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UNITED STATES ATOMIC ENERGY COMMISSION

RESUMÉ OF URANIUM ALLOY DATA-X

By

E. F. Losco

Z. M. Shapiro

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For The Atomic Energy Commission

H. R. Canale
Chief, Declassification Branch

November 14, 1955

Bettis Plant

Westinghouse Electric Corporation

Pittsburgh, Pennsylvania

Technical Information Extension, Oak Ridge, Tennessee

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Resume of Uranium Alloy Data - X

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November 14, 1955

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~~CONFIDENTIAL~~Resume of Uranium Alloy Data - XINTRODUCTION

The principal developments in the PWR uranium fuel program since the last panel meeting follow:

1. Bond-Line Attack in Coextruded U-3.8 w/o Si Alloy

Samples of U-3.8 w/o Si clad with Zircaloy-2 by coextrusion are subject to rapid attack in 650°F water at the core-clad interface. It has been definitely established through X-ray diffraction studies on samples which were heat treated to form thick interdiffusion layers that two important layers form. One of these is the silicide of zirconium, $ZrSi_2$, on the clad side of the interface. Just below this is a layer corresponding to alpha uranium. The presence of the alpha uranium layer adjacent to the core alloy is believed to be responsible for bond-line attack. It is apparent from this that fuel elements prepared by direct bonding of Zircaloy-2 to U_3Si will be subject to rapid bond-line attack unless a barrier layer is interposed between cladding and core. Furthermore, materials the free energy of reaction of which with silicon is greater than that of uranium are not suitable as barrier layers since the surface of the fuel would be depleted of silicon (to form the silicide) leaving behind the alpha uranium layer. Evidence for this was obtained in the X-ray studies of direct bonded elements and was confirmed by corrosion tests of elements prepared with niobium and molybdenum barriers. The latter barriers form silicides and result in bond-line attack at the core-barrier interface.

2. Aluminum-Base Alloys as Barrier Layers for U_3Si Fuel Elements

Aluminum-base alloys (which do not form silicides) show considerable promise as barrier layers. Fuel elements prepared by brazing extruded U_3Si rods to Zircaloy-2 tubes with Al-6 w/o Cu have satisfactorily withstood 35 days test in 650°F water. However, difficulties have been encountered in bonding of the brazing alloy to the Zircaloy at temperatures below about 950°C. Use of such high brazing temperatures is not practical because of excessive reaction of aluminum with the core alloy. To offset this difficulty and permit brazing to be carried out at reasonably low temperatures, it is necessary to precoat and bond the Zircaloy surface with a suitable metal prior to brazing. Preliminary tests indicate Al-5 w/o Si can be easily bonded to Zircaloy at about 950°C. Using this as a surface layer and also as the brazing alloy excellent joints were obtained between barrier and cladding and barrier and core at temperatures as low as 800°C. A more practical solution being investigated involves precoating the Zircaloy with zinc by deposition from a fused $ZnCl_2$ bath. Work is progressing along these lines.

3. Corrosion Mechanism for Bare U-3.8 w/o Si Alloy

Bare duplex melted samples of U-3.8 w/o Si ingots epsilonized for 7 days at 800°C in general display better corrosion resistance in 650°F water than extrusions prepared from the same ingots. Deterioration of the corrosion properties of extrusions is attributed to the presence of excess

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alpha which is strung out in the rolling direction. In the absence of residual alpha the extrusions corrode at a rate comparable to the cast ingots. The addition of 0.2 w/o Mo, Nb, or Cu seems to be beneficial in eliminating excess alpha and in making the silicon content of the alloy less critical for practical purposes. In any case, it appears that use of cast, rather than wrought, U_3Si rods is more desirable for fuel elements since any excess alpha would then be uniformly distributed throughout the matrix.

The presence of carbon as an impurity in U-3.8 w/o Si alloy markedly reduces the peritectoid transformation. Early work by G. Bitsianes (Alloys of Uranium and Silicon, II - The Epsilon Phase, CT-3309 dated June 1945) indicates that 1000 ppm of carbon reduces the peritectoid transformation temperature from 930°C to 830°C. Some ingots and extrusions prepared by Bettis and Nuclear Metals, Inc., contain from 400 to 1000 ppm of carbon introduced mainly during induction melting in BeO or ZrO₂ washed graphite crucibles. Heat treatment of these materials at 850 to 900°C definitely results in de-epsilonization. It is suspected also that partial de-epsilonization occurs during extrusion at 870 to 900°C. Even when the temperature is too low to cause de-epsilonization, precipitation of a second phase occurs at grain boundaries, and grain boundary attack in 650°F water is rapid. Improvements in melting practice designed to minimize carbon contamination are being investigated.

Corrosion of bare U_3Si in 650°F water appears to progress by the reaction $U_3Si + H_2O = (U,Si)O_2 + H_2 + U_3Si_2$. The delta phase U_3Si_2 appears as a white layer just below the oxide on the core surface. An attempt is being made to obtain positive X-ray identification of this surface layer. A similar phase appears to penetrate the grain boundary of extruded samples subjected to a long-time anneal (168 hr) at 800°C. This behavior is not observed on samples given the standard epsilonizing treatment. The attention of the panel members is directed to this phenomenon and its discussion at the next panel meeting is requested.

4. Irradiation Induced Transformation in U-Mo and U-Nb Alloys

Extensive metallographic examination at 250X of thermally transformed U-Mo and U-Nb samples which were irradiated in MTR reveals no indication of reversion to the gamma phase condition even though all other physical and mechanical changes (electrical resistivity, density, and hardness) are in the direction of gamma reversion. The samples are being checked at 1500X to determine if the second phase present is extremely fine grained gamma or alpha. The presence of alpha is highly unlikely in view of the high values of resistivity obtained. This was confirmed in heat treatment tests on unirradiated and thermally transformed samples of U-9 w/o Mo. It was found that simple disordering of the epsilon phase by heating in the alpha plus gamma field does not result in a resistivity increase comparable to that observed in irradiated samples. The resistivity corresponding to that obtained for irradiated samples is obtained only if alpha is completely dissolved by heating in the single phase gamma field.

5. Magnetic susceptibility measurements of UO_2 oxidized at low temperatures to O/U ratios of 2.03 to 2.67 show the persistence of the UO_2 phase to a ratio of 2.31 with oxygen ions entering interstitial positions in the lattice, and the presence of an orthorhombic phase (U_3O_8) at ratios in excess of 2.44, with the excess uranium created by oxygen lattice vacancies.

6. The fourth Chalk River UO_2 test is showing indications of specimen failure (presumably failure of the defected sample). Positive information will not be available until the specimen train is returned.

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I. U-3.8 w/o Si Alloy Development

Since the last resume (WAPD-PMM-262) was prepared, a clearer understanding was obtained of the problems involved in preparing Zircaloy-2 clad U-3.8 w/o Si fuel elements having satisfactory corrosion resistance in 650°F water. Work was continued on the investigation of coextruded rods, with and without metallic barrier layers, and elements prepared by brazing the fuel rods into extruded Zircaloy tubes with aluminum-base brazing alloys. In addition, work designed to establish the factors affecting the corrosion resistance of the fuel material itself was expanded to include heat treatment, composition, and microstructural effects.

A. U-3.8 w/o Si Coextruded with Zircaloy-2 Cladding

Simulated fuel elements were prepared from coextrusions numbered 26-1, 27-1, and 28-1. These were defected with a single 0.040 in. diam hole penetrating through the cladding into the fuel and tested in 650°F degassed water. All elements failed within 28 days of test. Metallographic examination of failed elements and corrosion tested bare-end samples taken from the same extrusions indicate that failure occurs by attack of the core-clad interface. Since directly bonded elements have consistently failed in this manner, no further corrosion tests are contemplated on fuel elements prepared in this manner.

An X-ray diffraction study was made of the interdiffusion layers formed between Zircaloy-2 and U-3.8 w/o Si. A section 1 in. long x 0.31 in. diam, cut from a Zircaloy-2 clad extrusion having a U-3.84 w/o Si core was sealed in a Vycor bulb in a helium atmosphere and annealed 14 days at 800°C. The sample was then sectioned longitudinally and a 3 degree cut made on one edge traversing all diffusion zones. Fig. 1 shows three diffusion zones revealed by the sectioning procedure. X-ray powder photographs were taken of each diffusion zone by a special adaptation of a Weissenberg camera. The photographs obtained were adequate but not of the best quality because the beam striking the sample was too large and overlapped the diffusion zones. To avoid this, small samples 0.5-1.0 mm long were cut from each diffusion zone, the Zircaloy cladding, and the U₃Si core. Patterns obtained from these samples were clear and easily interpreted.

The results obtained are listed in Table I and compared with available references. The following was noted:

1. The Zircaloy-2 cladding is normal.
2. Zone 1, the layer below the cladding (having a Widmanstätten structure), is essentially zirconium with lattice constants corresponding to those of Zr.
3. Zone 2, immediately below Zone 1, is ZrSi₂.
4. Zone 3, adjacent to the core alloy, corresponds to alpha uranium.
5. The U₃Si core is normal and checks well with the literature.

It is apparent from Fig. 1 and the above findings that uranium atoms diffuse into the Zircaloy cladding to form Zone 1. At 800°C the diffusion of U into Zr changes the composition to the extent that beta Zr is formed and on subsequent cooling this transforms back to alpha Zr. Silicon from the U_3Si core alloy builds up in the cladding to form $ZrSi_2$ -- or a mixed Zr-U silicide (Zone 2). Its migration from the core leaves a uranium-rich region that corresponds to alpha U (Zone 3). The surface area of the core is depleted in Si but this depletion is overcome by diffusion of Si from the residual epsilon (U_3Si_2) phase to maintain the U_3Si composition at the surface of the core. This may readily be seen in Fig. 1. The surface of the core is essentially free of U_3Si_2 particles. Below the surface these particles are still present.

It appears therefore that the presence of a corrodible layer of uranium in directly bonded extrusions is responsible for interface corrosion of defected fuel elements. Apparently, this layer must exist to some extent in as-extruded material. Defected fuel elements prepared in this way could hardly be expected to have satisfactory corrosion properties. Further, the migration of Si out of the core to form a metallic silicide with the consequent development of a highly corrodible layer of alpha uranium below it suggests that the corrosion resistance of clad U_3Si fuel elements can be improved only by interposing a barrier material which does not form a silicide preferentially to uranium.

Only a few elements may be suitable as barrier layers which do not form silicides. These include Al, Be, Ag, Sn, Sb, Au, Tl, and Pb. It is planned to investigate the most promising of these in connection with the barrier layer program.

B. U-3.8 w/o Si Coextruded with Metallic Barrier Layers

Defected fuel elements, prepared from coextrusions having barrier layers of niobium and molybdenum, interposed between Zircaloy-2 and the core alloy were tested in 650°F water with the results shown in Table II. Two extrusions were prepared with a niobium barrier. The first (No. 1) showed unbonding between barrier and core and failed quickly by bond-line attack. The other (No. 3) was well bonded but also failed early (within 14 days) by attack at the barrier-core interface. The same results were obtained for extrusion No. 8, containing a molybdenum barrier.

Other extrusions prepared with U-10 Nb, Mo, and Cu barriers have not been tested as yet. However, all of these barriers form silicides. Based on the X-ray findings indicated in the previous section it is not expected that these barrier materials will solve the bonding problem. Now that the requirements for a successful barrier are suspected, materials which do not form silicides will be investigated.

C. Clad U-3.8 w/o Si with an Aluminum Alloy Barrier

The preliminary tests involving preparation of fuel elements by brazing extruded rods of U-3.8 w/o Si inside Zircaloy-2 tubes using Al-6 w/o Cu as the brazing alloy appeared very promising. Several fuel elements were fabricated using Zircaloy-2 tubes closed at one end and capped after brazing. The U-3.8 w/o Si rods, coextruded with Zircaloy-2 cladding, were stripped of the cladding to within $3/8$ in. of one end. The latter were then inserted into the closed end tubes, the unmachined Zircaloy on the rods serving to center the rods in the tubing. Thus a 0.035 in. gap existed between the U-3.8 w/o Si rod and the inside of the tubing except at the bottom where the centering ring of Zircaloy was located. The Al-Cu alloy was melted (in vacuum) at the top end of the U₃Si rod and allowed to flow down to fill the gap between rod and tubing. After brazing, the open end of the element was end plugged, a 0.040 in. diam defect introduced through the cladding and brazing alloy and the elements corrosion tested in 680 and 650°F water.

Two of the above elements were tested in 680°F water. One of them failed on the 16th day of test when the autoclave was inadvertently overheated and the samples were exposed to 750 to 800°F steam for about 8 hr. Examination of the sample indicated that the Al-Cu alloy was well bonded to the U₃Si but was not bonded to the Zircaloy tube. Sections through this rod at the end cap and defect hole are shown in Fig. 2a and 2b respectively. (Note in Fig. 2a the longitudinal splitting of the rod which is related to alpha stringering of the extrusions.) Water apparently infiltrated the tube-barrier interface to the Zircaloy spacer ring (which was not bonded to the U₃Si rod) resulting in oxidation of the U₃Si rod and splitting of the tube cladding at this location. The relatively small amount of corrosion of the U₃Si and the apparent extrusion of Al into the defect should be noted in Fig. 2b. The other sample was not adversely affected and was continued in test in 680°F water. After 29 days of test slight bulging was noticed at the spacer ring location but creep of the Al-Cu barrier seemed to have essentially blocked the defect. Despite the bulge the rod has survived 36 days of test and is continuing in test.

Four similar elements are being tested in 650°F water. These have survived 35 days of test. One rod has bulged in several positions apparently because of incomplete filling by the brazing alloy. The others appear satisfactory except for slight pimpling at the defects. The rods are continuing in test.

Continued work on brazing indicates that good metallurgical bonds are obtained between the Al-6 w/o Cu alloy and the U₃Si rod at temperatures of about 800°C, but the brazing alloy will not wet and bond to Zircaloy unless the temperature is raised to about 950°C. When this is done, the Al-Cu alloy severely attacks U₃Si. It was also found that the Al-Cu alloy has relatively poor flowability and will not fill out gaps between cladding and core that are less than about 0.030 in. thick when brazing is carried out at 850-900°C. When gaps of this size are used shrinkage voids are observed within the Al-Cu barrier layer and bonding to Zircaloy is unsatisfactory.

Al-Si alloys have also been considered as brazing alloys. Welding rods of Al-5 w/o Si and Al-12 w/o Si appear to have good corrosion resistance, surviving 21 days in 680°F water. These alloys have better flowability than the Al-Cu alloy and lower thermal expansion coefficients. Wetting tests of Al-5 w/o Si on Zircaloy, however, indicate that 950°C is required for bonding as is the case for Al-6 w/o Cu. Brazing tests indicate that barriers may be made as thin as 0.010 in. at 850-900°C but no bonding to Zircaloy results.

In an effort to solve the brazing problem the feasibility of precoating the inside of the Zircaloy tube with a material that will permit brazing at 800-850°C or below is being investigated. A rod of the Al-5 w/o Si alloy was inserted in a closed end Zircaloy tube heated rapidly to 1000°C and immediately cooled. The alloy was well bonded to the Zircaloy and, in fact, resulted in a 0.007 in. reduction in the original 0.030 in. Zircaloy thickness by solution. The alloy was machined out, leaving 0.030 in. of cladding containing the bonded diffusion zone. Using the latter a rod of U_3Si was brazed as before at 800-850°C with Al-5 w/o Si and a 0.030 in. gap. The gap was easily filled with no shrinkage cavities appearing in the solidified brazing alloy. A cross section through this rod is shown in Fig. 3. It can be seen that a good bond exists at both the clad-barrier and barrier-core interfaces. Attempts are currently being made to form the initial aluminum layer by vapor deposition and a subsequent anneal.

Efforts are now under way to investigate the possibility of using deposited layers of Zn, Fe, and Ni on the Zircaloy, diffusing these to form a bond before carrying out the brazing operation with promising Al-base alloys. The use of a deposited layer of Zn appears quite promising in reducing the brazing temperature and permitting the use of very thin brazed joints.

D. Epsilonization of Duplex Melted U_3Si Ingots and Extrusions

It was previously indicated that it is necessary to homogenize and epsilonize the cast ingot structure prior to extrusion to attain good corrosion resistance. The standard epsilonizing treatment employed by Bettis and Nuclear Metals, Inc., to complete the peritectoid reaction of alpha (uranium) and delta (U_3Si_2) to form corrosion resistant epsilon (U_3Si) is 800°C for 7 days. To establish the effectiveness of this treatment, metallography, chemical analysis, and corrosion studies were carried out on eight duplex melted and epsilonized ingots. The results are shown in Table III.

It is apparent that none of the epsilonized ingots have a complete epsilon structure, but contain residual alpha, delta, or both. The chemical analysis does not always correlate with structure apparently because of the presence of some alloy segregation. For example, ingot No. 538, with a top and bottom analysis of 3.79 and 3.71 w/o Si, respectively, should contain residual alpha and actually contains residual delta. A detailed study of alloy segregation is planned on other ingots.

From the corrosion data in Table III it is apparent that alpha leads to a reduced or variable corrosion life. Where alpha is not present, no cracking of samples occurs and the corrosion rates are relatively uniform.

and the U_3Si corrodes in a normal manner. Of the six billet samples still in test, the 42-day corrosion rates vary from -0.62 to -0.87 mg/cm²/hr.

A program to verify whether the present epsilonizing treatment of 800°C for 7 days is optimum was initiated. A series of heat treatments were investigated on as-cast material and also on epsilonized material. The treatments are listed below.

As-Cast Alloy

800°C for 7, 14, and 21 days
850°C for 7 and 14 days
900°C for 3, 7, and 14 days
750°C for 14 days

Alloy Epsilonized for 7 days at 800°C

800°C for 7 days
850°C for 3 and 7 days
900°C for 1 and 3 days

These treatments were applied to samples from all the ingots (with the exception of ingot No. 454) listed in Table III. Only 7 days test time in 650°F water has been completed. The results, however, indicate that none of the treatments were beneficial. All the samples epsilonized at 900°C disintegrated in 0-3 days. With the exception of ingot No. 484, all the samples epsilonized at 850°C disintegrated in 0-3 days. The corrosion rates for samples from ingot No. 484 (still in test) are 2 to 4 times higher than samples from the same ingot epsilonized 7 days at 800°C. Increasing the epsilonizing time at 800°C beyond 7 days resulted in disintegration of some samples in 0-3 days. Most of these, however, remain in test but are corroding twice as fast as similar samples given the standard epsilonization treatment. The samples annealed 14 days at 750°C (after being given the standard anneal) behave similarly to those annealed for long times at 800°C.

Metallographic examination was completed on the heat treated samples from ingot No. 484 and representative structures are shown in Figs. 4 and 5. All samples came from the same ingot slice. Except for possible differences due to alloy segregation, it appears that increasing time at 750, 800, and 850°C resulted in little, if any, structural changes. However, annealing at 900°C produced a marked change in structure (Fig. 5c). It appears that the epsilon phase, U_3Si , transforms back to alpha plus delta. Samples heated 3 days at 900°C and having the indicated structure disintegrated in 650°F water in 0-3 days. Since the peritectoid transformation temperature is reported to be 930°C, the changes in microstructure obtained at 900°C were not expected.

Similar effects of heat treatment have been observed on Zircaloy-clad U-3.8 w/o Si extrusions fabricated by Nuclear Metals, Inc. Three such extrusions, which were originally intended for use on irradiation tests, were found to have a structure consisting of alpha stringers and islands of delta in a matrix of epsilon. In an effort to eliminate the alpha stringers, which deteriorate corrosion resistance (Fig. 2a) and may result in dimensional instability under irradiation, bare samples from one extrusion were heated at 800°C, 850°C, and 900°C for 3 and 7 days. The resultant microstructures indicated that de-epsilonization occurred at 850°C and 900°C (Fig. 6). The samples treated at 900°C appeared to have been in a

liquid-solid phase region. Corrosion data and density measurements were obtained on these samples (Table IV). The samples either disintegrated or had very high corrosion rates after 3 days in 650°F. Density decreased toward that of de-epsilonized material. For the 900°C treatment density was actually lower than that which would be expected for completely de-epsilonized U-3.8 w/o Si.

The unusual behavior of the above material on heat treatment below the 930°C peritectoid transformation temperature suggests that impurities are responsible for an appreciable and variable reduction in the peritectoid temperature. The NMI extrusion contained 900 ppm of carbon. Although early data on Bettis ingots indicated an average carbon content of 400-500 ppm, recent data indicate that the carbon content may reach in the neighborhood of 1000 ppm. Early work conducted at the Massachusetts Institute of Technology by G. Bitsianes (Alloys of Uranium and Silicon, II - The Epsilon Phase, CT-3309 dated June 1945) indicates that 0.1 w/o carbon in U-20 a/o Si lowers the peritectoid transformation to about 830°C. It is not surprising, therefore, to expect de-epsilonization of the subject extrusions at 850°C. Almost 200 ppm of carbon is introduced with the raw materials (U and Si). The remainder apparently is introduced during induction melting in ZrO₂-washed graphite crucibles. Attempts will be made to improve the melting practice (improved mold wash, short holding time, reduced superheating) to reduce contamination. Also some material will be melted in BeO crucibles for comparison.

E. Corrosion of Bare U-3.8 w/o Si Extrusions

Five duplex melted ingots, prepared by Bettis, were epsilonized for 7 days at 800°C and coextruded with Zircaloy-2 cladding. The ingot numbers and the extrusions made from these ingots are identified as follows:

Ingot No.	Silicon w/o	Extrusion No.	Residual Phase
452	3.93	26-1	alpha + delta
454	3.83	27-1	alpha + delta
484	3.69	28-1	delta
534	3.84	534-1	delta
548	3.83	548-1	delta

Part of each extrusion (except 548-1 which was extruded bare) was stripped of clad and corrosion tested in 650°F degassed water. Corrosion rates for the above ingots are reported in Table III and the rates for bare samples in the extruded and extruded and heat treated conditions are shown in Table V. It should be mentioned that 2 of 6 as-extruded samples from Ext. No. 26-1 cracked in less than 14 days and 3 of 6 extruded and epsilonized samples from Ext. No. 27-1 cracked in less than 14 days.

A comparison of the results in Table V with those shown for epsilonized ingots in Table III indicates that the corrosion rate of extruded material is higher than the rate for as-cast and epsilonized material and that further heat treatment on the extruded samples does not improve the material. Metallographic examination of the extrusions has disclosed that large differences in structure can be found in different locations of the same extruded

rod. Fig. 7 is an extreme example of structure variation found in Ext. No. 26-1 and shows the heavy alpha stringers found in one location (Fig. 6a). About 30 in. away from this sample the rod is practically free of alpha stringers (Fig. 7b). This would account for the short life of some of the samples and also indicates the variation of composition which exists in the ingot. A comparison of the total corrosion (mg/cm^2) versus time for two epsilonized ingots and the extruded material made from these ingots can be seen in Fig. 8. It is becoming apparent that the extruded material is poorer than the cast and epsilonized alloy. This is probably due to alpha stringers in the extruded samples leading to cracking compared to a dispersed alpha in the cast alloy. Because some of the alpha particles appear to be fine and discrete, rather than continuous stringers, it is suspected that partial de-epsilonization may occur during the extrusion operation carried out at 870 to 900°C. It is intended therefore in the future to devote attention to the use of U_3Si in the cast condition.

F. Mechanism of Corrosion of U_3Si

The mechanism of corrosion of unclad U_3Si is being investigated to obtain a better understanding of the factors controlling the corrosion reaction. It has been observed that an unidentified phase forms at the interface between the oxide and the U_3Si . Examples of this phase are shown in Figs. 9a and 9b. It is apparent that this phase varies in thickness and continuity and is penetrated by the oxide at various points along the interface. However, because the majority of the U_3Si surface is covered by this phase, it is probable that the corrosion reaction is influenced by its presence.

As may be noted in Figs. 9a and 9b, the unknown phase is very similar in appearance to the U_3Si_2 particles (the white particles) dispersed throughout the matrix. This latter observation indicates that the corrosion reaction enriches the matrix in silicon and hence causes the formation of U_3Si_2 at the oxide interface. Corrosion tested samples are being examined by X-ray diffraction techniques for a positive identification of this phase.

When unclad samples of U_3Si extrusions are annealed at high temperatures after extrusion it was shown that the corrosion resistance is adversely affected. For example, the 35-day corrosion rate for bare samples of extrusion No. 462 is increased from 0.70 $\text{mg}/\text{cm}^2/\text{hr}$ to 1.18 $\text{mg}/\text{cm}^2/\text{hr}$ by an annealing treatment of 168 hr at 800°C. This anneal causes a coarsening of the grains and an apparent precipitation of some impurity phase at the grain boundaries as seen in Fig. 9c. An unclad sample from this extrusion was annealed at 800°C for 168 hr and then corrosion tested in 650°F water for 168 hr. Examination of the corrosion surface reveals, as seen in Fig. 9d, that the corrosion attack is accelerated at the grain boundaries with grain boundary penetration of the phase which was tentatively identified as U_3Si_2 . It would appear that this penetration is the result of higher diffusivity of silicon at the grain boundaries with the result that U_3Si_2 tends to nucleate along these boundaries. As seen in Fig. 9d the oxidation is consuming the phase at the grain boundaries more rapidly than it attacks the U_3Si matrix. This results in whole grains being separated from the surface and hence a higher rate of metal consumption. Furthermore, since this U_3Si_2 phase is attacked more rapidly than U_3Si , the formation of this phase during corrosion may accelerate corrosion.

G. Corrosion of Bare Ternary Alloys

Arc melted buttons, supplied by Armour Research Laboratory, of U-3.8 w/o Si plus 0.2, 0.5, or 1.0 w/o Mo, Nb, Cu, Ta, or Pt are still being corrosion tested in 650°F degassed water. The results obtained to date are summarized in Table VI. Included in Table VI are current results being obtained for duplex melted and epsilonized U-3.8 w/o Si ingot castings for comparison purposes. In WAPD-PMM-262, U-3.8 w/o Si with additions of 0.2 Mo, Nb, and Cu were compared to old U-3.8 w/o Si data to point out the beneficial effect of these additions. At that time not enough data were available on the U-3.8 w/o Si now being made at Bettis by the duplex melting process. It can be seen from Table VI that cast and epsilonized U-3.8 w/o Si alloy in which no alpha is detected has corrosion rates as good as the best ternaries being tested at the present time. The effect of the ternary addition as previously shown was to eliminate the presence of alpha which is very difficult to control in the binary alloy. More data have been accumulated on the U-3.8 w/o Si alloy with tantalum additions with promising results on the 0.2 Ta addition.

An attempt to pinpoint the exact composition for the most promising ternary alloys was made at Bettis. Arc melted buttons of U-3.8 w/o Si plus 0.1, 0.2, or 0.3 w/o Mo, Nb, or Cu are being corrosion tested in 650°F degassed water. The corrosion test results and metallographic examination indicated that the buttons were extremely segregated and of poor quality. It is planned to repeat the above analyses with induction cast rods. In addition, rods with 0.1, 0.2, and 0.3 w/o Al and Ta will also be cast for corrosion testing. Data for these alloys should be available for the next resume.

H. Fuel Elements Containing U₃Si Particles

The presence of elongated alpha stringers in extruded rods of U₃Si causes a reduction in the corrosion resistance of the fuel and adversely affects the resistance to dimensional changes during irradiation. It is possible that the detrimental effects of the alpha phase can be eliminated by the use of crushed U₃Si particles for the fuel material. Such an element can possibly be impregnated with a corrosion resistant aluminum alloy to afford both a bond to the Zircaloy-2 cladding and a corrosion resistant material between the particles.

An element of this type was made with Zircaloy-2 cladding and irregularly shaped U₃Si particles which varied in size between 0.015 and 0.050 inches. The compact with a density of 55% maximum U₃Si density, was impregnated in vacuo with an Al-6 w/o Cu alloy. The specimen was overheated during the Al alloy melting cycle with the result that the U₃Si particles transformed to the alpha and delta phase and the specimen could not, therefore, be examined for corrosion characteristics. It was found by this test that good bonding was obtained both between the Al alloy and the Zircaloy cladding and between the Al alloy and the U-3.8 w/o Si particles. However, shrinkage cavities formed throughout the Al alloy upon solidification. The thermal contraction caused the separation of the Al alloy grains in regions where the low melting eutectic (Al-23 w/o Cu) had formed intergranularly.

Particles of U_3Si are now being prepared for a study of the effects of particle size and size distribution, compact density, Al alloy composition and fabrication techniques upon the quality of the fuel element.

I. Mechanical Properties of Extruded U_3Si

A method proposed for the fabrication of U_3Si fuel elements is the drawing of Zircaloy-2 cladding and aluminum alloy barrier materials, both in the form of tubes, over an extruded or cast rod of U_3Si at moderate temperatures. The melting point of the aluminum alloy limits the working temperature to approximately $600^\circ C$. Tensile tests were conducted to determine if the core alloy has sufficient ductility to permit drawing at such relatively low temperatures.

An epsilonized ingot, No. 502, was extruded without cladding to 0.50 in. diameter. Threaded-end tensile specimens with a 0.25 in. diam reduced section were machined from the extrusion. Some of the specimens were annealed prior to final machining for 168 hr at $800^\circ C$ and furnace cooled. Only four reliable tests were obtained because some specimens contained defects resulting from extrusion cracks. The results are shown in Table VII.

As was expected, there is no measurable ductility for any of the room temperature tests. The ductility at $390^\circ C$ is about 4%. Unfortunately, the maximum ductility and the presence, if any, of a brittle-ductile transition cannot be ascertained from these limited tests. When more material is obtained, a complete series of tests covering the range up to $600^\circ C$ will be run.

The $800^\circ C$ heat treatment has caused a decrease of nearly 50% of the room temperature ultimate strength. Grain size is known to have an appreciable effect upon the fracture stress of brittle specimens with fine grained samples having higher fracture stress when all other variables remain constant. However, the variation in grain size between the two specimen conditions is too small in relation to the large change in fracture stress to account for the difference. Strain rate may also be eliminated as a possible cause since the as-extruded specimen which was tested at a rate of 0.05 in. per minute had a strength nearly the same as the specimen tested at ten times this rate.

Microexamination failed to reveal any structural change (other than grain size) due to heat treatment. However, it is known from corrosion studies of the epsilon phase that some form of intergranular precipitation apparently does occur during high temperature annealing. It is possible that such a grain boundary phenomenon is responsible for the observed effect upon fracture stress. If this is the case, it is expected that fracture may occur intergranularly in the heat treated samples. The nature of these fractures is being determined.

II. Gamma Phase U-Mo and U-Nb Alloys

As indicated in the previous resume no new experiments were initiated to establish the effects of fabrication and heat treatment variables on the corrosion behavior of U-Mo and U-Nb alloys. However, those tests which were under way were continued. Additional data obtained are described below.

A. Corrosion Life of Extruded and Cold Drawn Alloys

The 650°F corrosion tests of defected clad fuel elements are being continued to determine the feasibility of heat treating subsequent to extrusion. The alloys still in test include U-Nb and U-Mo alloys, with and without ternary Pt additions, and U-Nb with a Nb barrier. The corrosion life data obtained to date are reported in Table VIII.

The corrosion life of the as-extruded alloys continues to be good in all cases except the U-8 Nb alloy. All the elements of this alloy have now failed, a maximum life of 168 days being attained in the as-extruded condition. The extruded elements of the other alloys have survived 154-182 days of test with the U-10 Nb alloy showing the only failures in this condition. The excellent corrosion life and lack of variability of corrosion life displayed for U-Mo alloys in the as-extruded condition indicate that the variables in the fabrication process were well under control.

Of the extruded samples homogenized at 900°C for 24 hr, only the U-10 Nb alloy with a Nb barrier has a corrosion life (182 days) equal to the best as-extruded elements, and two of three cold drawn and homogenized samples are still in test after 154 days. In contrast to these results, all the extruded and homogenized samples of the other alloys have failed in less than 140 days. In the homogenized and aged (400°C for 5 weeks) samples, the U-12 Nb alloy samples have the longest life in both the extruded and cold drawn conditions (greater than 126 days).

B. Effect of Annealing on Corrosion Life of Cold Drawn U-12 Mo

The study being conducted to determine the feasibility of a recrystallization and stress-relief heat treatment for cold drawn U-12 Mo defected clad fuel elements was continued. The induction melted and pre-homogenized elements have now completed 126 days of test on the material cold drawn 53% R.A. and 42 days on material cold drawn 40% R.A. The data for these elements are tabulated in Table IX. Although the tests on the 40% cold drawn material have not had sufficient test time, the lesser amount of cold reduction has already reduced the considerable variability found in 53% cold drawn material. From the 53% cold drawn data there are indications that the optimum heat treatment is about 5 to 10 minutes at 700°C. Such a treatment is long enough to recrystallize the fuel yet short enough to confine the growth of the diffusion zone between clad and core.

The above tests were repeated using extrusions prepared from duplex melted and pre-homogenized ingots and the results are recorded in Table X for 84 days of test time. The data indicate that the variability

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has been decreased and no failures have been recorded for the as-extruded samples and the 53% cold drawn samples annealed for 5 and 10 minutes at 700°C. It is becoming apparent that a short time recrystallization heat treatment, which confines the amount of core-clad diffusion, is necessary to increase the life of cold drawn U-12 Mo clad extrusions.

C. Effect of Internal Stress on Corrosion Life

1. Defected Clad U-12 w/o Mo Stressed Elements

The investigation of effect of residual macroscopic stress on the corrosion life of U-12 Mo has been completed. Two extruded rod samples with Zircaloy-2 cladding were stressed in a bending jig to about 95,000 psi and two other samples from the same extrusion were used as unstressed control samples. All the samples were defected with 0.040 in. diam holes through the cladding. One of the stressed samples was tested with the defect on the tension side, and the other was tested with the defect on the compression side in 650°F degassed water. The samples were removed after 126 days of test. No failures occurred during the test; however, the two unstressed control samples and the stressed sample with the defect on the tension side started to bulge indicating failure would have occurred in the next 14 day test period.

The samples were sectioned through the defect and examined metallographically. Longitudinal sections can be seen in Fig. 10. It will be noted that the tension-defected specimen is cracked while the compression-defected specimen has corroded normally with no evidence of cracking. Some hydride was present in the heavily cracked areas of the tension-defected sample. The control samples were also cracked and exhibited hydride precipitation. No evidence of hydride was found in the compression sample. Thus, it appears that when compressive stresses are present in a clad defected element, the corrosion life is extended.

2. Bare U-12 w/o Mo Stressed Samples

Two bare samples were stressed in a bending jig to 18,000 psi and 26,000 psi, respectively, and tested in 650°F degassed water together with two unstressed control samples. All the samples were taken from the same extrusion. The samples were homogenized prior to testing at 900°C for 24 hr. After 28 days of test, the samples were removed for examination. At the time of removal from the bending jig, it was observed that the samples had a permanent set thus relieving the originally imposed stress.

Upon examination of the microstructure of the tension and compression sides of the samples, it was apparent that the state of stress had considerable influence on the density and form of the hydride needles present. This can be seen in Fig. 11 showing photomicrographs of longitudinal sections at opposite tension and compression edges. Note the heavy density and random direction of the hydride on the tension side compared with the relatively fine needles, nearly perpendicular to the edge, on the compression side. The center of the rod, i.e., the location of zero

stress, had a hydride configuration resembling the tension edge. The unstressed control samples also resembled the tension side and the zero stress center of the rod. Thus it becomes evident that the optimum stress configuration in both clad and bare samples is a state of compression.

3. Cathodically Hydrided Bare Samples of U-12 w/o Mo

Bare samples of U-12 w/o Mo were cathodically hydrided at 100°C in a bending fixture with maximum fiber stress of 80,000 psi. The microstructure at the tension and compression surfaces is illustrated in Fig. 12. It is apparent that the hydride platelets tend to orient themselves perpendicular to the tension surface and parallel to the compression surface. This confirms the observation made in an earlier resume. This was not the case for the samples tested in 650°F water and may indicate a pronounced effect of test temperature.

D. Effect of Defect Size on Corrosion Life

Twenty rod samples from a single U-12 w/o Mo coextrusion are being corrosion tested in 650°F water to determine the effect of varying defect size on corrosion life. Four samples each were prepared with defects of 0.003 in., 0.013 in., 0.020 in., 0.040 in., and 0.080 in. diam. The results obtained so far (listed in Table XI) indicate that all four elements with 0.080 in. diam defects failed in 70-126 days and two of four with 0.040 in. defects failed in 84-98 days. The remaining elements have survived 154 days and are continuing in test. It is apparent that corrosion life of clad elements is related to the area of exposed fuel.

E. Corrosion of Zircaloy-2 Clad U-2 w/o Zr Extruded Elements

Six simulated fuel elements fabricated by Nuclear Metals, Inc., (NMI) using Zircaloy-2 clad extrusions of U-2 w/o Zr alloy, were supplied for corrosion testing in 650°F water. These elements had been defected with 0.007 in. diam holes drilled just through the cladding. They were then heat treated 2 hr at 1050°C and furnace cooled to develop a corrosion resistant U-Zr interdiffusion layer below the defects and at the core-clad interface. The samples had been corrosion tested for 1225 hr in 600°F water by NMI before being sent to Bettis for further tests.

One sample was sectioned at the defect and revealed the condition shown in Fig. 13a. Oxide had apparently built up at the base of the defect preventing accelerated attack of the core alloy. The remaining five samples were tested in 650°F water for two 14-day test periods without adverse effects. It was suggested at the last Uranium Alloy Panel Meeting that the defects be enlarged before continuing the test. This was done as follows:

Samples

1 and 2	Defect enlarged to 0.040 in. diam just through the cladding (0.030 in. from surface, oxide layer undisturbed)

- 3 and 4 Defect enlarged to 0.040 in. diam through a fraction of the oxide (0.035 in. from surface)
- 5 Defect enlarged to 0.080 in. diam just through the cladding (0.030 in. from surface, oxide layer undisturbed).

After the above modifications in defect size the samples failed catastrophically in the next 14-day test period in 650°F water. Failures are shown in Fig. 13b. In view of this, it is reasonable to question the effectiveness of the diffusion heat treatment in inhibiting corrosion attack of the fuel. It was shown for U-12 w/o Mo that very long life can be expected when defects smaller than 0.040 in. diam are drilled through the cladding into the fuel. It is felt that oxide build-up in small size defects is responsible for this. The same may be true for the subject samples.

III. Basic Metal Alloy Development

A. Alloying Behavior of Uranium

1. Specific Heat Measurements on U-Base Alloys

To gain a better understanding of the alloying behavior of uranium, an experiment is in progress to measure specific heats and latent heats of transformation and order-disorder of uranium alloys as a function of temperature.

Data has been obtained on one 10.5 w/o and three 21.5 w/o Mo alloys:

	J-740-A	L-142-B	L-142B-I	L-142-C
Composition w/o Mo	10.5	21.5	21.5	21.5
Heat Treatment	Stepwise 560°C → 400°C in 181 days	500°C for 96 days	500°C for 96 days	550°C for 95 days
Heating Rate °C/min	3.5	3.5	1.9	1.9
Phases before Heating (by X-ray spectrometer)	$\alpha + \epsilon$ + *S.L.	Mo + ϵ + S.L.	Mo + ϵ + S.L.	Mo + ϵ + S.L.
Phases after Heating (by X-ray spectrometer)	γ	$\gamma + \text{Mo}$	$\gamma + \text{Mo}$	$\gamma + \text{Mo}$

* Superlattice

The specific heat versus temperature curves for both alloys indicate that the epsilon phase is actually an ordered form of gamma. The specific heat curve shows the destruction of the long range order below the critical temperature. Thermocouple calibration and sample emissivity corrections are being made to obtain accurate values of specific heats at various temperatures.

The position of the peritectoid reaction isotherm has been located but accurate temperature values will be indicated after thermocouple calibration. It is apparent that the peritectoid reaction occurs at least 10°C higher than the latest phase diagram indicates (600°C + 10°C). The temperature obtained is not very sensitive to heating rate for the relatively slow rates used. Changing the rate from 1.9°C/min to 3.5°C/min resulted in less than 2°C upward displacement of the peritectoid isotherm.

Specific heat measurements will also be obtained for U-Nb samples corresponding to AB, A₃B and AB₃ compositions to determine if an ordering reaction occurs in this system. Suitable samples are being prepared

for heat treatment.

2. Resistivity of U-Alloys at Low Temperatures

A liquid nitrogen cryostat has been built for determining electrical resistivities of U-base alloys in the temperature range -196°C to R.T. These data will help explain the low temperature resistivity anomalies observed for U-Mo, U-Nb and U-Zr alloys. Final assembly of the cryostat is awaiting delivery of thin-walled monel tubing. In the meantime suitable samples of the alloys are being prepared and heat treated.

B. Uranium-Base Alloys with Transition Phase Structures

The crystal structure has been determined for the alpha prime phase found in a 5 w/o Mo-U single crystal water quenched from 900°C . The structure was determined from X-ray diffraction patterns obtained with a Weissenberg camera. The structure is base-centered orthorhombic Cmm2, with $a = 7.0 \text{ \AA}$, $b = 6.84 \text{ \AA}$ and $c = 3.42 \text{ \AA}$. There are eight atoms in the unit cell with the atoms located at $0, Y, Z_1$; $0, \bar{Y}, Z_1$; $1/2, 1/2 + Y, Z_1$; $1/2, 1/2 - Y, Z_1$; $X, 0, Z_2$; $\bar{X}, 0, Z_2$; $1/2 + X, 1/2, Z_2$; and $1/2 - X, 1/2, Z_2$, where $X = 0.25 \pm 0.03$, $Y = 0.25 \pm 0.03$, $Z_1 = 0 \pm 0.03$ and $Z_2 = 0.50 \pm 0.03$. A more precise determination of the atom locations is being determined by calculating the residuals with various assumed values of the coordinates. This structure is actually a slight perturbation of the body-centered cubic gamma phase and may be visualized as consisting of four body-centered cubes lying in a plane arranged in a square whose sides are composed of two cube lengths. The slight perturbations of the atoms from the body-centered cubic positions yield the observed orthorhombic structure.

Work is now in progress to determine the habit plane of the transformation and the orientation relationships as a result of transformation. From this information an attempt will be made to determine the atom movements during transformation.

C. Thermal Stability of Low Zr-Nb Alloys of U

In order to determine the phase boundaries, thermal stability and rate of diffusion in the low Zr-Nb ternaries of uranium, diffusion couples of Zr with different U-Nb alloys are being prepared which will cover a range of ternary compositions. An attempt to prepare a couple by roll bonding a specimen in vacuum at elevated temperatures proved unsatisfactory because of the difficulty in obtaining a plane interface between the components of the couple. Excellent bonding has been achieved, however, using pressure bonding techniques. The components, with prepared surfaces, are clamped together at room temperature with Kovar bolts. These are then heated in vacuum for diffusion, taking advantage of the difference in thermal expansion between the components of the couple and Kovar bolts. A series of couples are now being prepared by this method.

D. Effect of Stress upon Decomposition of the Gamma Phase

The effect of stress upon the decomposition of the gamma phase

in low Mo uranium alloys is being investigated on specimens held at 400°C under dead weight loading. In the 7 w/o Mo-U alloy a marked acceleration of transformation occurred at stress levels above 40,000 psi. The effect was noted by comparing the threaded ends of creep specimens with their gauge lengths. Above 40,000 psi stress on the gauge length, the threaded ends of the specimen, which are subjected to a relatively small stress, exhibited a gamma phase structure whereas the gauge length had transformed to alpha plus gamma. Metallographic examination of a polished and etched specimen reveals the presence of what appear to be slip bands throughout the transformed region, indicating that transformation had occurred along the slip planes. Experiments are now being conducted using a 5 w/o Mo-U alloy in which transformation to the alpha prime phase can be induced by the application of stress.

E. Magnetic Susceptibility of U-Mo Alloys

Equipment has been assembled for measuring the susceptibility of metal samples as affected by molybdenum content and the presence of hydride platelets formed during corrosion in high temperature water. The Curie method is being used, bismuth being used for calibrating the apparatus. The susceptibility of alpha uranium was determined to be $1.730 \pm 0.008 \times 10^{-6}$ at 27°C. This compares favorably with the latest value of 1.74×10^{-6} given in the literature.^{1/}

These results are expressed in cgs units per gram. The susceptibility per gram atom (X_A) as a function of Mo concentration is shown in Fig. 14. To the first approximation the value of X_A is proportional to the density of states for the spin paramagnetism. The results indicate that the density of states increases as the electron atom ratio increases, reaches a maximum at about 15 a/o Mo, and then rapidly falls off.

Stoner^{2/} has discussed in some detail the relation of the bond form and the temperature dependence of susceptibility. The temperature coefficient of susceptibility for alpha uranium has been shown to be positive by Bates and Hughes. Applying Stoner's results we conclude that alpha uranium is at or near a region of minimum energy-level density. The positive temperature coefficient arises from the possibility of the transfer of electrons between two bonds, so that the number of holes in the lower bond and the number of electrons in the upper bond both increase. This effect may more than compensate for the normal temperature decrease of susceptibility for the two bonds considered separately. For higher Mo concentrations the temperature coefficient of susceptibility should change sign and become negative. It is believed that it is possible to associate the energy bond overlap effect with the anomaly in the temperature coefficient of electrical resistivity.

1. Bates, L.F. and Hughes, D., Proc. Phys. Soc. (London) B67, 28 (1954).
2. Stoner, E.C., Proc. Roy. Soc. 154, 656, (1936).

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F. Identification of the Hydride Phase in Corrosion Tested Gamma-Phase U-12 w/o Mo Alloys

Decomposition pressure measurements of U-12 w/o Mo alloys corrosion tested for various times in 650°F water have been continued. Recent data show that earlier results, obtained from a run upon a single specimen corrosion tested for 14 days at 650°F, were in error. The log of decomposition pressure vs the reciprocal of the absolute temperature curve for this specimen, which was previously shown to have a slope differing from specimens corrosion tested for 21 days, is now known to superimpose upon the 21 day curve as do specimens corrosion tested for seven days. A summary of the data accumulated to date is contained in Fig. 15 and listed below:

$$\text{Log Pmm} = \frac{-1235}{T} + 5.315$$

This equation is valid for the U-12 w/o Mo hydride in a γ matrix and containing 270, 980 and 1350 ppm H₂. The heat of formation of the hydride is equal to 5650 cal/mol.

The equation for the system $2\text{UH}_3 \rightleftharpoons 2\alpha\text{U} + 3\text{H}_2$ is

$$\text{Log Pmm} = \frac{-4480}{T} + 9.20 \text{ (from literature)}$$

The experimentally determined heat of formation for this hydride is 20,800 cal/mol. In view of these results there is no doubt that the corrosion hydride in γ -phase U-Mo and pure UH₃ are fundamentally different.

It was previously reported that the heat of formation was dependent upon the amount of hydride formed during corrosion. Based on additional data this is now found to be in error. The realized behavior is to be expected in view of the fact that the decomposition pressure is independent of composition for a two-phase system.

The apparatus has been modified to accommodate a wider range of pressure, temperature and composition. As soon as sufficient pressure temperature, and composition data are obtained, the results will be analyzed both from the standpoint of kinetics and thermodynamics. Absorption and metallographic experiments will be conducted in order to determine the H₂ solubility limit and the limiting composition of the hydride as a function of temperature. The apparatus is now capable of admitting and removing measured amounts of hydrogen.

G. U-Nb Phase Diagram Investigation

It was reported in WAPD-PMM-262 that both U-Mo and U-Nb alloys thermally transformed at low temperatures are retransformed upon irradiation to the high temperature gamma phase which is corrosion resistance. In the case of U-Mo alloys this irradiation induced transformation may be explained on the basis of disordering of epsilon and simultaneous solution of alpha to form the gamma phase. In the U-Nb system, however, no ordered phase has been reported. The possibility of the existence of an ordered phase, which may be very sensitive to method of sample preparation, was investigated.

A sample of U-10 w/o Nb, in the gamma condition, was transformed at 550°C for 2 weeks to form alpha and γ_2 (approx. 90 w/o Nb) according to the existing phase diagram. The sample was partially dissolved in HCl to remove the alpha phase and subjected to X-ray examination. The powder pattern showed the presence of a BCC phase with $a_0 = 3.414$ Å and a FCC phase with $a_0 = 4.693$ Å. The BCC phase is presumed to be untransformed γ_1 . The lattice parameter corresponds to a Nb concentration of 20 w/o. The reason for the shift from the original 10 w/o Nb is unknown. The FCC phase observed does not appear in the equilibrium diagram. Vigorous cold working of the residue with a mortar and pestle fails to eliminate the FCC phase in powder patterns. Chemical analysis by X-ray fluorescent techniques indicates roughly 90 w/o Nb and 10 w/o U. An untransformed (gamma quenched) sample of U-10 w/o Nb, also treated in HCl, did not show the FCC phase when X-rayed.

Another sample of U-10 w/o Nb, was specifically prepared for X-ray examination before gamma quenching and transforming for 2 weeks at 550°C. With no further surface treatment the presence of alpha, UO_2 , UO, gamma and the FCC phase was observed. When the specimen surface was polished metallographically the FCC phase disappeared. Upon re-etching to remove mechanically disturbed metal the phase reappeared. Partial solution of the sample in HCl again showed the FCC phase.

It was not possible to evaluate changes in lattice parameter of the FCC phase in samples of U-10 w/o Nb by varying transformation time below 550°C because of line broadening attributed to small particle size. To better determine the composition of this phase, however, samples corresponding to U_3Nb , U_2Nb , UNb, UNb_2 and UNb_3 are being prepared. These will be heat treated to determine if the FCC phase is in an ordered or disordered state.

Since the residue of the first sample treated in HCl analyzed approximately 90 w/o Nb, samples of this composition were gamma quenched and transformed for various times at 500°C. Microexamination, Fig. 16 indicates no grain boundary precipitation in the gamma quenched condition, but a definite appearance of a grain boundary phase on annealing at 500°C for 1 week. Longer anneals shows increased thickening of grain boundaries. X-ray examination has not revealed this phase after 3 weeks at 500°C. Samples are being treated for longer periods in an effort to identify this phase by X-ray diffraction.

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IV. Irradiation Effects - Uranium-Base Alloys

A. Irradiation of Transformed Specimens of U-Mo Alloys

It was reported in Resume IX (WAPD-PMM-262, p. 22) that the electrical resistivity, hardness and density measurements on irradiated specimens of alpha-transformed U-9, 10.5, 12 and 13.5 w/o Mo indicated the alpha plus epsilon phase had reverted to gamma phase as a result of neutron bombardment. Examination of the microstructures of these irradiated specimens at relatively low magnifications (250X) revealed essentially no changes from their pre-irradiation structures. Since the nature of the radiation-induced transformation is to disorder the epsilon and to take alpha uranium into solution, it is postulated that the second phase observed in irradiated samples may be impurities precipitated at the grain boundaries and sub-grain boundaries during thermal transformation. These may not be affected by irradiation and the microstructure would then remain unchanged. Another possibility is that this phase may actually be extremely small grained gamma which may look like alpha at low magnifications. In the U-9 and 10.5 w/o Mo, alloy samples, which had the maximum amount of transformation products, the alpha and epsilon phases were finely dispersed in a lamellar structure.

To check if the same resistivity values observed for thermally transformed and irradiated samples could also be obtained on samples having an alpha plus gamma structure the following experiment was performed. A sample of U-9 w/o Mo, which had been gamma quenched and thermally transformed to produce alpha plus epsilon (as was the case for the irradiated samples) was heated at 580°C for periods up to 11 hours. Since this temperature is in the alpha plus gamma field, the ordered epsilon phase would be expected to disorder and only a small change in the amount of alpha present would result. The reaction was followed by electrical resistivity measurements, X-ray and microstructure. The results showed that resistivity at first increased rapidly and then leveled off, reaching an equilibrium value which was only half that observed after neutron bombardment of a similar sample. The increase was from 31.5 to 52.5 microhm-centimeters at -200°C as compared to the change from 31.5 to 71 microhm-centimeters for the irradiated sample. X-ray indicated the presence of alpha and gamma and no epsilon. The microstructure before and after the 580°C treatment appeared unchanged. The same sample was then held at 650°C (in the gamma phase field) for approximately 16 hours. After this heat treatment the resistivity reached that of gamma quenched material (72.5 microhm-centimeters) and the microstructure showed a normal gamma phase with no second-phase present. These results indicate that the high resistivity values observed on transformed and irradiated samples cannot be explained by disordering of epsilon phase alone. Alpha must be redissolved during irradiation to attain the observed resistivities.

The design of an in-pile experiment in which the kinetics of the above reaction will be followed by means of electrical resistivity measurements is continuing. It is planned to use a foil specimen, 0.020 in. x 0.125 in. x 2.00 in. onto which a thermocouple and other necessary electrical leads have been spot welded. The specimen will be sealed into a Nonax glass tube in a

helium atmosphere and the electrical connections sealed by means of a tungsten-to-glass press seal. This glass tube will then be sealed into an aluminum sleeve so that if the tube breaks no radioactive contamination will enter the Brookhaven reactor coolant air streams. This one capsule design permits versatility in obtaining the desired specimen temperature in-pile since it will fit into the liquid nitrogen facility, and can also be inserted into a regular test hole with or without a heating unit.

B. Pre-Post Irradiation Tests on Clad U-Mo Alloys

Because of the lack of test facilities to handle the increased load of corrosion tests of irradiated materials, it has become necessary to reevaluate the importance of the irradiation programs which require corrosion testing. Since the decision was made last spring that PWR Core #1 was to be composed of UO_2 elements, the PWR metal core development has tended toward core alloys having better nuclear properties than uranium-molybdenum. Consequently, the corrosion testing of irradiated clad uranium-molybdenum alloys is now restricted to the completion of programs currently in progress. At present, a number of control specimens from various programs are continuing in corrosion test. These results will be reported in detail upon completion of the testing of specimens from any one experiment.

C. In-Pile Tests of U-Mo Alloys

1. Irradiation of Clad U-Mo Alloys in X-Rod Loop

Of twelve samples of U-10.5 w/o Mo and two samples of U-12 w/o Mo, fabricated from the same extrusions as used for the CR-WAPD-Met 4A specimens and irradiated in MTR process water to a maximum exposure of 8830 MWD/T, eight samples of U-10.5 w/o Mo remain in test in 650°F water after 35 days test. All samples from the loop test failed in 21 days or less. No reason for the early corrosion failure of the loop specimens as compared to the better life indicated for the specimens exposed to MTR process water is apparent.

D. Tests of Other Alloys

1. Clad-U-Nb Alloys

Ten samples of Zircaloy-2 clad natural U-10 w/o Nb, fabricated in various manners as shown in WAPD-PMM-262 and irradiated in MTR process water to a maximum exposure of 2600 MWD/T, have been in corrosion test in 650° F water for 21 days without failure.

2. Unclad U-Nb Alloys

Metallographic samples have been cut from the seven unclad uranium-10 w/o Nb samples, which were irradiated in the MTR to an estimated maximum exposure of 1820 MWD/T. The physical properties, as shown in WAPD-PMM-262, apparently tended toward equilibrium values intermediate between the gamma condition and the fully transformed conditions. Metallographic

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examination of specimen F-3 (γ Q + irradiated) and specimen F-22 (partially transformed + irradiated) indicated no obvious changes in microstructure from the pre-irradiation structure.

Corrosion specimens have been cut from several of these samples. It is intended that the corrosion rate in 650°F water will be obtained for comparison with rates obtained for unirradiated material.

3. U-Si Alloys

The three extrusions of Zircaloy-2 clad U-3.8 w/o Si which were fabricated by Nuclear Metals, Inc. were found to contain alpha uranium stringers. Because of the dimensional instability displayed by alpha uranium under irradiation, it was decided to use these only if they could be heat treated to eliminate alpha. When this was attempted de-epsilon ionization and loss of corrosion resistance resulted. This was discussed in Section I, D. Therefore, this material will not be used for irradiation tests.

Bare samples, 3/16 in. diam by 2 in. long have been machined from a pre-epsilon ionized and extruded ingot fabricated at the Bettis Site. The corrosion rate of as-extruded samples tested in 650°F water is 1.1 to 1.2 mg/cm²/hr. This rate is considered normal for the alloy. The microstructure of this extrusion showed small quantities of excess alpha and delta with little or no alpha stringers. It is planned to irradiate samples from this extrusion in NaK filled capsules for determination of dimensional, physical property and corrosion resistant changes.

D. Volume Expansion Study

In recent weeks, a number of clad fuel elements, showing cladding failures, have been returned to Bettis Plant after high exposures in-pile. Of fourteen Zircaloy-1 clad uranium and U-2 w/o Zr alloy prototype fuel rods, which were irradiated in the Chalk River NRX reactor to a maximum burn-up of 10,500 MWD/T, the three samples which were exposed to greater than 9000 MWD/T exhibited transverse and longitudinal splitting of the cladding, as described in WAPD-PMM-262 as experiment CR-WAPD-Met 2A. The maximum measured volume change was 2.8%. Of seventeen Zircaloy-2 clad uranium-molybdenum samples, which were irradiated in MTR process water to a maximum calculated exposure of 28,000 MWD/T, one sample of U-10.5 w/o Mo exhibited longitudinal cracking in the clad. The measured volume change was 4.3%. All of these specimens were irradiated in low temperature water; therefore, it is doubtful that corrosion of the clad is a significant factor in the cladding failures. These failures are believed due to the volume expansion of the core alloys during irradiation. This volume expansion is not to be construed as anisotropic growth, which we know experimentally can be overcome by suitable alloying, heat-treatment, and/or cladding. It is presumed that the introduction of fission fragment atoms into the metal crystal lattice causes an expansion of the crystal lattice and a consequent volume expansion.

If it is assumed that the volume will change as an additive function of the atomic volumes of the atoms present and that two fission fragment atoms are produced per fission, the following relationship will indicate volume changes to be expected in bare materials not subjected to

external stresses.

$$\text{Vol. change/l a/o burnup} = \% \Delta V / 10,000 \text{ MWD/T} = \frac{2 \bar{V} - V_u}{V_u \text{ alloy}}$$

where $\bar{V}$ = mean atomic volume of a fission fragment atom = $25.6 \text{ \AA}^3$

V_u = atomic volume of a uranium atom

$V_u \text{ alloy}$ = mean atomic volume of uranium alloy

Using this relationship, the following percent change in volume per 1000 MWD/T are obtained.

100 a/o U	0.26
25 a/o U-75 a/o Zr	0.23
5 a/o U-95 a/o Zr	0.22
U-9 w/o Mo	0.27
U-10.5 w/o Mo	0.27
U-12.0 w/o Mo	0.27
U-13.5 w/o Mo	0.28
U-10 w/o Nb	0.26
U-3.78 w/o Si	0.29

These calculated values are lower by a factor of 2-3 than the experimentally determined values. Kittel of Argonne has found a value of 0.3-0.5 for the volume expansion of natural uranium. Data obtained at Bettis indicate values of 0.4-0.7 for various uranium-molybdenum alloys.

Approximate curves of volume expansion vs exposure for bare uranium alloys and clad alloys are shown in Figure 17. It is to be noted that apparently the rate of growth is dependent upon the restraining forces imposed. This restraining force can be increased by increasing the cladding thickness; however, the amount of deformation allowable in the cladding prior to rupture cannot be changed to any great extent by increasing the cladding thickness. In the range of cladding thicknesses we are interested in, the amount of volume expansion allowable before clad failure is dependent upon the ductility of the clad material. It is therefore suggested that a ductile cladding, such as aluminum, will allow longer fuel element life before clad failure. A second suggestion is that fuel elements having a hollow center would be beneficial, since the volume expansion could be forced to fill the hollow center before deforming the clad. Life of such an element can be represented in Figure 17 by a horizontal line along the abscissa until the center would be filled and then an upward slope while cladding deformed. Experiments are currently being designed to check the feasibility of both of these suggestions.

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V. Properties and Preparation of UO₂

A. Powder Preparation

Work was continued on the preparation of UO₂ by various methods for irradiation and fabrication tests as well as for investigation of the physical and chemical properties of the oxide itself.

1. Hydrogen Reduced UO₃

This method was used to produce uniformly enriched UO₂ and oxide containing ZrO₂ and SiO₂ additives for irradiation tests. In preparing the powders with additives, known amounts of zirconyl nitrate and ethyl orthosilicate were dissolved in the uranyl nitrate solution so that on pyrolysis and hydrogen reduction, the desired amount of additives would be uniformly dispersed throughout the resulting UO₂. Powders containing appreciable amounts of SiO₂ (up to 5%) were inclined to be pyrophoric. Metallographic examination of argon sintered specimens containing 500-1000 ppm silica indicates that SiO₂ tends to inhibit the grain growth of UO₂. This effect will be studied in more detail.

2. Activated UO₂

Work on sulfated UO₂ has continued in an effort to determine the reasons for its superior sinterability. A quantity of UO₃ in which the sulfate ions were added to the uranyl nitrate solution was received from the Mallinckrodt Chemical Works. After reduction with hydrogen at 790°C this material was found to contain 75 ppm sulfate and 1% U(VI). The sinterability of this powder will be determined during the next period. Sulfate additions (75-260 ppm) were found to have no effect on the X-ray pattern of UO₂.

Since the particle size of these active powders is thought to be smaller than that of the normal material, Mallinckrodt UO₂ was treated with uranyl sulfate (1% sulfate) in order to determine the contribution of the sulfate ion itself to the superior sinterability. After hydrogen reduction of the U(VI) at 800°C, the UO₂ was found to contain 54 ppm sulfate. This material will be compacted and sintered in the same manner as the other activated powders.

3. Precipitation Studies

A series of ammonium diuranate powders was pyrolyzed at 250, 500 and 800°C and then reduced with hydrogen at 800-900°C. In most cases the final products were pyrophoric and had to be stabilized by using longer reduction periods and/or higher reduction temperatures. X-ray patterns of these powders are being run. A study of their pressing and sintering characteristics is in progress.

4. X-Ray Studies

An investigation was made in order to determine whether the uranium oxides and their parent compounds could be characterized by their X-ray diffraction patterns as to their method of preparation. Samples of ammonium

diuranate, U_3O_8 , UO_3 , and UO_2 prepared by various methods were examined. Work to date indicates that most of the compounds studied had normal powder patterns. The powder lines of UO_2 made by high pressure steam oxidation of uranium were broader than those of oxide prepared by other methods, indicating that this process produces a UO_2 with a very small crystallite size. However, U_3O_8 made by oxidation of this UO_2 yielded the sharpest X-ray pattern, indicating that some sintering occurs during oxidation.

5. Iron, Carbon, and Silica Doped UO_2

Because of the distillation of silica from the sintering furnace and the consequent possibility of the interaction of SiO_2 with the UO_2 pellets, silica was added in amounts of 250, 400 and 800 ppm to determine the sensitivity of sintered UO_2 to this impurity. Pellets were sintered in atmospheres of argon and hydrogen at $1450^\circ C$ for two hours. Corrosion testing was carried out in de-gassed water at $650^\circ F$ and 2600 psi. No visible changes were observed after an exposure of 15 days.

Since the UO_2 is prepared in stainless steel apparatus, and iron has been found in certain samples of UO_2 , oxides containing 100, 250, 500, 1000, 2500 and 5000 ppm iron have been prepared by dissolving ferric nitrate in the uranyl nitrate solution. These powders will be compacted, sintered and corrosion tested.

It has been observed that pellets which were sintered at $1750^\circ C$ for 10 hours in a graphite resistor furnace with an argon atmosphere repeatedly failed during corrosion testing while pellets sintered in hydrogen did not. Accordingly, it was postulated that formation of uranium carbide to the grain boundaries caused the corrosion failure of the argon sintered pellets and that carbon is removed during hydrogen sintering.

To test this hypothesis, pellets containing carbon in amounts up to 650 ppm were prepared sintering the doped powders in argon at $1450^\circ C$ for two hours and in hydrogen at $1450^\circ C$ for 2 hours. As expected, hydrogen sintering was found to reduce the carbon content to approximately 20 ppm while the pellets sintered in argon retained their carbon. However, contrary to expectations, the specimens containing carbon did not desinter even after a 15 day exposure. It is felt that this behavior may be due to insufficient time and temperature in sintering. Accordingly, this program will be continued by sintering the carbon doped powders at temperatures up to $1550^\circ C$ for 10 hours in an attempt to form uranium carbide as a second phase. Pellets prepared in this manner will be tested for corrosion resistance.

B. Fabrication and Sintering of UO_2

In connection with the study of the relative importance of the various factors affecting the sinterability of UO_2 a number of compacts having various as-pressed densities were sintered at $1750^\circ C$ in hydrogen for times ranging from 2 to 64 hours.

To date, the closed porosity determinations have been completed. These indicate that (1) the closed porosities in as-pressed compacts (densities ranging between 51 to 70% theoretical) made from as-received MCW powder as well as in 70% theoretical green compacts made from MCW + 325 and -325 sieve size

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fractions are in the range of 6.5 to 7.5%; (2) during the first 2 hours of sintering the closed porosity decreases 2 1/2 to 4% depending on the history of the compact; (3) between 2 and 16 hours there is first a slight increase and then a decrease in the closed porosity (1-3%); (4) between 16 and 64 hours the closed porosities do not change more than 0.5% and are essentially the same as they were at the end of 2 hours; (5) there seems to be no relation between terminal closed porosity and sieve size of the original powder, but there does seem to be correlation between the as present density and the terminal closed porosity. Compacts having green densities of 51%, 60% and 70% theoretical had final closed porosities of 2.2%, 3.0% and 4.7% respectively. The ratios of open-to-closed porosities are currently being measured.

C. Stability of UO₂ in Water

Long term tests on the stability of UO₂ compacts in water have continued. Specimens have now been exposed for 52 days in degassed water at 650°F, 45 days in degassed water maintained at pH of 10.5 at 650°F, and 58 days in degassed steam at 750°F. In general, the appearance and weights of the compacts have not changed significantly.

Several hydrogen sintered compacts, to which PVA and Sterotex had been added prior to pressing, were exposed to each of the above conditions for 10 days. The compacts appeared unaffected by the exposures. Chemical and spectrographic analyses of duplicate samples indicate that the addition of Sterotex and PVA does not increase the level of impurities in the hydrogen sintered compacts. The following concentrations of impurities were found: 0.1% U(VI), 0.002% carbon, 0.0002% silicon, 0.005% water, and 0.002% fluoride.

The test in degassed water containing 25 cc H₂kg has been discontinued since no differences have appeared between compacts tested under this condition and compacts tested in degassed water.

D. Surface Area and Density of UO₂ Powders

Surface area and density measurements have been performed on 3 ammonium diuranate (ADU) powders in an effort to understand the effect of preparative method on the nature of the resultant UO₂ powder.

The ADU powders were prepared from uranyl nitrate by precipitation with either ammonium hydroxide (ADU-13), urea (ADU-14) or ammonium carbonate (ADU-17). The surface area and density values obtained for the 3 powders are 72.4 m²/cc and 5.06 g/cc for ADU-13, 28.2 m²/cc and 5.15 g/cc for ADU-14, and 31.0 m²/cc and 5.66 g/cc for ADU-17.

The ADU prepared by precipitation with ammonia has the largest specific area, the lowest density and, on the basis of X-ray measurement, the smallest crystallite size. This is consistent with the fact that precipitation with ammonia occurs rapidly, very likely forming a finely divided, poorly crystallized powder. Precipitation by the other two methods occurs slowly, yielding a material of larger crystallite and particle size and smaller specific surface.

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Although both the ADU-14 and ADU-17 powders were prepared by methods involving slow precipitation, microscopic examination shows the ADU-17 to be composed of uniform spherical particles whereas the ADU-14 particles are somewhat distributed in size and shape. This probably indicates that the ADU-17 is a more carefully crystallized powder than the ADU-14 and contains fewer closed pores and amorphous regions—a conclusion which is borne out by the greater density of the ADU-17. The similarity in surface area may indicate that the average particle size of the ADU-17 is smaller than that of the ADU-14.

The above results will be compared with data obtained on the 3 derivative UO_2 powders.

E. $\text{UO}_2\text{-O}_2$ Equilibria

A study of the equilibrium and kinetics in the system $\text{UO}_2\text{-O}_2$ has been initiated in an effort to gain more information regarding the structure and stability of UO_2 under various conditions. In this connection, the equilibrium phase diagram for the region $\text{UO}_2\text{-U}_3\text{O}_8$ is being checked in order to determine the accuracy of the different phase diagrams prepared by Perio and Gronvald. Samples of oxides having O/U ratios of approximately 2.10, 2.20, 2.25, 2.35, 2.45, 2.51, 2.58 and 2.67 have been prepared by controlled oxidation of Mallinckrodt UO_2 ($\text{UO}_2\cdot 0.3$) in air using a thermal balance. The samples were placed in Pyrex or Vycor bulbs and were sealed off in vacuo. The samples in Vycor bulbs were subsequently annealed at 700°C for 7 or 14 days; the other samples were annealed at 360°C for 10 or 20 days.

The O/U ratios of the annealed samples have been determined and the samples have been sent for X-ray diffraction analysis to determine their crystal structures.

A number of samples of various O/U ratios are also being prepared by controlled oxidation in air at 180°C . One group of these samples will be sent directly for crystal structure determinations. Duplicate sets will be annealed in evacuated sealed bulbs at 180°C for periods of 9 and 16 days prior to submitting them for X-ray analysis.

By the above experimental procedure it is hoped to obtain information on the nature of the immediate products of the slow oxidation of UO_2 and the phase changes that occur on annealing.

F. Kinetic Studies in Water

Work has continued on the rates of oxidation and reduction of UO_2 and U_3O_8 in 650°F water containing oxygen and hydrogen in order to gain a better understanding of the reactivity of UO_2 in water under various conditions.

As was reported previously, UO_2 and U_3O_8 powders exposed for 45 days in degassed water retained their initial compositions. Samples exposed for 35 days in degassed water maintained at a pH of 10.5 also appeared unaffected. Further experimental work on these samples has been discontinued since UO_2 and U_3O_8 appear to be stable under these conditions.

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Samples of U_3O_8 have been removed from water containing 100 cc H_2/kg after exposure periods of from 5 to 55 days. The samples removed after the longest exposure had O/U ratios of 2.27. This test was discontinued temporarily when a leak developed which permitted oxygen to enter the system. The test will be continued with a new group of samples.

Results on samples exposed to water containing either 2 cc O_2/kg or 5 cc H_2/kg will be reported in the next resume.

G. In-Pile Fission Gas Release Experiment

Construction of apparatus for the determination of the rate of release of fission gases from a specimen of UO_2 under irradiation, described in WAPD-PMM-262, is approaching completion. The apparatus will be installed at Brookhaven National Laboratory and experimental work begun about November 18.

H. Magnetic Susceptibility Studies

The magnetic susceptibilities of a series of uranium oxide compounds having oxygen to uranium ratios between 2.00 and 2.67 have been measured, and the theoretical significance of the results discussed by Goldman and Arrot of the Carnegie Institute of Technology. Conclusions of their report are as follows:

1. The phase boundary of the lower oxygen phase (CaF_2 structure) is at an oxygen-to-uranium ratio of 2.31 ± 0.01 .
2. The phase boundary of the higher oxygen phase (orthorhombic) is at an oxygen-to-uranium ratio of about 2.44. This figure is not as reliable as the foregoing.
3. Dawson and Lister (J. Chem. Soc., 1950, 2177; ibid., 2181) appear to have an error in their O/U analysis for $2.0 < O/U < 2.3$. This means that the X-ray data of Anderson and Albeman (J. Chem. Soc. London, 1949, S 303) is also in error as to composition since the same sample was used in both measurements.
4. When oxygen is added to UO_2 , the anion lattice is undisturbed and oxygen enters in interstitial positions, increasing the exchange interaction between U(IV) ions.
5. With increasing oxygen content it is most likely that U(IV) ions are replaced by U(VI) ions.
6. The difference between U_3O_8 and oxide of slightly lower O/U compositions in the same phase is brought about by creation of oxygen lattice vacancies.
7. The upper limit on the presence of UH_3 in the samples studied is on the order of one part in 10^5 .
8. $UO_{2.03}$ has an antiferromagnetic transition at $28^\circ K$. This transition is not evident in oxides of greater oxygen concentration; in the latter, anomalies exist in the region 2 to $28^\circ K$.

I. Diffusion Studies

Construction of equipment and the development of experimental techniques for the determination of the self diffusion of uranium and oxygen in UO_2 has begun.

Two types of diffusion couples are being investigated — a compound pellet, consisting of a layer of enriched UO_2 pressed onto a layer of natural UO_2 , and a sintered natural UO_2 pellet upon which a thin layer of enriched UO_2 has been vacuum deposited.

An evaluation of the two types of diffusion couples is being made.

Various methods for obtaining and analyzing very thin layers of UO_2 from the couples are also being investigated.

VI. UO₂ Irradiation Program

Hanford has tentatively approved the pre-post irradiation program planned with the aid of Naval and Westinghouse statisticians, Miss B. Day and Mr. W. Horton which, by the application of statistical methods, will yield a maximum amount of information with a minimum number of samples. In order to achieve maximum uniformity, the samples will be irradiated in the Hanford piles in NaK capsules. The only design change requested by Hanford involved the addition of an aluminum sleeve on the NaK capsule to maintain proper water flow and provide added protection against galvanic corrosion.

As currently planned, the program involves the irradiation of four groups of 32 samples for periods up to 16 months. The Hanford Special Irradiations Request is being prepared and will be submitted for final approval in the next four weeks.

1. MTR-WAPD 14 Met 1 Irradiation of PWR Reference Rods in NaK Capsules

In order to ascertain the behavior of UO₂ specimens when irradiated under maximum anticipated PWR blanket conditions, four 4½" long specimens made according to the PWR reference specifications (except that Sterotex was not added) were canned in NaK capsules and irradiated in position L-45 of the MTR for three weeks. This location is the highest flux position in the MTR, producing a thermal neutron flux of approximately 4×10^{14} nv. The maximum calculated temperatures produced in the specimens at this flux are as follows:

Central temperature of UO ₂	2660°C
Interface temperature	660°C
Temperature at surface of cladding	540°C
Heat Flux	762,000 Btu/hr-ft ²

The samples have been returned to the Bettis Plant, and the NaK capsules opened. Gross changes were observed in the appearance of the two high flux samples. The cladding bulged and cracked, and in one of the samples the Zircaloy-2 cladding apparently melted and allowed the UO₂ to escape. The two lower flux samples showed no evident change in appearance as a result of the irradiation.

The fission monitors are currently being analyzed, and the results should permit a more accurate calculation of the average temperatures existing in the samples during the irradiation. After photographing and measuring the dimensions of the samples, the samples will be sectioned and both the UO₂ and Zircaloy cladding examined metallographically.

2. MTR-WAPD-19-Met-4 A-Piece Cyclor

The eight natural Mallinckrodt samples with various combinations of diametral fit and end cap clearance which were irradiated in an A-Piece cyclor at MTR have been returned to WAPD and the capsules opened. These specimens were cycled into and out of a thermal neutron flux of 1×10^{14} neutron/cm² approximately 10,000 times over a period of 12 weeks. They received an estimated exposure of 1250 MWD/T. Preliminary measurements indicate no appreciable warpage.

3. MTR-WAPD-19-Met-5 A-Piece Cyclor

The four full length 5.5 w/o enriched UO_2 specimens being irradiated in the second A-Piece cyclor at an unsuppressed flux of 3.5×10^{13} neutrons/cm² have been rescheduled for three more cycles. They will be discharged December 5, 1955 instead of October 3. This should give an exposure based on revised estimates of the MTR suppressed fluxes of approximately 5,000 MWD/T. The total number of cycles into and out of the maximum thermal neutron flux will be 17,500. These samples are similar to MTR-WAPD-19-Met-4.

4. MTR-WAPD-19-Met-6

Eight natural samples similar to those reported in MTR-WAPD-19-Met-4 but compacted without PVA are being irradiated in the first A-Piece cyclor. These will be discharged on December 5, 1955 also after an exposure of 1650 MWD/T and 10,000 cycles into and out of the high thermal neutron flux.

5. CR-WAPD-Met-7 Third Chalk River UO_2 Defect Test

This test, containing one 95% dense defected UO_2 sample approximately 3.5" long has operated for an estimated total of 5000 MWD/T exposure in the X-1 loop facility over a period of approximately five months.

The activity levels in the water have remained relatively low for the entire test with no indications of gross specimen failure. The gross beta-gamma counts have averaged 1×10^6 disintegrations/minute/ml. The test will be discharged November 7, 1955 and will be replaced by CR-WAPD-Met-9.

6. CR-WAPD-Met-8 Fourth Chalk River UO_2 Defect Test

A full length UO_2 specimen (93% of theoretical density) has been operating in the X-3 loop at Chalk River since the middle of September, 1955, along with other defect U-Zr specimens. The specimen was made of 4.7% enriched UO_2 compacted without PVA and sintered in H_2 at 1750° for 10 hours. The defect is a .005" diam hole 1 1/2" from the top end. The specimen was assembled with 0.0015" diametral clearance and press fit end caps similar to the specimen in CR-WAPD-Met-7.

Operation began with a level of activity comparable to X-1 and it rose gradually to a level approximately three times that of X-1, possibly because of the larger specimen size assuming no contribution from the U-Zr samples. On October 28, 1955 there was a pile shutdown and upon start up the gamma monitor on the X-3 loop went off scale then settled back down to approximately twice the previous level. This was repeated on several succeeding shutdowns in the next two days. Indications are that the UO_2 specimen is splitting similar to the first UO_2 defect test in the X-1. Although there is a possibility of failure of the U-Zr specimens, the bursts of activity on startup followed by a return to a lower level of activity would seem to have been more typical of a progressively worse defect in a clad UO_2 specimen.

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Uranium analyses are now being performed on the water in an attempt to determine whether fission products are removed from UO_2 by the leaching process, or if they are associated in part with the presence of UO_2 in solution or suspension in the water.

10. WAPD-MTR-14-Met-6

One MCW specimen was punctured and the release of fission gas up to 250°C was measured. A release of about $7 \times 10^{-1}\%$ of Kr^{85} at room temperature and about $2 \times 10^{-3}\%$ at 250°C was found. The results are based on calculations of the total amount of Kr^{85} in the specimen using an estimated burnup of 3000 MWD/T. The specimens were then removed to be sectioned for metallographic studies. The specimens will then be returned to the fission gas apparatus for measurement of the release of gas at temperatures up to 1000°C . Total fission gas analyses will also be made.

11. WAPD-MTR-19-Met-1

A measurement of the release of fission gases from an 81% dense MCW sample at temperatures up to 1000°C has been completed, but the fraction of gas liberated cannot be reported until the results of the total gas analysis are available.

12. Out-of-pile Thermal Cycling

Five UO_2 filled Zircaloy-2 tubes and one empty clad altered in the following respects after being thermally cycled tubes in NaK between 550 and 1050°F :

- a. Lengths have increased from 0.1 to 1.0% with the empty clad having changed the greatest amount.
- b. Diameters have changed from -0.03 to +0.9%. Again the empty clad suffered the greatest change.
- c. Center runout of the rods has changed from 0.002 to 0.044" with the average of the six rods equal to 0.011".
- d. X-rays of the rods show one broken end pellet in four of the rods as well as a few small particles in the spaces between many of the pellets.
- e. In general, the rate of change in these measurements appears to be decreasing. No evidence of ratcheting due to the differential coefficient of expansion of Zircaloy-2 and UO_2 had been found. The rods have been returned to test for an additional 2000 cycles.

7. CR-WAPD-Met-9 Fifth Chalk River UO₂ Defect Test

Six PWR reference size rods have been prepared for the fifth UO₂ defect test in the X-rod loops at Chalk River. Five of the rods will be placed in the X-1 loop in-pile section replacing the samples from CR-WAPD-Met-7. The sixth rod will be placed in the out-of-pile section as a control sample. The five in-pile samples will operate at heat fluxes of 300,000, 400,000 and 500,000 Btu/hr ft² °F. One defect specimen and one undefected control specimen will be irradiated at each of the two lower flux positions. The center sample, operating at the highest heat flux, will also be defect. The two specimens at the highest heat fluxes have 0.005" diam. defects, while the other specimen is defect with a crack starting at the weld.

The specimens were fabricated from enriched UO₂ prepared by the decomposition of uranyl nitrate to UO₃ and subsequent reduction to UO₂. The samples were compacted with PVA, and Sterotex was added to the 500,000 Btu/hr ft² specimen. All were sintered at 1750°C in a hydrogen atmosphere furnace according to PWR reference specifications. The out-of-pile test period begins November 7, 1955 and the samples will be inserted in the reactor on the following shutdown. The test is scheduled to run for one year.

8. MTR-WAPD-13-6

A defect UO₂ specimen cold pressed and sintered to 93% of theoretical density and enriched to 4.7 w/o uranium-235 in UO₂ has been inserted in the HB-5 facility at MTR. It is operating at a maximum heat flux of 600,000 Btu/hr ft² and is scheduled to remain in the reactor for twelve weeks. During the first three weeks the specimen remained essentially stationary in the high flux position except for daily checks on the cycling mechanism. The specimen started operation on October 10, 1955 and the gross beta-gamma activity in the loop water one hour after sampling was 7×10^5 cpm. By October 19, 1955 this activity increased to 1.6×10^6 cpm. The specimen will remain stationary for the second three weeks and then it will be cycled in and out of the flux at 10 minute interval for three weeks. Following these the time interval for cycling will be increased to eight hours.

9. WAPD-MTR-14-Met-1

The release of fission products by leaching from a 95% dense specimen into degassed water at 650°F and 2200 psi has been determined by analyzing the UO₂ specimen, the autoclave water and the activity on the autoclave walls. The data are summarized in the following table.

<u>Element</u>	<u>% in H₂O</u>	<u>% on Walls</u>	<u>% Loss from UO₂</u>
Sr	3.0×10^{-2}	3.2×10^{-4}	0.03
Zr	2.1×10^{-2}	9.9×10^{-2}	.12
Ru	4.3×10^{-2}	9.0×10^{-2}	.13
Cs	5.7×10^{-1}	9.7×10^{-1}	.15
Ce	4.9×10^{-3}	6.1×10^{-3}	.01

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Table I

Interpretation of Powder Films of Diffusion Zones of
U₃Si-Zr Clad and Comparison with Literature References

<u>Material</u>	<u>Experimental U₃Si-Zr</u>	<u>Literature References</u>
Zirconium	Hexagonal a = 3.230 Å c = 5.139 Å Space Group P6 ₃ m2	Zr ⁽¹⁾ Hexagonal a = 3.229 Å c = 5.141 Å Space Group P6 ₃ m2
Zone 1	Hexagonal a = 3.230 Å c = 5.139 Å Space Group P6 ₃ m2	Zr ⁽¹⁾ Hexagonal a = 3.229 Å c = 5.141 Å Space Group P6 ₃ m2
Zone 2	Orthorhombic a = 3.54 b = 14.40 c = 3.68 Space Group Cmcm	ZrSi ₂ ⁽²⁾ Orthorhombic a = 3.72 b = 14.61 c = 3.67 Space Group Cmcm
Zone 3	Orthorhombic a = 4.936 Å b = 5.875 Å c = 2.860 Å Space Group = Amam	U ⁽³⁾ Orthorhombic a = 4.945 Å b = 5.865 Å c = 2.852 Å Space Group = Amam
U ₃ Si	Tetragonal a = 6.000 Å c = 8.632 Å Space Group I4/mcm	U ₃ Si ⁽⁴⁾ Tetragonal a = 6.000 Å c = 8.661 Å Space Group I4/mcm

References:

(1) Donnay, J. D. H. and Nowacki, W. (1954) Crystal Data, Memoir 60, Geological Society of America, pg. 427

(2) Ibid., pg. 266

(3) Ibid., pg. 316

(4) Ibid., pg. 367

Table II

Preliminary Results from the Corrosion Testing of
U₃Si Coextrusions with Metallic Barrier Layers

<u>Extru- sion No.</u>	<u>Ingot No.</u>	<u>Barrier Material</u>	<u>Clad-Barrier and Barrier-Core Bonds</u>	<u>Corrosion of Defected Clad Specimens in 650°F Water (Two samples)</u>
1	462	Nb	Partially unbonded at barrier-core interface	Both samples failed in 14 days due to bond line attack at barrier-core interface
3	460	Nb	Examination showed no unbonding	" "
8	504	Mo	Examination showed no unbonding	" "
12	536-2	U-10 Nb	-----	No results to date
41	538-1	Mo	-----	No results to date
42	538-2	Cu	-----	No results to date

41 Table III

Metallographic Examination, Residual Phases, Silicon Analysis and Corrosion
Rates of U₃Si Ingots Epsilonized at 800°C for 7 Days

Ingot No.	Residual Phase and (Sample Location)	Silicon Content w/o and (Sample Location)	Corrosion Rate (mg/cm ² /hr) after days -----								
			1	3	7	14	21	28	35	42	
454	alpha / delta (B)	3.83 (T)	-2.71	-1.39	-1.31	-1.16	-1.14	-1.05	Dis.*		
			(1 sample cracked up 7-14 days)								
484	delta (T)	3.69 (B)	-1.15	-0.81	-0.70	-0.80	-0.73	-0.83	-0.81	-0.84	
524	alpha / delta (B)	3.64 (T) 3.60 (B)	-0.16	Dis.							
530	delta (T)	3.79 (T)	-0.82	-0.73	-0.57	-0.55	-0.49	-0.49	-0.55	-0.62	
534	delta (B)	3.84 (T)	-0.86	-0.59	-0.51	-0.81	-0.85	-0.84	-0.86	-0.87	
536	cored alpha (T)	3.80 (T) 3.76 (B)	-1.19	-0.66	-0.63	-0.87	-0.77	-0.78	-0.80	-0.80	
			(1 sample cracked up 7-14 days)								
538	delta (B)	3.79 (T) 3.71 (B)	-1.28	-0.86	-0.68	-0.75	-0.62	-0.60	-0.64	-0.70	
548	cored delta (T)	3.83 (T)	-1.23	-1.03	-1.00	-0.96	-0.79	-0.75	-0.74	-0.75	

* Dis. indicates disintegrated
T indicates top of ingot
B indicates bottom of ingot

42 Table IV

Effect of Heat Treatment on the Density
and Corrosion Resistance of Uranium-Silicon Alloy

<u>Condition</u>	<u>Density* - (gm/cm³)</u>	<u>Corrosion Resistance in 650°F Water</u>
As-extruded	15.28	disintegrated in 24 hours
800°C - 3 days	15.43	-3.36 mg/cm ² /hr after 72 hours
850°C - 3 days	15.15	disintegrated in 72 hours
900°C - 3 days	14.34	disintegrated in 72 hours
800°C - 7 days	15.19	-3.23 mg/cm ² /hr after 72 hours
850°C - 7 days	14.96	disintegrated in 72 hours
900°C - 7 days	14.21	disintegrated in 72 hours

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* The calculated density for this alloy, assuming complete epsilonization, is 15.51 gm/cm³.
Assuming all delta plus α -U, the calculated density is 14.65 gm/cm³.

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Table V

Corrosion Properties of Bare Extruded U-3.8 w/o Si in 650°F Water

<u>Ingot No.</u>	<u>Extru- sion No.</u>	<u>Condition</u>	<u>3</u>	<u>7</u>	<u>14</u>	<u>21</u>	<u>28</u>	<u>35</u>	<u>42</u>	<u>49</u>
452	26-1	As Extruded	-1.82	-1.34	-1.00	-0.82	-0.75	-0.74	-0.74	
		Ext. / 800°C/7 day	-1.81	-1.50	-1.10	-0.94	-0.92	-0.91		
454	27-1	As Extruded	-0.81	-1.39	-1.39	-1.29	-1.26	-1.33	-1.18	Cracked
		Ext. / 800°C/7 day	-1.61	-2.24	-1.69	-1.66	-1.47	-1.32		
484	28-1	As Extruded	-0.80	-0.82	-1.15	-1.42	(Cracked up 21-28 days)			
534	534-1	As Extruded	-1.22	-1.52						
548	548-1	As Extruded	Disint.							

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Table VI

Corrosion Properties of Various Uranium-Silicon Alloys in 650°F Degassed Water

Alloy	Corrosion Rate (mg/cm ² /hr) after											
	<u>1 da</u>	<u>4 da</u>	<u>7 da</u>	<u>14 da</u>	<u>21 da</u>	<u>28 da</u>	<u>35 da</u>	<u>42 da</u>	<u>49 da</u>	<u>56 da</u>	<u>63 da</u>	
U / 3.8 w/o Si (#530)	-0.82	-0.73	-0.57	-0.55	-0.49	-0.49	-0.55	-0.62*				
U / 3.8 w/o Si (#538)	-1.28	-0.86	-0.68	-0.75	-0.62	-0.60	-0.64	-0.70*				
U / 3.8 w/o Si / 1.0 w/o Pt	-10.2	-11.6	-8.5	-4.7	-3.6	disint.						
/ 0.5 w/o Pt	-15.0	-6.6	-6.1	-4.4	-3.4	disint.						
/ 0.5 w/o Mo	-1.1	-2.1	-2.5	-2.2	-1.9	-1.9	-1.8	disint.				
/ 0.2 w/o Mo	-0.63	-0.46	-0.44	-0.46	-0.49	-0.54	-0.61	-0.64	-0.67	-0.68	-0.70*	
/ 0.5 w/o Nb	-0.97	-1.64	-1.89	-1.67	-1.54	-1.50	-1.46	-1.40	-1.39	-1.30	disint.	
/ 0.2 w/o Nb	-0.16	-0.19	-0.33	-0.53	-0.58	-0.60	-0.68	-0.70	-0.76	-0.77	-0.79*	
/ 0.5 w/o Cu	-0.64	-1.15	-1.95	-1.64	-1.55	-1.50	-1.42	-1.33	-1.33	-1.21	-1.19*	
/ 0.2 w/o Cu	-0.14	-0.29	-1.10	-0.74	-0.77	-0.81	-0.83	-0.83	-0.92	-0.90	-0.91*	
/ 1.0 w/o Ta	-3.75	-2.59	-2.27	-1.66	-1.51	-1.41*						
/ 0.5 w/o Ta	-6.9	-4.7	-3.36	-2.32	-1.85	-1.67*						
/ 0.2 w/o Ta	-1.69	-1.05	-1.02	-0.82	-0.74	-0.72*						

* still in test

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Table VIIMechanical Properties of Extruded U₃Si

<u>Specimen No.</u>	<u>Post Extrusion Heat Treatment</u>	<u>DPH</u>	<u>Mean Grain Diam. (mm)</u>	<u>Test Temp. (°C)</u>	<u>Ultimate Strength (psi)</u>	<u>0.2 % Yield Strength (psi)</u>	<u>Total Elong. (%)</u>	<u>Red. in Area (%)</u>	<u>Strain Rate (in/min)</u>
502 A-7	As Extruded	229	0.015	Room T.	85,000	---	nil	nil	0.5
502 A-9	As Extruded	229	0.015	Room T.	86,500	(fractured in shoulder section)			0.05
502 B-2 ⁽¹⁾	800°C-168 hr-F.C.211		0.034	Room T.	45,000	---	nil	nil	0.05
502 B-4 ⁽¹⁾	800 C-168 hr-F.C.211		0.034	Room T.	44,000	---	nil	nil	0.05
502 B-3	800 C-168 hr-F.C.211		0.034	390	41,000	18,900	4	4	0.05

(1). Specimen fractured at the extreme end of the reduced section.

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Table VIII

Corrosion Life of Simulated Fuel Elements Tested
in 343°C Degassed Water with 0.040 in. Defect in Clad

<u>Alloy Content</u> <u>w/o</u>	<u>Sample</u> <u>Condition*</u>	<u>No. of</u> <u>Samples</u>	<u>Corrosion Life</u> <u>Days</u>
8 Nb	A	2	84-168
	B	2	56-84
	C	2	14-70
	D	4	0-14
	E	4	42-84
	F	3	0-14
10 Nb	A	3	>182
	A	3	84-168
	B	6	28-70
	C	2	>140
	C	2	98-126
	D	8	0-14
	E	9	14-70
	F	1	>140
12 Nb	F	6	0-98
	A	2	>154
	B	3	112-140
	C	2	>126
	D	5	0-56
	E	1	>154
	E	4	56-126
	F	4	>126
10 Nb-Nb Barrier	A	2	>182
	B	2	>182
	C	2	0-28
	D	4	0-14
	E	2	>154
	E	1	56-70
	F	4	0-14
10 Nb / 1 Pt	A	3	>182
	B	3	70-112
	C	3	14-98
	D	2	>154
	E	2	28-84
	F	2	98-112

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Table VIII (Contd.)

<u>Alloy Content</u> <u>w/o</u>	<u>Sample</u> <u>Condition*</u>	<u>No. of</u> <u>Samples</u>	<u>Corrosion Life</u> <u>Days</u>
8 Mo / 1 Pt	A	3	> 182
	B	3	14-28
	C	3	0-14
	D	4	0-14
	E	4	28-112
	F	4	0-14
12 Mo / 0.5 Pt	A	2	> 182
	B	2	70-140
	C	2	56-98
	D	4	0-14
	E	4	56-98
	F	4	56-112

* A - As Extruded

B - Extruded / 900°C/24 hr/W.Q.

C - Extruded / 900°C/24 hr/W.Q. / 400°C/5 wk/W.Q.

D - Extruded / Cold Drawn

E - Extruded / Cold Drawn / 900°C/24 hr/W.Q.

F - Extruded / Cold Drawn / 900°C/24 hr/W.Q. / 400°C/5 wk/W.Q.

Table IX

Effect of Annealing on the Corrosion Properties in
650°F Water of Cold Drawn U-12 Mo Defected Fuel Elements
Using Induction Cast and Pre-homogenized Fuel

<u>Condition</u>	<u>Heat Treatment</u>	<u>Corrosion Life - Days</u>
As Extruded		>126
Cold Drawn - 40%		>42
Cold Drawn - 53%		7-14
Cold Drawn - 40%	600°C - 10 min - W. Q.	0-14
"	" 30 min "	>42
"	" 1 hr "	>42
"	" 7½ hr "	>42
"	700°C - 5 min - W. Q.	>42
"	" 10 min "	>42
"	" 30 min "	>42
Cold Drawn - 53%	600°C - 10 min - W. Q.	14-42
"	" 30 min "	14-42
"	" 1 hr "	14-42
"	" 8 hr "	(1) 28-42 (1) >126
"	" 24 hr "	28-42
"	700°C - 10 min - W. Q.	(1) 14-28 (1) >126
"	" 30 min "	(1) 14-28 (1) 84-98
"	" 1 hr "	(1) 14-28 (1) 70-84
"	" 8 hr "	7-14
"	" 24 hr "	0-7
"	900°C - 10 min - W. Q.	14-42
"	" 30 min "	14-28
"	" 1 hr "	14-28
"	" 24 hr "	0-7

Table X

Effect of Annealing on the Corrosion Properties in
650°F Water of Cold Drawn U-12 Mo Defected Fuel
Elements Using Duplex Melted and Pre-homogenized Fuel

<u>Condition</u>	<u>Heat Treatment</u>	<u>Corrosion Life - Days</u>	
As Extruded		(1) >84	>84
53% Cold Drawn	600°C - 10 min - W. Q.	(2) 14-28	28-42
"	" 30 min "	(1) 14-28	(1) 42-56
"	" 1 hr "	(1) 28-42	(1) 42-56
"	" 4 hr "		42-56
"	" 7 hr "	(1) 28-42	(1) 42-56
"	" 24 hr "	(1) 42-56	(1) 56-70
"	700°C - 5 min - W. Q.		>84
"	" 10 min "		>84
"	" 30 min "	(1) 28-42	(1) 42-56
"	" 1 hr "	(1) 0-14	(1) 14-28
"	800°C - 5 min - W. Q.	(1) 28-42	(1) 70-84
"	" 10 min	(1) 14-28	(1) 28-42
Extruded	400°C - 5 wk - W. Q.		>56
53% Cold Drawn	" " "	(1) 42-56	(1) >56

Table XI

Effect of Defect Diameter on the Corrosion Life of
Clad U-12 Mo Fuel Elements in Degassed Water at 650°F

<u>Defect Diameter</u> <u>inches</u>	<u>No.</u> <u>Elements</u>	<u>Corrosion Life</u> <u>Days</u>
0.003	4	> 154
0.013	4	> 154
0.020	4	> 154
0.040	2	> 154
	2	84-98
0.080	1	70-84
	2	98-112
	1	112-126

Zirconium Clad

Zone 1

Zone 2

Zone 3

U_3Si Core

(+ U_3Si_2 Particles,
Epsilon)

Fig. 1. Photomicrograph of Diffusion Zones in Zr- U_3Si

Neg. No. 2827-B-1

100X

Fig. 2a. Section through U-3.8 w/o Si rod with Al-6 w/o Cu barrier after 16 days in 680°F water. Acid Polish. Neg. No. 2990-B-2

8X

Fig. 2b. Section through defect hole of U-3/8 w/o Si rod with Al-6 w/o Cu barrier after 16 days in 680°F water. Note extrusion of barrier layer tending to seal defect hole in Zircaloy-2. Acid Polish. Neg. No. 2990-B-1

20X

- a. Left to right:
Zircaloy-2, Al-5 w/o
Si, U-3.8 w/o Si

Neg. No. 3345-B1-1
75X

- b. Left to right:
Zirc-2, Al-5 w/o Si

Neg. No. 3345-B1-2
500X

- c. Left to right:
Al-5 w/o Si,
U-3.8 w/o Si

Neg. No. 3345-B1-3
500X

Fig. 3. Section through U-3.8 w/o Si rod brazed to Zircaloy-2 with Al-5 w/o Si at 800-850°C after precoating the Zircaloy-2 with Al-5 w/o Si at 1000°C

a. As cast + 800°C/7 days
Neg.No. 2587-B1-2 250X

b. As cast + 800°C/14 days
Neg.No. 3214-B-1 250X

c. As cast + 800°C/21 days
Neg.No. 3214-B-2 250X

Fig. 4. Effect of heat treatment on structure of ingot #484.

a. As cast + 750°C/14 days
Neg.No. 3214-B-6 250X

b. As-cast + 850°C/7 days
Neg.No. 3214-B-3 250X

c. As cast + 900°C/3 days
Neg.No. 3214-B5-1 250X

Fig. 5. Effect of heat treatment on structure of ingot #484.

a. U-3.8 w/o Si heated 3 days at 800°C
Neg.No. 3032-B-1 250X

b. U-3.8 w/o Si heated 3 days at 850°C
Neg.No. 3033-B-1 250X

c. U-3.8 w/o Si heated 3 days at 900°C
Neg.No. 3034-B-1 250X

Fig. 6. Effect of annealing temperature on microstructure of U-3.8 w/o Si extrusion #6489 (fabricated by NMI).

a. Stringered alpha in Ext. No. 26-1
Neg.No. 2667-B-2 250X

b. Location 30 in. from "a" in Ext. No. 26-1.
Neg.No. 2668-B2 250X

Fig. 7. Variations in Extruded Microstructure in Extrusion No. 26-1.

FIGURE 8
TOTAL CORROSION CURVES
FOR EPSILONIZED U-3-8 ^{w/o}
SI ALLOY
INGOTS AND EXTRUSIONS

a. Extrusion No. 502. Corrosion tested in as-extruded condition for 96 hr in 650°F water. Neg.No. 3076-B2-1 500X

b. Extrusion No. 462. Corrosion tested in as-extruded condition for 72 hr in 650°F water. Neg.No. 2894-B-1 1000X

c. Extrusion No. 462. Annealed for 168 hr at 800°C. As heat treated, no corrosion. Neg.No. 2830-B-2 500X

d. Extrusion No. 462. Annealed for 168 hr at 800°C and corrosion tested for 168 hr in 650°F water. Neg.No. 3076-B3-1 500X

Fig. 9. Corrosion of Unclad U₃Si.

a. U-12 Mo clad stress sample with defect in tension.
Neg.No. 3056-B-1 13X

b. U-12 Mo clad stress sample with defect in compression.
Neg.No. 3055-B-1 13X

Fig. 10. Effect of stress on U-12 Mo clad defected fuel elements.

a. Tension side of extruded U-12 Mo stress sample.
Neg.No. 3051-B-2

100X

b. Compression side of extruded U-12 Mo stress sample.
Neg. No. 3051-B-1

100X

Fig. 11. Effect of stress on the hydride formation in bare
U-12 Mo rod stressed at 18,000 psi.

Tension Side

Neg.No. 2916-B-1-1

Compression Side

Neg.No. 2916-B-1-2

Fig. 12. Cathodically produced hydride in a bar bent to produce an outer fiber stress of 80000 psi.
Electrolytic Etch 250X

Fig. 13a. Corrosion at 0.007 in. defect in U-2 w/o Zr element after 1235 hr at 600°F.

Neg.No. 2614-B-1

150 X

Fig. 13b. Failure of U-2 w/o Zr after 14 days in 650°F water using 0.040 in. and 0.080 in. defects.

Neg.No. M 1735-1

1.5 X

FIGURE 14
MAGNETIC SUSCEPTIBILITY OF U-Mo ALLOYS

FIGURE 15
HYDROGEN DECOMPOSITION PRESSURE OVEN
GAMMA PHASE U-12 W/o Mo ALLOY
CORRODED IN 650 °F WATER

Fig. 16a. 90 Nb-10 U, homogenized at 900°C for 24 hr.
Neg.No. 3155-B-1 1000X

Fig. 16b. 90 Nb-10 U, homogenized at 900°C for 24 hr,
annealed at 500°C for 1 week.
Neg.No. 3156-B-1 1000X

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EFFECT OF IRRADIATION ON THE VOLUME CHANGE OF CLAD AND UNCLAD U AND U ALLOYS

FIGURE 17

CONFIDENTIAL

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