specimens irradiated to lower exposures, indicating that 9000 MWD/T may be the upper limit of exposure before the cladding is excessively stressed by expansion of the core alloy.

Identification of Hydride Phases Formed During Corrosion of Gamma Phase Alloys

An experiment is in progress to identify the hydride phase formed during hot water corrosion of U-Mo alloys. The experiment consists of measurement of the decomposition (hydrogen) pressure as a function of temperature over samples of 12 w/o Mo alloy which were gamma-quenched and then hydrided by autoclave corrosion treatment in 650°F water.

Preliminary measurements on specimens which were corroded for 7 and 21 days, resulting in a hydrogen concentration of 270 and 1350 ppm, respectively, have been completed and indicate reproducible equilibrium pressures. The values closely approximate the relation:

$$\log P_{mm} = \frac{-1235}{T} + 5.350$$

as compared with:

$$\log P_{mm} = \frac{-4480}{T} + 9.20$$

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for the equilibrium between UH_3, hydrogen, alpha-uranium. Plots of log P versus $\frac{1}{T^{\circ}K}$ were made and extrapolation of this plot revealed a pressure of about 38 mm at room temperature for the U-Mo specimens. The value for alpha U is 0.6 mm at 200°C. Decreasing the temperature revealed that some of the gas is reabsorbed, at preceptible rates even at room temperature, indicating reversibility of the reaction on a qualitative basis.

It appears from the above that much higher hydrogen pressures and lower heats of formation are associated with the decomposition of the corrosion hydride compared with pure UH_3. The corrosion hydride, therefore, is different than pure UH_3.

Table I

Changes in Dimensions and Density of Irradiated U-Mo Alloys

Sample No.	Composition	Heat Treatment	Estimated Exposure (MWD/T)	(Diameter in.) Pre	Post	% Change	Density (g/cm^3) Pre	Post	% Change
A-6	U-9 w/o Mo	#1	310	.1880	.1887	n.s.	17.35±.01	17.34	0
A-8	U-9 w/o Mo	#4	920	.1875	.1887	+0.6)	17.47±.01	17.29	-1.0
A-9	U-9 w/o Mo	#4	700	.1870	.1882	+0.7)		17.24	-1.3
B-1	U-10.5 w/o Mo	#1	600	.1880	.1888	n.s.	17.15±.01	17.16	0
B-12	U-10.5 w/o Mo	#4	540	.1880	.1880	0	17.18±.01	17.16	-
B-9	U-10.5 w/o Mo	#4	500	.1870	.1874	n.s.		17.11	-0.4

Notes:

1. All specimens of U-12 w/o Mo and U-13.5 w/o Mo having heat treatments #1 to #4 respectively showed no change in dimensions or density.

2. n.s. indicates an insignificant change.

3. Heat Treatments

   #1 - 900°C in kinetic vacuum for 24 hours, water-quenched.
   #2 - #1 plus 525°C in evacuated Vycor tubes for 8 hours, water-quenched.
   #3 - #1 plus 525°C in evacuated Vycor tubes for 48 hours, water-quenched.
   #4 - #1 plus 525°C in evacuated Vycor tubes for 312 hours, water-quenched.

Table II

Changes in Electrical Resistivity of Irradiated U-Mo Alloys

Sample No.	Composition	Heat Treatment	Exposure (MWD/T)	ρ at -196°C Pre	ρ at -196°C Post	% Change
A-6	U-9 w/o Mo	#1	310	72.20	71.36	-1.2
A-8	U-9 w/o Mo	#4	920	31.17	70.81	+125
A-9	U-9 w/o Mo	#4	700	31.15	67.52	+114
B-1	U-10.5 w/o Mo	#1	600	74.48	72.22	-3.0
B-12	U-10.5 w/o Mo	#4	540	59.75	68.52	+14.6
B-9	U-10.5 w/o Mo	#4	500	59.74	72.68	+21.7
C-6	U-12 w/o Mo	#1	310	76.41	74.15	-3.0
C-8	U-12 w/o Mo	#2	770	76.23	74.71	-2.0
C-9	U-12 w/o Mo	#2	600	75.99	74.09	-2.5
C-15	U-12 w/o Mo	#3	475	75.62	74.46	-1.5
C-16	U-12 w/o Mo	#3	640	75.70	74.85	-1.1
C-22	U-12 w/o Mo	#4	780	70.15	73.03	+4.1
C-23	U-12 w/o Mo	#4	610	69.67	72.98	+4.8
D-2	U-13.5 w/o Mo	#1	310	79.57	76.32	-4.1
D-14	U-13.5 w/o Mo	#2	700	78.20	76.56	-2.1
D-9	U-13.5 w/o Mo	#2	500	78.77	76.33	-3.1
D-15	U-13.5 w/o Mo	#3	475	76.78	75.81	-1.2
D-16	U-13.5 w/o Mo	#3	820	76.48	76.26	-0.3
D-22	U-13.5 w/o Mo	#4	810	72.09	76.40	+6.0
D-24	U-13.5 w/o Mo	#4	720	72.96	75.77	+3.9

Note:

Heat Treatments

#1 - 900°C in kinetic vacuum for 24 hours, water-quenched.
#2 - #1 plus 525°C in evacuated Vycor tubes for 8 hours, water-quenched.
#3 - #1 plus 525°C in evacuated Vycor tubes for 48 hours, water-quenched.
#4 - #1 plus 525°C in evacuated Vycor tubes for 312 hours, water-quenched.

Table III

Hardness of Irradiated U-Mo Alloys

Spec. No.	Comp. w/o Mo	Pre-Irrad. Heat Treatment	Exposure* (MWD/T)	Hardness (5 kilo load) Pre	Post	DPH
A-1	U-9 w/o Mo	#1	310	287±4	299±11	n.s.**
A-5	U-9 w/o Mo	#4	920	440±20	303±6	-197±26
A-7	U-9 w/o Mo	#4	700	444±22	299±9	-145±31
B-1	U-10.5 w/o Mo	#1	600	305±1	300±10	n.s.
B-6	U-10.5 w/o Mo	#4	540	384±17	334±9	-50±26
B-7	U-10.5 w/o Mo	#4	500	361±7	326±14	-35±21
C-1	U-12 w/o Mo	#1	310	324±5	307±12	n.s.
C-6	U-12 w/o Mo	#2	770	320±9	323±5	n.s.
C-7	U-12 w/o Mo	#2	600	321±4	--	--
C-11	U-12 w/o Mo	#3	475	326±6	350±8	24±14
C-12	U-12 w/o Mo	#3	640	333±3	323±14	n.s.
C-16	U-12 w/o Mo	#4	780	344±6	353±16	n.s.
C-17	U-12 w/o Mo	#4	610	336±8	353±15	n.s.
D-1	U-13.5 w/o Mo	#1	310	341±4	348±4	n.s.
D-7	U-13.5 w/o Mo	#2	700	311±2	339±10	-28±12
D-8	U-13.5 w/o Mo	#2	500	339±16	345±6	n.s.
D-11	U-13.5 w/o Mo	#3	475	333±5	327±14	n.s.
D-12	U-13.5 w/o Mo	#3	820	325±6	334±16	n.s.
D-16	U-13.5 w/o Mo	#4	810	348±7	--	--
D-11	U-13.5 w/o Mo	#4	720	343±4	347±10	n.s.

* From flux monitor data
** n.s. - not significant

Table IV

Physical Property Changes of Irradiated Unclad U-10 w/o Nb Alloys

Specimen No.	F-3	F-4	F-5	F-22	F-19	G-4	G-1
Pre-Irradiation Condition	900°C-24 Hrs-W.Q.	900°C-24 Hrs-W.Q.	900°C-24 Hrs-W.Q.	900°C-24 Hrs+400°C-2 Wks	900°C-24 Hrs+450°C-2 Wks	900°C-24 Hrs+500°C-2 Wks	900°C-24 Hrs+550°C-2 Wks
Exposure (MWD/T)	1820	1660	1220	222	446	1060	1660
Pre-Length (in.)	1.992	1.993	1.995	1.983	1.983	1.984	1.984
Post-Length (in.)	1.995	1.995	1.996	1.995	1.993	2.014	2.052
% Change in Length	+0.2	+0.1	+0.1	+0.6	+0.5	+1.5	+3.4
Pre-Density (gm/cm^3)	16.51	16.49	16.49	16.71	16.76	16.79	16.82
Post-Density (gm/cm^3)	16.49	--	16.50	16.55	16.58	16.53	16.47
% Change in Density	-0.1	--	-0.1	-1.0	-1.1	-1.6	-2.1
Pre-Elect. Resist. at 196°C	71.19	71.22	71.19	35.71	21.73	18.04	16.57
Post-Elect. Resist. at 196°C	61.55	69.96	72.64	58.99	41.58	34.20	27.76
% Change	-13.5	-1.8	+2.0	+65.2	+91.4	+89.6	+67.5
Pre-Elect. Resist. at R.T.	65.59	65.76	65.66	52.22	44.24	41.56	40.14
Post-Elect. Resist. at R.T.	63.87	66.12	68.54	63.67	58.26	53.44	50.27
% Change	-2.6	+0.6	+4.4	+21.9	+31.7	+28.6	+25.2