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UNITED STATES ATOMIC ENERGY COMMISSION

URANIUM ALLOY NEWSLETTER

Edited by
Edward Epremian

September 1956
[TISE Issuance Date]

Division of Research
Washington, D. C.

Technical Information Service Extension, Oak Ridge, Tenn.

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AMES LABORATORY - Reported by H. A. Wilhelm

Uranium-Niobium Phase Diagrams

The phase diagram of the U-Nb alloys already is established in outline. However, an effort is being made to place some of the boundary lines as exactly as possible.

One line that is under re-investigation is the solidus. Melting point measurements are being made on niobium-rich alloys that have been homogenized more thoroughly than the specimens used previously. This work should determine whether or not the sudden lift in the line near 70 per cent niobium actually exists.

Solvus lines on both sides of the diagram are being re-studied by x-ray and metallographic methods. This work indicates that the solubility of niobium in alpha uranium may be a little lower than previously believed. Determination of the solvus line bounding the niobium-rich solid solution area is difficult because of the sluggishness of the changes in the alloys. Apparently, this boundary line lies slightly above 50 per cent niobium; that is, at a little higher niobium content than it was previously placed.

Preparation of Ingots of Uranium-Niobium Alloy

A study was made of the preparation of a U-10 w/o Nb alloy by the bomb reduction of a suitable form of niobium and uranium tetrafluoride with calcium. A series of reductions were attempted in which Nb_2O_5 , Nb metal powder, NbCl_5 and salt mixtures having the formulae of K_2NbOF_5 and Na_2NbOF_5 were used in the reaction charge. Na_2NbOF_5 proved to be most satisfactory as the source of niobium. High yields and good metal-slag separations were obtained. Chemical analysis and density determinations showed the composition to 10 ± 0.5 w/o niobium. Fifteen pounds of alloy were melted in BeO and cast into a graphite mold to give an ingot of fairly uniform composition. No data have been obtained on the amounts of such

impurities as carbon, oxygen and hydrogen, nor has the method been applied to anything larger than a three-pound reduction scale. Further work is needed on the study of these and other factors before a final evaluation of the method can be made. Such studies are currently in progress.

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ARGONNE NATIONAL LABORATORY - Reported by L. T. Lloyd

Uranium-Silicon Alloys

The effect of irradiation on the corrosion resistance of cast U-3.8 w/o Si alloy is difficult to evaluate because of non-reproducible results on unirradiated material. Samples with burn-ups up to 0.71 total a/o have had satisfactory corrosion resistance at 290°C for periods up to four days. Failure does not appear to be due to simple corrosion as the irradiated material is very brittle and cracks even when not subjected to corrosion conditions. (Greenberg)

Uranium-Titanium Alloys

A program is underway to study the mechanisms by which the aqueous corrosion of uranium is improved by alloying. To date, a uranium-2.7 w/o titanium alloy quenched from the gamma phase has shown promising corrosion resistant properties. After 11 days at 290°C the corrosion rate is about 30 mg/cm²/day. (Greenberg)

Uranium-Zirconium Alloys

Phase Diagram: A series of arc melted and cast U-Zr alloys (4.7 to 32.0 w/o Zr) were homogenized at 1000°C to 1150°C, oil or water quenched, then annealed for 1 week at 750°C, 724°C, 700°C, and 690°C and water quenched. From micro-examination of the alloys the following observations can be made:

1. The existence of a mono-eutectoid transformation and a $\gamma_1 + \gamma_2$ region as reported by other investigators is confirmed.
2. The mono-eutectoid transformation temperature is between 690°C and 701°C.
3. At 701°C the low zirconium boundary of the $\gamma_1 + \gamma_2$ region is above 6.9 w/o Zr. The high zirconium boundary appears to be between 21.3 and 22.97 w/o Zr.
4. At 690°C the beta uranium + γ_2 region extends to between 24.96 and 26.98 w/o Zr. (Zegler)

Irradiation: Efforts to stabilize wrought U-2 w/o Zr alloys by proper heat treatment have been continued. The good results obtained by soaking for a week at 690°C after an initial 800°C gamma quench have been mentioned (WASH-198). Study of this heat treatment has been continued by irradiation of a series of

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specimens quenched and tempered at 690°C for various lengths of time. As shown in Table I, the growth rate of the specimens is dependent on transformation time, with minimum growth rate shown by the specimens held at temperature 6 hours. Measurements were made at a burnup level of about one percent of total atoms. (Kittel and Paine)

TABLE I

Effect of Transformation Time on the Irradiation Growth Rate
of U-2 w/o Zr Alloy Specimens
(Previously Quenched from 800°C)

Hours Tempered at 690°C	Growth Rate, G_1^*
0.17	21.2
1	8.4
6	5.1
24	20.7
128	38.9

*Microinches per inch per part per million burnup.

Uranium-Plutonium-Fissium Alloys

Dilatometric studies of several uranium-20 w/o plutonium-fissium alloys resulted in the expansion coefficients in Table II. The 10.8 w/o fissium alloy is the equilibrium alloy and contains 4.3 w/o Ru, 2.8 w/o Mo, 2.5 w/o Pd, 0.7 Rh, and 0.5 w/o Zr. The other fissium alloys contain the same ratio of fissium product elements. (Baumrucker)

TABLE II

Expansion Coefficients of U-Pu-Fissium Alloys

Nominal Fissium Content (bal. U-20 w/o Pu)	Temperature Range °C	Expansion Coefficient $\alpha \times 10^6/^{\circ}\text{C}$ (average)	Number of Runs
10.8 w/o	20-500	15.2	4
8.1 w/o	25-367	17.11	5
5.4 w/o	25-367	16.99	9
5.4 w/o	25-438	17.53	9

Uranium-Zirconium-Niobium Alloys

Phase Diagram: A phase has been observed in the U-Nb binary, in the range of 6 to 8 w/o Nb but not in the 10 1/2 w/o alloy, whose X-ray diffraction pattern can be indexed seemingly on the basis of a tetragonal unit cell. This tetragonal

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cell can be derived from the body centered cubic high temperature γ phase by slightly distorting this latter phase. The degree of tetragonality is apparently a function of the Nb content and may reach a maximum at approximately 7 w/o Nb. This phase has been found in these binary alloys in the as cast, as cast and rolled, and also after an interrupted quench. There is evidence that if these alloys are annealed at high temperature and quenched rapidly, it is possible to retain the high temperature body centered cubic γ phase without transforming to the tetragonal phase.

This tetragonal phase has also been observed to date in the following ternary alloys: U-6 w/o Nb-2 w/o Zr, U-6 w/o Nb-4 w/o Zr, U-4 w/o Nb-6 w/o Zr, and U-7 w/o Nb-10 w/o Zr. (Dwight and Mueller)

Corrosion: As measured by corrosion resistance at 260°C and higher, it appears that the critical radiation exposure of the gamma quenched U-5 w/o Zr-15 w/o Nb alloy is that producing between 0.046 and 0.074 total a/o burn-up. Samples which had been irradiated up to the lower dosage had corrosion resistance comparable to that of the unirradiated material at temperatures up to 315°C. However, a sample, (wt. of about 45g) irradiated to a burn-up of 0.074 total a/o was about half corroded after 1.71 days at 260°C. (Greenberg)

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

Continuous Cooling Transformation Studies For The U-10 w/o Nb Alloy

Continuous cooling transformation kinetics have been investigated for the U-10 w/o Nb alloy. Samples encapsulated in Vycor tubes were solution annealed in the gamma field at 950°C and cooled at linear rates. The specimens were water quenched from temperatures between 300° and 550°C.

Metallographic investigation showed that the decomposition ($\gamma \rightarrow \alpha + \gamma_2$) structure was similar to that obtained by isothermal annealing. The continuous cooling transformation diagram is presented in Figure 1. The indicated times represent the cooling times from 950°C to the quenching temperature. The data points indicate annealing treatments required to produce incipient transformation. Many heat treatments were performed and the microstructures contained up to 80% transformation.

The previously determined transformation curve based upon isothermal data has been shown for comparison. For the isothermal studies the specimens were solution annealed in the gamma field at 1050°C. Based upon the above findings, the critical cooling rate to prevent decomposition is 8.5°C/minute.

Vickers hardness data (10 Kg) were obtained on the metallographic samples. Plots of hardness vs log time were constructed. The times required to produce a hardness increase are shown in Figure 2. The findings are similar to the metallographic results, although with isothermal annealing, pre-precipitation hardening occurs at the lower temperatures. The critical cooling rate determined from the hardness data is 8°C/minute.

Similar continuous cooling transformation diagrams were constructed for samples continuously cooled after a solution anneal at 700°C for both 1 and 4 hours. This treatment represents annealing in the $\gamma + \gamma_2$ field. The hardness data were somewhat complicated by annealing in the two phase field.

As in the case of the 950°C solution treatment, the nose of the transformation curves occurs at about 500°C. The critical cooling rates for the solution treat-

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ments at 700°C for 1 and 4 hours are 8° and 10°C/minute, respectively. These results indicate that the longer holding time at 700°C may accelerate the monotectoid decomposition.

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FIG. 1 - ISOTHERMAL AND CONTINUOUS COOLING TRANSFORMATION DIAGRAMS FOR U-10% Nb ALLOYS BASED ON METALLOGRAPHIC RESULTS (SOLUTION TREATED FOR 1 HOUR AT 950°C)

FIG. 2 - ISOTHERMAL AND CONTINUOUS COOLING TRANSFORMATION DIAGRAMS FOR U-10% Nb ALLOYS BASED ON HARDNESS DATA (SOLUTION TREATED FOR 1 HOUR AT 950°C)

ATOMICS INTERNATIONAL - Reported by B. R. Hayward

Our main effort has been procuring experimental fuel slugs for our SRE. To date we have procured 500 slightly enriched U-Mo (1.2 w/o) alloy slugs 3/4 inch diameter by 1 inch long which were cold pressed, sintered and machined at Sylvania. A centrifugal casting program at Fernald is producing enriched 3/4 inch by 6 inch slugs of U-2 w/o Zr, U-1.2 w/o Mo and U-1.8 w/o Mo. Severe subsurface porosity was observed on the first fabrication cycle of enriched material, but this has been minimized in subsequent development work by a higher rpm, longer heat cycle and cleaner feed material. The enriched alloy slugs are now being recast using the improved techniques. The machinability of cast U-1.2 w/o Mo has been greatly improved by annealing for two hours at 1100°F.

Development work has been completed and extrusion of enriched Th-U (5.4%U) rods has commenced at Nuclear Metals. Analysis of uranium in the thorium still shows minor variations between sites, more direct comparative analyses are planned. The exact location of the uranium in the thorium is unknown.

Capsule tests on experimental fuel alloys are ready for irradiation at the MTR.

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BATTELLE MEMORIAL INSTITUTE - Reported by F. A. Rough

PHYSICAL PROPERTIES AND TRANSFORMATION STUDIES

Investigation of the Tensile Transition in Alpha Uranium

The tensile transition in alpha uranium is being studied in an effort to learn more about the nature of the process resulting in the loss of tensile ductility. The variation of microstructure with the temperature of deformation has provided evidence that the transition phenomenon is not simply related to a change in the primary mode of deformation.

Replicas obtained from the surface of deformed specimens indicate that the deformation process at 200°F, nominally above the transition, is exceedingly complicated. The microstructure exhibits many twinning systems and a complete array of slip structure that is most difficult to analyze. At room temperature (75°F), the microstructure of specimens which are strained the same amount as at 200°F is comparatively much simpler. Both twinning and slip processes are active at 75°F. However, the slip structure at 75°F generally consists of single lines, whereas at 200°F there is a tendency for the slip structure to band together in a manner common to most metals. Another item observed is that the deformation markings with respect to twinning versus slip are very inhomogeneous from grain to grain, suggesting a possible orientation dependence for the various deformation modes.

At -75°F, the microstructure (light microscopy) revealed twinning and final fracture by cleavage. A secondary cleavage crack that did not result in specimen fracture followed the twin edges, jumping from one to another as each twin stopped. The point of departure was generally associated with a region of intersecting twins. This behavior at -75°F along with the behavior at 75°F and 200°F is remarkably similar to the behavior of iron on passing through its tensile transition.

Transformation Kinetics and Hardening Reaction of Zirconium Alloys

Containing up to 20 w/o Uranium

The time-temperature-transformation relationships are being determined in an effort to evaluate heat treatments and their effect upon mechanical properties in Zr-U alloys containing up to 20 w/o uranium.

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Isothermal Transformation. Heat treatments are essentially completed. Temperatures investigated include: 350, 400, 450, 500, 550, 580, 650, 700, and 750°C.

X-ray diffraction analysis has been performed on representative samples in order to identify the phases present. A phase previously designated as "omega" has been detected in samples quenched after transformation both above and below the eutectoid temperature. In order to determine whether this phase forms during continuous-cooling or during isothermal treatment, X-ray patterns were taken on a 20 w/o alloy transformed at 550°C for 20 min and 3900 min employing manganese radiation instead of the customary iron radiation. This choice of a target resulted in a separation of lines in the back-reflection area. It was then observed that the "omega" lattice appeared to be an expanded epsilon and approached epsilon by contracting upon isothermal holding. This would seemingly indicate that omega is merely a supersaturated epsilon lattice, and that the phase identified as omega must form upon quenching from temperatures above the eutectoid rather than forming upon isothermal treatment at temperatures greater than the eutectoid.

Microstructures of alloys containing the omega phase showed an equiaxed substructure within the prior beta grains. Upon longer holding, the equiaxed structure gradually changed to a Widmanstatten pattern. Electron micrographs of the substructure revealed a fine precipitate.

The shifting of diffraction lines of the omega phase on isothermal transformation will be followed further by use of the high-temperature camera on both a Zr-20 w/o and a 10 w/o U alloy.

Transformation Kinetics of Zirconium Alloys Containing 20 to 70 w/o Uranium

The transformation kinetics of the beta zirconium-to-epsilon decomposition is being studied in alloys containing 20 to 70 w/o uranium.

An X-ray diffraction study of specimens of the 22, 50, and 70 w/o uranium alloys, isothermally transformed at 575°C, and of the 50 w/o uranium alloy isothermally transformed at 500°C, has been made 628 014 .

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The 50 and 70 w/o uranium specimens showed only beta when quenched, while the 22 w/o specimens treated similarly showed a tract of epsilon. The lattice constants of the beta phase for the 22, 50, and 70 w/o uranium samples were 3.580, 3.558, and 3.553 Å, respectively. After 10 sec at 575°C, the 22 and 70 w/o uranium alloys showed apparent changes in lattice constant which were interpreted as a transformation to epsilon even though the intensities of superlattice reflections became well developed after 100 sec at 575°C in the 22 and 50 w/o samples, and after 10 sec in the 70 w/o sample.

Interplanar spacings of superlattice reflections were found to be dependent on uranium content, with the reflections in all the alloys showing a tendency toward assuming the same spacings with prolonged annealing. Accompanying the change in interplanar spacings in the 22 and 70 w/o uranium specimens is the precipitation of alpha zirconium and alpha uranium respectively. A tract of alpha zirconium is detected in the 22 w/o alloy specimen after 100 sec at 575°C and a trace of alpha uranium is detected in the 70 w/o uranium specimen after the same period of transformation. However, after as little as 0 and 10 sec, a broad shoulder in the diffraction curve near the strongest reflection indicates that some sort of preprecipitation phenomenon is taking place although its nature is not known.

These results are interpreted as indicating that the epsilon phase is a solid solution with a broad range of composition under nonequilibrium conditions. These composition limits narrow as either alpha zirconium or alpha uranium precipitate and the supersaturated epsilon phase begins to approach its equilibrium solubility limits.

Properties of Fissium Alloys

A program for the determination of the physical properties of fissium alloys is in progress. Those properties which have been determined and previously reported include the melting point, the hot hardness, and the fluidity of the alloy. Other properties which have been investigated are the thermal stability, the thermal conductivity, and the tensile properties.

The tensile properties of the alloy were investigated at 550, 1000, and 1275°F in vacuum with a strain rate above yield of 0.02 in. per min cross-head speed. Flat-plate-type test bars were used for these tests and the results are listed in Table I.

The data reported in Table II are taken from a mean curve drawn through the experimental points. The absolute accuracy of the reported values is estimated to be ± 5 per cent.

Thermal-stability tests were conducted on specimens of the alloy approximately 1/2 in. in diameter by 1 in. long. Four such specimens were cycled 250 times in the temperature range from 150 to 725°C. Two of these specimens exhibited very little visual deformation, but they had grown approximately 3.1 per cent in length. The other two specimens were deformed to a greater extent and had grown >6 per cent in length with a corresponding decrease in diameter.

TABLE I

Tensile Properties of Fissium Alloys at 550, 1000, and 1275°F

Testing Temperature, °F	Young's Modulus, psi	0.2 Per Cent Offset Yield Strength, psi	Ultimate Strength, psi	Elongation in 2 in., per cent	Reduction in Area, per cent
550	9.9×10^6	(a)			
1000	8.6×10^6	12,640	32,630	13.3	25
1000	9.8×10^6	15,000	43,300	8.6	17
1275		(b)	2,120	100(c)	
1275		(c)	2,330	100(c)	

- (a) Broke in grip before reaching 0.2 per cent offset. Stress in reduced section at maximum load was 77,500 psi.
 (b) Temperature too high for satisfactory yield-strength determination.
 (c) Specimens elongated to limit of vacuum tank but did not fracture.

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TABLE II

Thermal-Conductivity Data From Fissium Alloy

Temperature, °C	Thermal Conductivity, w per cm ² per cm per °C
20	0.110
100	0.143
200	0.163
300	0.211
400	0.260
500	0.296
600	0.334
700	0.370
800	0.407
900	0.441

CORROSIONMechanism of Uranium Corrosion

As part of the program to develop corrosion-resistant fuel elements for atomic reactors, a study of the mechanism of aqueous uranium corrosion is under way. Preliminary experiments have been carried out to investigate the effect of the pH of the corroding water on the corrosion rates.

The pH of the corroding water was varied by making buffered sodium acetate-acetic acid solutions. Samples tested in such solutions at 60°C developed a blue film after 8 hr but did not change weight significantly during 48 hr. No visible difference could be seen over the pH range of 6.5 to 5.0. Similar tests carried out in boiling solutions showed no significant difference when the pH was varied.

An experimental program has been planned aimed at determining basic information as to the mechanism of the reaction of uranium with water. Various physical-chemical approaches are being considered. The initial study consists of an investigation of the characteristic reactions of high-purity uranium and water under different conditions. Equipment is being assembled to study the reaction of uranium at elevated temperatures in moist-argon atmospheres. The corrosion will be followed by measurements of the evolved hydrogen and weight changes and by X-ray examination of the resulting films.

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RADIATION EFFECTS

Radiation Stability of Zirconium-Uranium Alloys

Work is continuing on the radiation-effects program for the Zr-U alloys. The initial investigation was carried out on both Zr-22 and 40 w/o U alloy specimens which had been irradiated by KAPL at about 500°C.

An objective of this investigation was to determine whether postirradiation heat treatment could give usable information about the radiation stability of Zr-U fuel alloy at elevated temperatures. Three irradiated specimens were heated isothermally at 725°C for 50 hr. Two irradiated specimens were given a thermal-cycling treatment from 400 to 700°C for 226 cycles and 100 cycles.

Data obtained from this experiment are given in Table III together with the irradiation data. Two specimens which were 1-in.-long center sections [redacted] [redacted] composed of a 22 w/o uranium core clad with zirconium exhibited marked volume expansions of about 25 per cent after heat treatment. The fuel element had expanded about 2.4 vol per cent during its irradiation. Other fuel elements irradiated and cycled above 600°C had volume expansions ranging from 10 to 20 per cent. After postirradiation heat treatment, the specimen core was bulged beyond the cut end of the cladding. There was extensive cracking in the cut core ends.

The 41 w/o uranium alloy specimens had been irradiated by KAPL as single bare rods. Two specimens were isothermally heat treated. The volume expansion of the two specimens measured about 45 per cent. They both showed extensive surface cracking.

These data indicate that postirradiation heat treatment can produce physical deformation equal to and even greater than that produced by irradiation at temperature.

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TABLE III

Postirradiation Heat Treatment of Zirconium-Uranium Alloys^(a)

Uranium Content (Balance Zirconium), w/o	Burnup Total a/o	Heat Treatment	Expansion, volume per cent	
			From Density	From Dimensions
22	1.2	50 hr at 725°C	24	26
22	1.2	226 cycles, 400 to 700°C	29	22
41	4.65	50 hr at 725°C	33	46
41	1.68	5 hr at 725°C	36	46

(a) Irradiated at approximately 500°C.

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HANFORD ATOMIC PRODUCTS OPERATION - Reported by S. H. BushTensile Properties of Irradiated Uranium Alloys

A preliminary examination of certain alloys of uranium has been completed; this investigation is one aspect of the evaluation of damage occurring in fissionable materials on irradiation. Three alloys were selected for testing. Two specimens of epsilonized U-23 a/o Si, two specimens of natural uranium, and one each of U-0.35 a/o Cr and U-0.5 a/o Ti were prepared. Standard 0.250-inch ASTM tensile specimens were machined from available stock. Each sample was inserted in a Zircaloy-2 capsule containing NaK as a heat transfer medium, and the completed assemblies were irradiated at the Materials Testing Reactor for one cycle. The estimated integrated neutron flux was 2.7×10^{20} nvt; this has not been confirmed by burnup measurements.

The specimens were returned to HAPO and examined in the Radiometallurgy facility. Pronounced warping occurred in four of the tensile bars. Such warping has been observed in previous exposures of natural uranium specimens and has been tentatively attributed to flux asymmetry. Due to this warping, results are semiquantitative at best.

An examination of the surfaces of the specimens was made. In the case of the U-23 a/o Si, one of the two specimens was unwarped and both specimens' surfaces retained their pre-irradiated smoothness and brightness. The uranium controls were warped, and the surfaces were roughened, essentially the same as observed in previous irradiations. The U-0.35 a/o Cr specimen was only slightly warped and relatively smooth, whereas the U-0.5 a/o Ti was very severely warped and roughened, so much that it could not be tested.

The room temperature tensile properties were obtained on a Tinius Olsen Tensile Tester using a microformer extensometer so that the stress-strain curve could be autographically recorded to fracture. The tensile properties of the unwarped U-23 a/o Si alloy were very low; the ultimate tensile strength was 31,000 psi and elongation was 0.17 per cent in a one-inch gage length. The fracture was typical

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of a ceramic material rather than of a metal. The natural uranium had an ultimate of 51,000 psi and an elongation of 0.18 per cent, quite a bit lower than observed previously; this is attributed to warpage. The U-0.35 a/o Cr had the highest ultimate strength observed in any irradiated uranium specimen tested to date. The ultimate was 106,000 psi; 0.2 per cent yield strength was 106,000 psi, and elongation was 0.64 per cent.

Control specimens of the alloys in the unirradiated state are awaiting burnup measurements. After these have been made, and the temperatures calculated, capsules will be heated to simulate the thermal history; then the samples will be tested at room temperature.

This testing program will be extended to other alloys to observe the effects of such alloy additions on the properties of irradiated fissionable materials. (L. F. Kendall, R. G. Wheeler, W. S. Kelly, and S. H. Bush)

KNOLLS ATOMIC POWER LABORATORY - Reported by C. A. Bruch

Zirconium-Uranium Equilibrium Phase Diagram

The work on the delta phase region of the zirconium-uranium equilibrium system has been completed.

The diagram shown in Figure 1 has been constructed from data obtained by thermal and microscopic analyses of alloys over the composition range 34 to 64 weight percent zirconium. The phase fields have been labeled according to the diagram presented by Saller and Rough¹. The system as shown is characterized by a peritectoid reaction occurring at 617°C and approximately 39 weight percent zirconium, and a eutectoid reaction occurring at 606°C and approximately 54 weight percent zirconium. The single phase delta regions extend from 42.5 ± 1 weight percent zirconium to 50.3 ± 1 weight percent zirconium at 550°C, with an increase in the solubility of the delta phase on the uranium rich side to approximately 39 weight percent zirconium at 617°C. (J. F. Duffey)

Comments on the Structure of the δ Phase in the U-Zr System

The δ phase in the U-Zr system is an ordered state of the γ phase. The reflections added by ordering can be indexed on the basis of a body-centered unit cell with an a_0 three times the a_0 of the γ unit cell.² Of these new reflections, none have been observed, either at this laboratory or at Argonne,³ with a zero index; that is:

$$I_{hko} = 0$$

unless

$$h/3 = n \text{ and } k/3 = n^1$$

1. BMI-1000, (1955), p.66.

2. M. H. Mueller, Acta Crystallographica 8 849-850 (1955).

3. M. H. Mueller, Argonne Lab. Personal Communication.

where n and n^1 are integers. In other words, the reflections $h k o$ observed are the reflections which are present in the unordered state, and which on the basis of the new large cell have their original indices multiplied by three.

This leads one to believe that, if the structure is a true superlattice, three times the original length in each direction, then there is no change after ordering in the electron density when projected along any one of the major axes of the supercell. For $\rho(xy)$ the projected electron density is given by:

$$\rho(xy) = \sum_h \sum_k F_{hko} e^{2\pi i(hx + ky)}$$

and:

$F_{hko} = 0$, Structure factors for any new superlattice reflections

$F_{hko} \neq 0$, Structure factors for original γ reflections indexed on basis of new cell.

Thus, since the Fourier coefficients of the projected electron density are the same before and after ordering, there is no change in projected electron density. Inasmuch as there are three lattice points containing atoms above each projected point in the superlattice, and all points in projection have equivalent electron densities, one is led to conclude that the ideal composition of the δ phase is UZr_2 (U_2Zr being ruled out by the phase diagram). By ideal composition it is meant that composition which can give perfect ordering throughout the crystal.

This ideal composition UZr_2 is about the limiting composition on the U side of the δ phase region⁴. There is a large solubility of Zr, but little or none of U, in the UZr_2 .

This argument for the ideal composition being UZr_2 does not depend upon whether the large cubic supercell is the primitive unit cell. The true primitive unit cell may not be cubic at all.* This argument for the UZr_2 composition depends only on

4. J. Duffey, Uranium Alloy Newsletter (This issue).

*For example, the true unit cell may be rhombohedral, and consequently can be indexed on a body-centered cubic basis.

whether the cubic cell chosen is a permissible crystallographic cell. Inasmuch as our powder patterns and the Argonne single crystal data can be well indexed on the large cube basis, it is reasonable to assume that the cell is a crystallographically permissible one. (E. R. Boyko)

Irradiation of Uranium-Zirconium Alloys

The purpose of this investigation was to compare the irradiation behaviors of the gamma and delta phases in the uranium-zirconium alloy system at two substantially different temperatures.

A zirconium-47.5 wt % uranium alloy was prepared by arc-melting and casting in a small cylinder. The "finger" was sealed in a steel jacket and hot swaged to about 0.150" diameter. The jacket was removed by pickling and the alloy given one cold-swaging pass and then centerless ground to 0.100" diameter. Samples, about three-quarters inch long, were cut for irradiation purposes.

These samples were given various heat treatments, after which they were irradiated in NaK for three cycles at MTR in a position with a flux of about 1×10^{14} nv. Some of the samples were irradiated in capsules with a controllable calrod-type heater wound on the outer surface. In this way the temperature could be maintained at selected elevated temperatures with variations of less than 5°C during pile operation.

Two samples were irradiated at 650°C , which is about 50° above the order-disorder transformation temperature: that is, well in the γ -phase region. These samples required no prior heat-treatment. Two samples were gamma-annealed for about an hour (at 800°C) and were quenched to retain the gamma phase. These were irradiated at ambient water temperature (about 90°C). Four samples were gamma annealed and then furnace cooled to ensure complete ordering to the delta-phase. The ordering of these samples was confirmed by metallographic examination using polarized light. Two of these specimens were irradiated at 90°C and two were irradiated at 550°C , about 50° under the transformation temperature.

The table gives the results of post-irradiation measurements on the samples together with the calculated burn-ups.

Sample	at % B.U.	Temp.	$\frac{\Delta d}{d} \times 100$	$\frac{\Delta L}{L} \times 100$
Ordered No. 1	0.25	550°C	.8	.29
Ordered No. 2	0.25	550°C	.4	.61
Untreated No. 1	0.29	650°C	.3	.98
Untreated No. 2	0.29	650°C	.6	.64
Gamma quenched 1	0.38	90°C	-.3	.59
Gamma quenched 2	0.38	90°C	.1	.88
Ordered No. 3	0.38	90°C	.6	-.10
Ordered No. 4	0.38	90°C	1.1	.29

There was no evidence on any of the samples of blistering or surface roughening.

Except for the dimension changes, the only evidences of damage were:

(1) The ordered samples irradiated at 550°C developed scaly surface marking for a distance of 50 to 80 mils from each end.

(2) Both of the ordered samples irradiated at the lower temperature were warped into a curvature of about 4 inches radius.

In the absence of further X-ray diffraction and metallographic studies (both of which are planned), the following conclusions seem warranted:

(a) There is no significant difference in the irradiation stability of the ordered and disordered phases at temperatures near the transformation temperature.

(b) At low temperatures, it is possible that the ordered phase disorders under irradiation. If there is a volume change on ordering, this could account for the warping observed, since one side of the sample would receive a greater nvt than the other due to flux anisotropy and self-shielding effects.

(c) If the conclusion in (b) is correct, then there is no ordering of the quenched gamma phase produced by irradiation. (D. D. Van Horn)

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FIGURE 1 THE DELTA PHASE OF THE URANIUM-ZIRCONIUM EQUILIBRIUM SYSTEM

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NATIONAL BUREAU OF STANDARDS - Reported by John J. ParkUranium-Platinum System

The uranium-platinum system apparently contains only one congruently melting compound, UPt_3 , which melts in excess of 1500°C . Two melting point determinations on an alloy apparently consisting entirely of this compound were run, with results indicating that it melts at approximately 1700°C . The results of back reflection X-ray data show that the solid solubility of uranium in platinum is about 4.5 a/o at room temperature and is about 4.7 a/o at 1000°C . Additional X-ray data of powders of uranium-platinum alloys give further basis for previous conclusions that this system contains three compounds in the region of 45 to 70 w/o of platinum; these compounds are the UPt , UPt_2 , and UPt_3 phases.

Uranium-Rhodium System

Results of thermal analysis thus far indicate that rhodium lowers the melting point of uranium to about 872°C with the formation of a eutectic at about 8 w/o of rhodium. In addition, rhodium lowers the gamma-beta transformation temperature to about 685°C and it lowers the beta-alpha transformation temperature to about 625°C . A reaction horizontal is also detected at about 720°C .

Uranium-Ruthenium

Tentative results on quenching experiments reveal fusion at 900°C , and thermal analyses indicate that the previously proposed eutectic arrest at about 885°C is essentially correct. Other quenching experiments to ascertain the decomposition temperature of the proposed Ru U compound indicate that this compound is stable up to at least 1140°C .

Uranium-Osmium

Additional thermal analysis results have shown the possibility of a compound of approximately 16 a/o osmium. Between this compound and the next phase of higher osmium content a eutectic arrest is noted in the region of 27 a/o osmium at a temperature of about 960°C . Osmium alloys of 18 and 29 a/o osmium have shown a highly pyrophoric nature upon removal of the alloys from the thermal analysis furnace;

however, only these two alloys reacted in this manner, while a 24 a/o alloy was not reactive. In addition, alloys sealed in vycor tubing under a partial pressure of argon and placed in a furnace for homogenization treatment have reacted in such a way as to shatter the vycor tubing.

Uranium-Palladium

Thermal analysis results indicate the transformations of uranium are detectable at least as far as 40 w/o or 60 a/o palladium. The first compound in this system appears to be situated at 75 a/o palladium, or UPd_3 .

Uranium-Iridium

Quenching experiments indicate that the solid solubility of iridium in the gamma-uranium field is at least 2.5 w/o or 3.1 a/o.

NUCLEAR METALS, INC. - Reported by Saul Isserow

Uranium-Silicon Epsilon Alloy

Water quenching of a coextruded rod as it left the press did not reduce the core-clad interaction. The quenching embrittled the U-Si core alloy considerably, leading to fracture during subsequent machining. Interposition of a copper electroplate was also an ineffective method of suppressing the core-clad interaction. The copper plate apparently disrupted the flow pattern to such an extent that the resulting rods had numerous bulges. (S. Isserow)

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

The study of the interdiffusion heat treatment (diffusion anneal) of U-2 w/o Zr rods with defective Zircaloy-2 cladding has continued. Diffusion annealed specimens which are continuing to survive corrosion testing in 650°F water or 750°F steam include the following: rods with 10 mils thick cladding containing a 2 mils diameter drilled hole defect; rods with 30 mils thick cladding containing either uranium wire pre-extrusion defects, or 7, 16 or 31 mils diameter drilled hole defects. Both the annealed and unannealed rods with the previously described defective end seals show no signs of corrosion failure. Diffusion anneals at 800°C for 24 hours or at 950°C for 2 hours show promise of providing good corrosion resistance. A treatment at 950°C for 1 hour apparently cannot be relied on to prevent corrosion failure.

Diffusion annealed specimens have been given additional heat treatments, which are adapted from treatments reported to promote irradiation stability. The preliminary results indicate that a supplementary treatment at 800°C does not harm the corrosion resistance while a 690°C treatment may reduce the corrosion resistance.

The amount of uranium in the 0.05-0.06 mil thick outermost layer of diffusion annealed rods with 5 and 10 mils thick cladding has been determined to be about 0.18 w/o and 0.13 w/o respectively. Additional tests will be made to determine whether a uranium-free surface can be obtained without sacrificing corrosion resistance.

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Heating to 1050°C for as little as 20 minutes forms a strong bond in rods where little or no bond existed between the core and the cladding. A diffusion anneal at 1050°C for 16 hours appears to have prevented the immediate corrosion failure of such rods whose cladding contained drilled hole defects.

SYLVANIA ELECTRIC PRODUCTS, INC. - Reported by Fred E. Bowman

Thermal Cycling

Preliminary thermal cycling results have been obtained on some of the ternary alloys which are currently receiving consideration. Samples were cycled from 200°C to 720°C at an intermediate rate, i.e., the samples, in evacuated Vycor tubes, spend ten minutes in the hot furnace and ten minutes in an air blast. The time-temperature relationships have been established with thermocouples attached to the specimens.

Table I lists the data which have been accumulated to date. All length and diameter changes noted are those resulting from 500 cycles between 200°C and 720°C. The maximum dimensional change noted for each sample is that reported.

TABLE I

The Effect of 500 Cycles From 200°C to 720°C on
Low Addition Ternary Uranium Alloys

Composition	$\Delta L, \%$	$\Delta D, \%$	Remarks
0.5 Mo; 0.5 Si	1.3	0.3	Smooth surface.
0.5 Mo; 0.5 V	1.9	1.8	Surface slightly roughened.
0.5 Mo; 0.5 Ti	3.4	3.9	Surface slightly roughened.
0.5 Mo; 0.5 Nb	1.3	3.4	Surface rough.
0.5 Mo	11.0	3.4	Warped, surface rough.
1.0 Mo; 0.1 C	3.4	3.2	Warped, cracked.
0.5 V; 0.5 Nb	3.0	2.5	
0.5 V; 0.5 Si	3.0	1.9	Surface smooth.
0.5 V; 1.0 Cr	1.6	5.3	Surface rough, barreled.
0.5 Nb; 0.1 C	50.0	3.8	
1.0 Nb; 0.25 Si	2.2	0.3	Surface good, ends dished.
1.0 Nb	16.0	4.8	Rough surface.
0.5 Ti; 0.5 Nb	1.9	8.3	Surface rough, ends distorted.
0.75 Mo; 0.75 Nb	8.5	6.1	Surface very rough.

The above results were, for the most part, obtained from single specimens. All represent material which has been cold pressed and sintered to maximum density.

Although no definite conclusions can be drawn as yet, several of the combinations of alloy additions appear promising and worth further investigation. This is particularly true of the silicon additions in combination with molybdenum and niobium.

As mentioned in the previous issue of the Newsletter, a major effort is being directed toward the combination of an alpha soluble material with an essentially insoluble addition. Silicon, carbon and aluminum are typical examples of the latter. The meager evidence to date suggests that basically, the principle may be valid, based upon the behavior of the silicon alloys in combination with molybdenum and niobium. In the case of the carbon additions, however, it appears that with such carbide formers as molybdenum and niobium, the alpha strengthening effects of these elements are lost, due to their tendency to come out of solution and form carbides. It is expected that the aluminum will behave in a manner similar to silicon.

Thorium-Uranium Alloys

Preliminary studies of solid solubility have been conducted employing a vapor deposited layer of uranium on a thorium surface cleaned by sputtering. After 13 days at 700°C, all evidence of the uranium on the surface had disappeared, indicating some solubility. Additional experiments are being performed to determine relative solubility and diffusion rates.

Powder densification by powder rolling techniques have been demonstrated to be satisfactory for an 80 w/o Th-U alloy. Densification is excellent and the distribution of uranium is far better than that obtainable by working cast alloys. Microexamination of this material at 1000 magnification has revealed a third phase appearing as a peripheral layer surrounding each uranium particle. This layer remains intact after the uranium is removed by etching. Attempts are being made to identify this material.

Once a technique for decreasing the circumferential distortion of the core has been established, powder rolling appears to offer an excellent method for the simultaneous fabrication and cladding of a thorium-uranium fuel element.

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WESTINGHOUSE ATOMIC POWER DIVISION - Reported by J. Hino

Uranium-Zirconium Alloys

Dilatometer measurements of the transformation behavior of gamma-quenched 50 and 60 w/o U + Zr alloys reveal that the metastable phase starts transforming to ϵ in a few seconds when reheated to temperatures between 200°C and 500°C. The reaction goes to completion in a few minutes at temperatures between 425°C and 500°C.

Hardness measurements on a 50 w/o U + 50 w/o Zr alloy quenched and transformed between 260°C and 575°C show a continuous increase as transformation proceeds to completion. The quenched (γ phase) hardness value is 200 DPH. This value is raised to about 400 DPH for fully transformed material. The magnitude of the hardness increase appears to be the same regardless of the transformation temperature, therefore hardness is thought to be a dependable measure of transformation. Calculation of the activation energy from a $\ln$ time vs $1/T$ ($^{\circ}\text{K}^{-1}$) graphical plot at an arbitrarily chosen hardness level yielded a value of 46,000 cal/mole. This value is between the reported activation energy values (43,000 and 47,000) for diffusion in U-Zr.

X-ray diffraction examination of the 50 w/o Zr alloy show that the appearance of the unique reflections (superlattice lines) of the ϵ phase is not a sensitive method for following the $\gamma \rightarrow \epsilon$ transformation. These reflections are diffuse and become distinct only after considerable transformation has occurred. (J. J. Kearns)

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Irradiation of U-Zr

In recent irradiation tests conducted by KAPL on Zircaloy-clad zirconium-8 w/o uranium fuel elements _____, it was observed that the irradiation induced volume changes resulting from 1.1 to 1.5 a/o burnup caused the Zircaloy-2 cladding to rupture. In order to establish the irradiation stability of fuel plates in the PWR seed core, two irradiation experiments consisting of both bare and clad Zircaloy-2 - 6, 8, 10, and 12 w/o uranium (fully enriched) alloys have been initiated.

The first experiment consist of the irradiation of 24 Zircaloy-2 clad platelets and 48 bare platelets are in the as-fabricated condition, while the bare platelets are being irradiated in the following:

- a. As fabricated
- b. 900°C - 2 hrs - W.Q.
- c. 900°C - 2 hrs - W.Q. + 360°C - 2 days - W.Q.

These samples were charged into reflector positions in the MTR on June 11, 1956, and are scheduled for a one year irradiation, which should yield a maximum burnup of 40% of the U^{235} atoms. Temperatures of fuel and cladding during irradiation approximate the temperature conditions expected in PWR.

The second experiment consists of the irradiation of 16 clad platelets in MTR process water. All platelets are in the as-fabricated condition. These samples are scheduled for insertion in an MTR reflector position July 23, 1956, and are to be irradiated 4 reactor cycles to an estimated maximum burnup of 40% of the U^{235} atoms. This experiment permits rapid burnup, but the temperatures of the fuel and cladding are considerably lower than PWR design conditions.

Following irradiation, measurements of dimensions, density, and degree of warpage will be made on all samples. (L. J. Jones)