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QUARTERLY PROGRESS REPORT FUELS DEVELOPMENT OPERATION

JULY, AUGUST, SEPTEMBER, 1962

OCTOBER 15, 1962

HANFORD LABORATORIES

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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of 196 pages.

QUARTERLY PROGRESS REPORT
FUELS DEVELOPMENT OPERATION
JULY, AUGUST, SEPTEMBER, 1962

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October 15, 1962

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HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

Work performed under Contract No. AT(45-1)-1350 between the
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PHYSICAL METALLURGYIRRADIATION EFFECTS IN REACTOR METALS

Oxidation tests in a CO₂ environment on R-27, a nickel base alloy, indicate the alloy has a slightly better oxidation resistance than austenitic stainless steels. Initial corrosion testing and tensile testing of R-27 has also been completed.

The in-reactor creep test of Zircaloy-2 at 250 C and 30,000 psi stress was terminated after three reactor cycles by a heater failure. The creep curve was similar in appearance to the previous test at 310 C and 30,000 psi, exhibiting creep rates lower than out-of-reactor rates during reactor on conditions and accelerated rates during reactor outages, but differed in having a peculiar transient 265 hours after reactor shut down in the second and third cycles. A new experiment at 350 C and 30,000 psi has been initiated. Additional measurements of activation energies agree well with previous measurements showing $\Delta H_{250-300\text{ C}} = 60,000$ cal/mole, $\Delta H_{325-425\text{ C}} = 86,000$ cal/mole and $\Delta H_{450} = 60,000$ cal/mole. A creep theory based on a dislocation climb mechanism and creep retarding defects produced by irradiation has been formulated and shown to be consistent with experimental observations.

Irradiation in the G-7 hot water loop facility of the ETR has been completed on 786 tensile and bend test specimens of Zircaloy-2, AM-350, and 304, 348 and 410 stainless steels. Integrated values of fast flux obtained from iron, titanium, and nickel monitors encapsulated with these samples gave reasonable agreement, with nickel readings generally high and iron readings generally low.

Corrosion studies of Zircaloy-2 specimens in an aqueous medium showed the weight gain for in-reactor samples was four times greater than out-of-reactor tests in an identical environment.

Tensile tests were conducted on irradiated Zircaloy-2 sheet at room temperatures and at 300 C in a salt bath. As previously noted the increase in tensile strength at room temperature was much larger for specimens strained in the transverse working direction than in the rolling direction. This directional effect was eliminated, however, in 300 C tests where slip predominated over twinning as the mode of deformation in both orientations. It was also found that damage recovery was more pronounced in initially strain free material and the critical temperature for damage accumulation was ascertained to be 300 C. In spite of a marked reduction in necking strain, fracture was ductile in both annealed and cold worked specimens irradiated to 4×10^{20} nvt. A fracture mechanism for transverse specimens which is shear activated and shear controlled is proposed and the mechanism of void nucleation and growth in the necked region is also treated.

Tensile testing was completed on irradiated specimens of Armco ingot iron decarburized and denitrogenized and specimens to which 100 ppm nitrogen was added. Property changes with neutron dose in the two specimens were similar and typical for the material.

SWELLING OF IRRADIATED FISSIONABLE MATERIALS

Testing of a prototype capsule for thermal cycling of uranium metallographic specimens is in progress. Observations on samples of uranium in the extruded condition and beta heat treated condition show the damage to be a function of grain size with more extensive damage in the beta heat treated samples. Optical and electron microscopy revealed a network of pores around the original grains in the as-extruded material and a series of cracks formed in the later stages of irradiation which did not follow these networks. It was concluded that the pores were not a source of structural weakness in the material.

Zircaloy-2 clad coaxial uranium (enriched) - uranium (depleted) diffusion couples for study of fission gas diffusion showed no gas migration into the uranium (depleted) core after annealing at 950 C for 100 hours. A diffusion coefficient for the fission product gases under these conditions was estimated at $3 \times 10^{-15} \text{ cm}^2/\text{sec}$. A needle-like second phase formed in the Zircaloy-2 cladding at temperatures from 650 to 950 C.

NEUTRON DAMAGE TO METALS

A combination of swaging and upsetting operations has been determined to be the best method of introducing cold work in molybdenum samples for stored energy measurements.

Studies of the deformation mode of single crystal molybdenum samples by Laue asterism methods during tensile testing show deformation occurs by slip on $\{112\}$ planes in the $\langle 111 \rangle$ directions in low-carbon (10 to 20 ppm) samples.

Studies of irradiated molybdenum foil by electron transmission microscopy have produced the first documented observations of defects in this material. The defects appear as fine spots and loops 25 to 150 Å in diameter and are tentatively identified as clustered vacancies and/or interstitials.

SUPPORTING STUDIES

Tensile testing of 48 notch samples of Zircaloy-2 rolled sheet and coextruded cladding are in progress.

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METALLIC FUELS TECHNOLOGY

NEW FUELS DESIGN

Fabrication of experimental Zircaloy-2-clad single-component N-reactor fuel elements, shaped to allow for uranium swelling, was initiated. Three test elements, 10.5 inches long, are to be irradiated in the ETR Test Loop facilities.

Examination of test specimens having striated Zircaloy-2 cladding, irradiated to 0.2 at.% burnup at cladding temperatures in excess of 400 C, is underway. No cladding failures were observed at cladding strains up to 3.5 percent, although localized straining of the cladding was observed.

New fuel fouling detectors (crud probes) are being fabricated using Zircaloy-2-clad thermocouples. These thermocouples will be sealed into the assembly by use of a Be-Zr braze.

A series of tests have been conducted to determine the compatibility of the steel clad outer support for the N fuel element with the Zircaloy process tube. Tests involving charging elements through 5- and 17-foot sections of process tubing were successful and showed no damage to the process tube. Tests in which elements were charged through a 52-foot process tube resulted in scratching in the first test, successful charging of 17 subsequent elements, and scratching during three final tests. While the mechanism of damage is not definitely established, the most probable source of damage is the presence of minute hard particles which become embedded in the steel support surface.

The N fuel element supports are being evaluated in terms of their ability to withstand high loads, impact, and vibration. The effects of support spring constants and fluid dampening on vibration characteristics have been investigated. Analyses are underway to study the effects of geometry changes on the mechanical properties of the supports.

FABRICATION DEVELOPMENT

Development of a closure which employs copper as a bonding medium, and mechanically applied pressure combined with electrical heating to complete the bonding process, has been modified by the use of a duplex layer of copper and nickel to replace the original copper. Fully bonded closures have been achieved. Elimination of uranium contamination of the bonding layer is now under study.

In studies of alternate braze alloys, closures have been successfully made in the 1000 C range using copper as the brazing medium.

Studies are in progress to modify the hot-headed, projection-welded closure discussed in previous reports so that the final closure is fully bonded. Completion of the bond will be achieved by the use of a brazing metal between cap and clad. Experimental work indicates that preplaced materials can be encapsulated by projection welding.

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A die has been designed and fabricated for coextrusion of single-tube fuel element design with an eight-fluted external surface. Initial extrusions were successful, with cross-section dimensions within 5 percent of the design geometry.

A program is under way to develop Zircaloy-2 strip having sufficient ductility to meet the needs in the fabrication of N-fuel supports. Laboratory studies have shown that bend ductility is directly relatable to sheet texture. Fabrication procedures have been found which will produce acceptable sheet. A rapid test has been developed to measure bend ductility, and the results of this bend test have been correlated with other measurable sheet properties.

FUEL EVALUATION

A dual-enriched single-tube N-reactor size fuel element, now undergoing its fourth cycle of irradiation in the M3 Loop of the ETR, has reached a maximum exposure of approximately 700 Mwd/ton. This fuel element has shown no deleterious effects of irradiation with the exception of having warped towards the reactor core as a result of the severe radial flux gradient. Decreasing power generation rates have reduced the maximum fuel operating temperature to approximately 450 C from an original 520 C.

Fuel elements containing closures utilizing the 5 wt % Be-95 wt % Zircaloy-2 alloy have been irradiated in cold water and destructively examined. All end closures have been checked and found free of cracks in the braze or other signs of incipient failure. Several sections show extensive cracking and void formation in the uranium in the heat-affected zone adjacent to the braze. The voids may have occurred by grain boundary tearing during irradiation, or may in part have been caused by the brazing process prior to irradiation.

An empirical model for the incremental swelling of the uranium fuel has been developed as an aid in interpreting experimental fuel swelling results. This model, which contains seven parameters which can be adjusted to give the best agreement between estimated and measured fuel swelling, has been used to evaluate the swelling data from 12 irradiated test elements. The evaluation indicates that uranium fuel swelling is extremely temperature sensitive in the range of 400 C with the "swelling temperature" decreasing with increased fuel burnup.

METALLIC FUEL ELEMENT DEVELOPMENT

Pilot billets of Th-2.5 wt % U and Th-2.5 wt % U-1 wt % Zr have been fabricated by double vacuum arc melting. Work on these pilot ingots to date indicates that the Zr addition lowers room temperature hardness, improves ingot sidewall quality, improves uranium distribution, and adds to the strength of welded joints in the primary electrode.

Three pilot thorium alloy ingots were successfully coextruded into Zircaloy-2-clad 0.875-inch OD x 0.495-inch ID tubes. The coextrusions were performed at a temperature of 760 C with an extrusion ratio of 17.4:1. The billet which contained 1 wt % Zr addition produced the best quality coextrusion. The addition of the 1 wt % Zr does not show any significant effect on the extrusion force at 760 C preheat temperature. At the 760 C extrusion temperature, significant bonding occurs between the copper shell and the Zircaloy cladding. Initial difficulty was encountered in obtaining uniform bright etching of the Zircaloy cladding. A more stringent copper stripping procedure has produced an etch of satisfactory quality.

The two Th-2.5 wt % U and the Th-2.5 wt % U-1 wt % Zr alloy pilot coextrusions, clad in Zircaloy-2, have been evaluated for cladding uniformity, clad-core bonding, and fuel structure. Cladding uniformity is extremely good in both alloys with both inner and outer cladding thickness variation being approximately 0.0005 inch. This indicates that as-cast machined billets may be used without primary working. Clad-core interfaces were clean with no evidence of unbonding. The thorium alloy cores were only partially recrystallized at this extrusion temperature. Fuel sections are undergoing a series of heat treatments to observe the effect on clad-core diffusion and fuel structure.

A brazed closure will be utilized for thorium-uranium metallic fuel specimens. This closure requires recessing the cut ends of each specimen. The chemical milling solution normally used for metallic uranium coextrusions (a mixture of HNO_3 , H_2SO_4 , and CuSO_4) did not attack the thorium alloy. A 10 percent aqueous HCl solution at 50 C appears to be a satisfactory milling bath. Addition of 1 percent Zr to the thorium-uranium alloy appears to enhance both surface characteristics and etch rate.

An initial irradiation test has been planned utilizing the ETR P-7 pressurized loop facility. Three 8-inch-long tubular elements will be irradiated in high temperature pressurized water. Initially these test elements will operate with a maximum fuel temperature of 620 C and a maximum heat flux of $1.1 \times 10^6 \text{ Btu}/(\text{hr})(\text{ft}^2)$. Dimensional stability and fuel swelling will be determined during the course of irradiation by interim measurement in the ETR canal.

CERAMIC FUELS DEVELOPMENTCERAMIC (URANIUM) FUELS RESEARCH

Graphite was welded by the magnetic force resistance welding technique to produce a high quality weld zone in which the graphite had smaller crystal size and higher density than the base material.

Physical properties of UO_2 that were determined include: the heat of fusion, 3300 cal/mol; the surface tension of liquid UO_2 at 2800 C, 490 dyne cm^{-1} ; the change in volume on melting, 9.6 percent; and the coefficient of thermal expansion between 2800 C and 3100 C, $3.7 \times 10^{-5} \text{ C}^{-1}$.

The increase in melting point that was observed earlier after low irradiation exposures of sintered, polycrystalline UO_2 was not observed with irradiated single crystal specimens, presumably because of the lack of excess oxygen over that required for the stoichiometric compound.

The liquidus of the UO_2 - ThO_2 system was measured, although extensive separation of the UO_2 and ThO_2 by sublimation caused experimental difficulties.

Measured thermal conductivity (to 1200 C) of a UO_2 single crystal is described by the equation,

$$k_{\text{obs}} = 0.0323 + 17.9 T^{-1} + 4.1 \times 10^{-12} T^3 + 7.5 \exp \frac{-9580}{T} \text{ watt cm}^{-1} \text{ } ^\circ\text{K}^{-1}.$$

Long irradiation (e. g. to 15,000 MWD/T) produced different microstructures in UO_2 than did short irradiations, presumably by continued, slow void migrations to the center after the fuel temperature profile was modified by columnar grain formation early in the irradiation.

Segregation of fission fragments in swaged UO_2 irradiated to more than $4 \times 10^{20} \text{ f/cc}$ was less than previously observed in specimens irradiated to $0.08 \times 10^{20} \text{ f/cc}$. This is attributed to a continual change in the relative effectiveness of each of several contributing mechanisms as the irradiation proceeds.

The microstructural features of UO_2 irradiated at high heat rating were correlated with the radial limit of the initially molten region by observations of which tungsten markers moved.

A proper combination of impact pressure, preheat temperature, specific surface and O/U ratio permits high energy-rate impaction of UO_2 to greater than 99 percent TD. The effects of changing these variables were studied. UN was compacted to 99.5 percent TD and a 50 wt % UO_2 -W powder mixture was compacted to 96 percent TD to produce a uniform dispersion of UO_2 in a continuous tungsten matrix.

Sintering between particles of swaged UO_2 during irradiation at temperatures as low as 300-400 C was confirmed by microscopic examination of irradiated PRTR fuel rods.

A 2.33-inch diameter single rod test element that split-failed during irradiation was observed during postirradiation examination to have developed much greater fuel temperatures than anticipated from the results of previous tests in the same facility. Out-of-reactor tests of the collapse characteristics of the 2.33-inch OD by 0.060-inch wall Zircaloy show that the 400 C collapse pressure can be increased from 1100 psi to 1500 psi by "hanging" the cladding tube from an external, coaxial process tube by electron beam welding five equally spaced, radially oriented ribs between the two tubes.

The second phase of the remote fuel element fabrication study, now begun, will involve remote refabrication demonstrations using techniques and equipment developed during the initial phase.

PHYSICAL METALLURGYIRRADIATION EFFECTS IN REACTOR METALSAlloy Selection - T. T. Claudson

The alloys to be studied under the program to determine the irradiation effects in reactor metals must represent both the alloys being used in existing reactors and those alloys whose properties indicate they will be suitable for future reactors. Several such alloys have been selected for study as part of a coordinated effort by several research laboratories to determine the effects of irradiation on reactor structural materials. All alloys which will be tested by more than one participating site will be procured, stored and distributed by Hanford Laboratories. A survey has been made to determine program needs for the various alloys, their quantity, and their required form.

Specifications have been written for material in the form of plate and bar for 304 SS, 348 SS, AM355, Inconel X-750, Inconel 600, Zr-2, A302B, and A212B alloys. Of these, 10 tons each of A212B and A302B pressure vessel plate 4 inches in thickness will be purchased from United States Steel Corporation. In 1959, the United States Steel Corporation Research Laboratories obtained heats of A212B, HY80, and T-1 low alloy steel plates. An extensive program was completed by USS in order to fully document these heats which were subsequently given to laboratories for various testing programs. Hanford Laboratories was able to obtain only 3-square feet of 4-inch A212B plate, 4-square feet of 6-inch A302B plate, and 20-square feet of 3-inch HY80 plate for the coordinated program. The small quantities of the A212B and A302B are insufficient for the present program needs but will be used to correlate data obtained from the additional material to be procured.

Corrosion, oxidation, and tensile specimens have been prepared for testing of an experimental alloy developed by Allegheny Ludlum Steel Corporation. Similar specimens are being prepared from sheets of Rene' 41, Hastelloy N, and R-235 alloys for similar testing. The R-27 alloy heated to 1700 F and 1800 F has been exposed to CO₂ environments and the oxidation determined by continuous weight-gain measurements. The data for these two tests are shown in Figure 3.1. These data indicate that the alloy has a slightly better resistance to oxidation in this environment than austenitic stainless steels. The results of one additional test are shown in Figure 3.2. In this case, six specimens were subjected to deoxygenized water at 1022 F and 3000 psi and the data plotted for the average values. Similar data are plotted for 304 L stainless steel, Inconel X-750, and 17-4 ph alloys for comparison purposes.

Initial tensile tests have been completed on sub-size sheet tensile specimens of the R-27 alloy. Specimens were heated in two ways: first,

FIGURE 3.1

Oxidation of Alloy R-27 in Carbon Dioxide Environment at Temperatures of 1700 F and 1800 F

FIGURE 3.2

Corrosion of R-27 Alloy in 3000 psi, 1022 F Deoxygenized Water

by heating the specimens to 1400 F for 8 hours, air cooling and then holding at 1400 F for 24 hours and final air cooling (condition A), second, specimens were heated to 1400 F for 24 hours, air cooled, reheated to 1200 F for 24 hours and air cooled (condition B). Annealed specimens were also tested. These data are presented in Table 3.1. No significant difference was evident due to the longer aging time in condition B.

TABLE 3.1

INITIAL TENSILE TESTS

<u>Heat Treatment</u>	<u>0.2% Yield Strength, psi</u>	<u>Ultimate Tensile Strength, psi</u>	<u>Uniform Elongation, %</u>
Annealed	51,000	110,000	52
Condition A	182,000	210,000	10.0
Condition B	175,000	209,000	9.5

In-Reactor Measurements - L. J. Chockie, J. J. Holmes and J. A. Williams

The objective of the in-reactor measurements program is to define the effects of fast neutron irradiation on the physical and mechanical properties of reactor structural materials. At the present time the in-reactor creep properties of 20% cold worked Zircaloy-2 are being studied. Four in-reactor creep tests have been conducted to date. The initial test was conducted in a prototype creep capsule at 260 C and 285 C at a stress of 30,000 psi. Subsequent tests have been conducted in improved in-reactor creep apparatus at 600 C, 310 C and 250 C. The details of all the above creep tests are presented in previous quarterly progress reports.

In-Reactor Studies

The in-reactor creep test at 250 C and 30,000 psi stress which was described in part in the progress report for April, May and June 1962 was continued through another shutdown. Further continuation of this test was precluded by a heater failure. The entire creep curve is shown in Figure 3.3. The features of the 250 C and 30,000 psi test are similar to those of the 310 C and 30,000 psi test^(1, 2) in that creep rates during reactor on periods are generally less than out-of-reactor and increase with time during reactor outages to rates greater than out-of-reactor.

The major differences between the two curves, ignoring the difference in total strain due to differences in stress, are the transients which occurred about 265 hours after the second and third shutdowns in the 250 C test. Similar transients were not seen in the 310 C test. Detailed analysis of all available experimental variables which might account for the transients has not revealed a variable which could produce such large changes in creep rate. It must therefore be concluded that the transients are the result of the buildup of some irradiation produced defect which reduces creep resistance. It has been suggested that an excess vacancy build up could increase creep rates.⁽³⁾

FIGURE 3.3

Creep of Zircaloy-2 at 250 C and 30,000 psi Stress

In order to continue the study of the in-reactor creep properties of Zircaloy-2 a creep test at 350 C and 30,000 psi was started. The test has been running about 320 hours during which time one reactor outage occurred. The creep rate in-reactor before the first outage was generally higher than in a similar out-of-reactor test (Figure 3.4). A quantitative description of the in-reactor and out-of-reactor creep rates prior to the first shutdown cannot be given as both tests were in primary creep where rates are changing rapidly. During the reactor outage, the in-reactor creep increased from the rate existing just before the outage to about 5.5 times that of the out-of-reactor rate. During startup the rate decreased to a steady state rate of 3.9 times the out-of-reactor rate. The total plastic strain in-reactor to date is about 1.86% while the out-of-reactor strain is about 0.76%.

The study of the activation energies for in-reactor creep of Zircaloy-2 has been continued. A new series of measurements has been made by the temperature cycle method. The measurements were made in a creep capsule having one lead heater out. One end of the specimen was, therefore, always 10 to 40 degrees below the control temperature of the specimen. The effect of the temperature gradient along the specimen is not very important in activation energy calculations as long as the test temperatures do not include those at which strain aging occurs.

Activation energies were calculated with data from both positive and negative temperature changes. The values found are shown in Table 3.2 and are presented in Figure 3.5 along with activation energies determined

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FIGURE 3.4

Creep Curves for 20% Cold Worked Zircaloy-2 of 30,000 psi and 350 C

FIGURE 3.5

In-Reactor Activation Energies for Zircaloy-2 Creep

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in a previous in-reactor capsule.⁽²⁾ The agreement between the two sets of data is good.

TABLE 3.2

ACTIVATION ENERGIES			
<u>Temperature, C</u>	<u>Stress, psi</u>	<u>$\Delta T^{\circ}C$</u>	<u>ΔH, cal/mole</u>
275	36,000	+16	62,100
301	39,200	+21	66,800
326	30,250	+21	57,100
348	30,500	+25	86,400
373	30,500	-22	87,900
375	30,700	+22	86,000

In-Reactor Creep Theory

Analysis of in-reactor and out-of-reactor creep behavior has lead to a consistent theory for in-reactor creep. The theory is based on a number of mechanistic interpretations of experimental data. These are as follows:

1. The creep of Zircaloy-2 at 30,000 psi and temperatures including 250 C and above is controlled by the climb of dislocations over barriers of various types.
2. During irradiation, creep rates are less at 310 C and 250 C than in similar out-of-reactor tests. The low creep rates during irradiation can be interpreted as being the result of an increase in obstacle density due to radiation damage.
3. During reactor outages, creep rates increase with time to a value which is greater than that of the out-of-reactor tests. The increase in rates during outages is attributed to the removal of the obstacles through annealing. The fact that the rates attained after annealing are greater than out-of-reactor is a very important result. This can only mean that the cold worked substructure loses much of its creep resistance during irradiation.
4. The number of irradiation produced climb obstacles at steady state can be expressed as

$$P(\varphi) = K(N_1)$$

where $P(\varphi)$ is the rate of obstacle production, N_1 = number of obstacle per unit volume and K is the reaction constant for obstacle removal through annealing. K can be written as $k \exp(-\Delta H_p/RT)$

where ΔH_p is the activation energy for obstacle annealing and k is another constant. Solving for N_i

$$N_i = \frac{P(\varphi)}{k} \exp\left(\frac{+\Delta H_p}{RT}\right).$$

Following Weertman⁽⁴⁾, ex-reactor creep rates in the climb range can be expressed as

$$\dot{\epsilon} = A \frac{1}{N^{2/3} h} \exp\left(\frac{-\Delta H_c}{RT}\right)$$

where A is a near constant for a material, N is the volume concentration of climb obstacles, ΔH_c is the activation energy for creep and is about equal to the activation energy for self-diffusion, and h is the height of the obstacle.

In-reactor in the presence of annealing and production of obstacles the steady state creep rate can be written

$$\dot{\epsilon}_i = A_i \left(\frac{1}{N^{2/3} h + N_i^{2/3} h_i} \right) \exp\left(\frac{-\Delta H_c}{RT}\right).$$

Since creep rates during irradiation are much lower than in the absence of irradiation,

$$N_i^{2/3} h_i \gg N^{2/3} h$$

hence

$$\dot{\epsilon}_i = A_i \frac{k}{P(\varphi)} \frac{1}{2/3} \exp\left(\frac{\Delta H_c + 2/3 \Delta H_p}{RT}\right). \quad (1)$$

The activation energies determined in-reactor tend to confirm equation (1). In the damage annealing range where dislocation climb controls creep, the activation energy calculated by the temperature cycle technique will depend upon the speed with which the steady state number of obstacles is established. Thus, at the lower end of the annealing range, annealing rates are slow and the obstacle concentration will be approximately the same before and after the temperature change. The activation energy in this case will be ΔH_c . ΔH_c out-of-reactor has a value of 58,500 cal/mole. The in-reactor activation energy below 325 C is near 60,000 cal/mole which agrees with equation (1) when annealing rates are slow. At higher temperatures annealing rates are fast and the equilibrium number of defects is established during the temperature change and activation energy of $\Delta H_c + 2/3 \Delta H_p = 86,000$ cal/mole is observed.

In summary the low in-reactor creep rates are believed to be the result of an increase in obstacle density with irradiation. In the temperature range where the in-reactor creep properties of Zircaloy-2 are being studied the irradiation produced obstacles can anneal. The steady state obstacle production is equal to the rate of annealing. The increase in rate during reactor outages is the result of a decrease in the density of irradiation produced obstacles through annealing. In addition, the original creep resistance is reduced by irradiation.

In view of the temperature dependence of the irradiation produced obstacle concentration and the loss of the preirradiation creep resistance it must be concluded that a temperature exists where in-reactor creep rates are greater than out-of-reactor rates. The 350 C in-reactor test confirms this latter conclusion.

Irradiation and Environmental Effects - A. L. Bement, J. E. Irvin, and D. A. Jones

The purpose of this phase of the program is to investigate the combined effects of radiation and reactor environment on the mechanical properties of structural materials. Special attention will be given to the determination of mechanical property changes produced in metals by irradiation at elevated temperatures in contact with water. Materials to be investigated include zirconium-base alloys and other alloys showing promise for high-temperature applications. Tensile and bend properties of these metals after irradiation under specific reactor operating conditions are to be determined. Notched tensile specimens will be employed to determine brittle failure tendencies in the irradiated materials, with the objective of predicting conditions under which catastrophic failure is likely to occur.

Loop Operation

With the conclusion of ETR Cycles 46, 47, and 48, 14 quadrants containing tensile and bend test specimens were discharged from the G-7, hot-water loop facility. The estimated neutron fluxes and exposures (>1 Mev) for these quadrants are given in Table 3.3. At the conclusion of Cycle 48, a total of 786 specimens have been irradiated to exposures ranging from about 5×10^{19} to about 4×10^{21} nvt (fast) and discharged. A total of 514 of these specimens have been shipped to HAPO for testing. Of the 238 specimens discharged at the conclusion of Cycles 46, 47, and 48, 22 were Zircaloy-2 bend test specimens and the remainder were tensile specimens of AM-350 and types 304, 348, and 410 stainless steels.

The operating temperature and history for the G-7 loop for Cycles 46 and 47, are shown in Table 3.4. Deviations from the normal control temperature of 540 F occurred in both cycles; however, temperature efficiency as a whole was satisfactory considering the fluctuating nature of Cycle 47. Temperature efficiency is expected to improve with re-establishment of the regular 6-week cycles beginning approximately October, 1962. A total of 170 effective days of loop operation at about 540 F have been realized from Cycle 39 through 47, corresponding to 289 calendar days.

Operating cycles for the out-of-reactor loop simulating G-7 loop operation for ETR Cycles 39 through 45 have been completed. The temperature-time and discharge schedules for Cycles 43, 44, and 45 are given in Table 3.5.

TABLE 3.3
QUADRANT DISCHARGES FOR ETR CYCLES 46, 47, AND 48

Quadrant Number	Number and Type of Specimens	Estimated Flux, nv	Estimated Exposure nvt
19	18 Zircaloy-2 Tensile	1.7×10^{14}	4.3×10^{21}
20	18 Zircaloy-2 Tensile	1.7×10^{14}	4.3×10^{21}
43	18 348-SS Tensile	1.73×10^{14}	3.24×10^{21}
44	12 304-SS Tensile	1.73×10^{14}	3.24×10^{21}
	6 348-SS Tensile	1.73×10^{14}	3.24×10^{21}
49	18 348-SS Tensile	1.73×10^{14}	5.4×10^{20}
50	12 304-SS Tensile	1.73×10^{14}	5.4×10^{20}
	6 348-SS Tensile	1.73×10^{14}	5.4×10^{20}
53	18 AM-350 Tensile	6.9×10^{13}	5.9×10^{19}
54	12 410-SS Tensile	6.9×10^{13}	5.9×10^{19}
	6 AM-350 Tensile	6.9×10^{13}	5.9×10^{19}
55	18 AM-350 Tensile	1.2×10^{14}	1.8×10^{20}
56	12 410-SS Tensile	1.2×10^{14}	1.8×10^{20}
	6 AM-350 Tensile	1.2×10^{14}	1.8×10^{20}
63	11 Zircaloy-2 Bend Test	1.6×10^{14}	4.5×10^{20}
64	11 Zircaloy-2 Bend Test	1.6×10^{14}	4.5×10^{20}
67	18 AM-350 Tensile	1.6×10^{14}	4.5×10^{20}
68	12 410-SS Tensile	1.6×10^{14}	4.5×10^{20}
	6 AM-350 Tensile	1.6×10^{14}	4.5×10^{20}

Total number of specimens discharged, Cycles 46, 47, and 48 = 238

Total number of specimens discharged to date = 786

TABLE 3.4
TEMPERATURE AND OPERATING HISTORY OF ETR G-7 FACILITY

Cycle	46	47
<u>Reactor</u>		
Startup ⁽¹⁾	5-9 -62	7-8-62
End of Cycles	6-11-62	8-6-62
Megawatt Days	5000	4118
Effective Days at 175 Mw	28.6	23.5
Number of Scrums ⁽²⁾	3	15
Number of Shutdowns	1	2
<u>G-7 Loop</u>		
Maximum Temperature	550 F	525 F
Effective Days Above 200 F	28.4	23.9
Effective Days at Operating Temp	25.7	19.1
Operating Efficiency ⁽³⁾	99.2%	100.2%
Temperature Efficiency ⁽⁴⁾	90.5%	80.0%

(1) Includes flux run

(2) Power drops below half full power with immediate recovery.

(3) Effective days of loop operation above 200 F with respect to effective days of reactor operation.

(4) Effective days of operation between 500 and 550 F with respect to effective days of operation above 200 F.

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TABLE 3.5
TEMPERATURE - TIME AND DISCHARGE PROGRAM FOR
OUT-OF-REACTOR LOOP

	<u>ETR Cycle Number</u>		
	<u>43</u>	<u>44</u>	<u>45</u>
Time at Temperature (hours)			
392 F	3.0	35.5	20.5
425 F	1.0	27.0	3.0
475 F	10.0	28.0	6.5
540 F	<u>292.5</u>	<u>528.5</u>	<u>531.5</u>
Total Time (hours)	306.5	619.0	561.5
Discharges (number and type of specimen)			
Zircaloy-2			
Tensile		24	
Notch Tensile			48
Corrosion Blanks	<u> </u>	<u>4</u>	<u> </u>
Total Number of Specimens	0	28	48
<u>Flux Measurements</u>			

Specific activities of nickel, titanium, and iron flux dosimeters have been determined for ETR Cycles 39, 40, and 41. The dosimeters, one of each metal, were enclosed in stainless capillary tubing, which was in turn spot welded to each quadrant of specimens. The tubes of dosimeters were removed from the various quadrants upon discharge, and transferred to the MTR-ETR counting laboratory for activation measurements. The specific activities for nickel and titanium are given in Table 3.6.

In calculating the fast flux from the $Ti^{46} (n, p) Sc^{46}$ reaction, the preferred value for the effective cross section, σ , was 8 mb. The following constants were also used: (1) the fraction of neutrons in a fission spectrum having energies above 1 Mev is 0.692, (2) the isotopic abundance of Ti^{46} is 7.93%, and (3) the half life of Sc^{46} is 84.1 days. Manganese was separated from iron by ion exchange prior to measuring specific activity from the $Fe^{54} (n, p) Mn^{54}$ reaction.

TABLE 3.6

SPECIFIC ACTIVITIES FOR TITANIUM AND NICKEL FLUX DOSIMETERS

Quadrant Number	ETR Cycles	Ti ⁴⁶ (n, p) Sc ⁴⁶ d/s/mg (10 ⁴)*	Ni ⁵⁸ (n, p) Co ⁵⁸ d/s/mg (10 ⁵)**
1	39, 40	2.7	3.55
2	39, 40	0.76	3.87
3	39, 40, 41	- -	14.3
4	39, 40, 41	2.3	12.7
5	39, 40	- -	6.7
6	39, 40	1.4	7.5
7	39, 40, 41	5.5	25.0
8	39, 40, 41	- -	21.5
13	39, 40	2.9	41.6
14	39, 40	2.5	- -
15	39, 40, 41	10.6	30.8
16	39, 40, 41	7.9	26.7

* Counted 6-21-62

** Counted 4-24-62

- - Flux wire not recovered

Correction for the burnout of Co⁵⁸ in the Ni⁵⁸ (n, p) Co⁵⁸ reaction required knowledge of the thermal flux for the various positions within the loop. Although Al-Co dosimeters were not attached to the quadrants, adequate thermal flux data from previous flux mappings of the GEH-20, 6 x 9 core position (which accommodates the G-7 loop) were available. These values are given in Table 3.7 along with the value of fast flux calculated from the following equation developed by Yoshikawa and Morgan:⁽⁵⁾

$$nvt (E > E_1) = K_{E_1} \frac{A e^{\lambda_2 t_0} (\lambda_1 + \sigma_1 \varphi_{th}) (\lambda_2 + \sigma_2 \varphi_{th}) t_i}{N \lambda_2 [\sigma (\lambda_1 + \sigma_1 \varphi_{th}) + \sigma_m \lambda_1] [1 - \exp^{-(\lambda_2 + \sigma_2 \varphi_{th}) t_1}]}$$

where:

$$K_{E_1} \text{ (for } E_1 = 1 \text{ Mev)} = 1.07$$

nvt = integrated fast neutron flux

σ = activation cross section for Ni⁵⁸ (n, p) Co⁵⁸
(stable isomer)
= 64 mb

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- σ_m = activation cross section for Ni^{58} (n, p) Co^{58m}
 (metastable isomer)
 = 28 mb
 λ_2 = Co^{58} decay constant ($t_{1/2} = 71.3$ days)
 = $9.72 \times 10^{-3} \text{ day}^{-1}$
 λ_1 = Co^{58m} decay constant ($t_{1/2} = 9.1$ hrs) 1.83 day^{-1}
 σ_2 = Co^{58} thermal neutron absorption cross section
 = 1650 b
 σ_1 = Co^{58m} thermal neutron absorption cross section
 = $1.7 \times 10^4 \text{ b}$
 N = Number of Ni^{58} atoms/mg = 6.95×10^{18}
 ϕ_{th} = Thermal neutron flux

TABLE 3.7

INTEGRATED FAST NEUTRON FLUXES FOR Ni^{58} (n, p) Co^{58} REACTION

Quadrant Number	Position (Inches from top of Core)	Thermal flux (10^{14} nv)	Integrated Fast* Neutron Flux (10^{20} nvt)
1	3	1.1	0.514
2	3	0.93	0.530
3	3	0.93	1.20
4	3	1.1	1.12
5	8	2.2	1.31
6	8	1.9	1.31
7	8	1.9	3.83
8	8	2.2	3.96
13	18	3.5	10.7
14	18	3.0	- -
15	18	3.0	7.2
16	18	3.5	7.13

* $E > 1 \text{ Mev}$

- - Flux wire not recovered

The densities and integrated values of fast flux for iron, titanium, and nickel are compared in Table 3.8. In this table it is seen that reasonable agreement (with exception of quadrant 1) is obtained in the integrated fast neutron flux among the three different dosimeters. By comparison,

nickel gives relatively high values and iron relatively low values over the flux range represented. The percentage difference varies from about 10 to 25% for lower to higher thermal fluxes, respectively, suggesting that present calculations may overestimate Co⁵⁸ burnout.

TABLE 3.8

INTEGRATED FLUX COMPARISONS FOR IRON, TITANIUM,
AND NICKEL DOSIMETERS

Quadrant Number	Ti ⁴⁶ (n, p) Flux (10 ¹³ nv)	Sc ⁴⁶	Fe ⁵⁴ (n, p) Flux (10 ¹³ nv)	Mn ⁵⁴	Integrated Fast Flux (10 ²⁰ nvt)	
					Fe	Ti
						Ni
1	7.4	- -	- -	- -	1.8	0.51
2	2.1	2.1	0.52	0.52	0.52	0.53
3	- -	2.5	1.1	- -	- -	1.2
4	2.9	2.1	0.94	1.3	1.3	1.1
5	- -	- -	- -	- -	- -	1.3
6	3.8	3.7	0.92	0.94	0.94	1.3
7	6.9	6.5	2.9	3.1	3.1	3.8
8	- -	- -	- -	- -	- -	4.0
13	8.0	- -	- -	2.0	- -	- -
14	6.8	- -	- -	3.0	- -	- -
15	13.0	12.0	5.4	5.9	7.2	7.2
16	10.0	10.0	4.5	4.5	7.1	7.1

The relatively higher measurement of fast neutron flux from nickel dosimeters is evident from the plot of Figure 3.6. This plot represents neutron flux (determined from nickel, titanium, and iron dosimeters) for ETR Cycle 45 as a function of distance from the top of the core filler piece in the G-7 loop. The dosimeters for these measurements were placed in an aluminum rod which was in turn encapsulated in a stainless-steel tube. Figure 3.7 shows a layout of the aluminum container, the stainless-steel capsule, the specimen-quadrant rod hanger, and the flow basket. Figure 3.8 shows the components assembled, ready for insertion into the G-7, hot water loop. In addition to the Fe, Ti, and Ni fast flux monitors, an Al-0.1 wt % Co thermal flux monitor was placed in each of the seven quadrant levels. A new capsule of monitors was inserted each cycle in order to obtain a cycle-by-cycle record of neutron flux in addition to the accumulative record obtained over several cycles from the quadrant dosimeters.

Reference to Figure 3.6 shows that titanium gives higher flux values for the upper half of the loop, nickel gives higher values for the lower half and iron gives lower values throughout. Titanium occasionally gives spurious values (see, for example, Table 3.8, Quadrant 1 and Figure 3.6, Quadrants 4 and 7) which are as yet unexplained.

FIGURE 3.6

Variation of Fast Flux ($\phi > 1$ Mev) Over the Length of the GEH-20 Loop, ETR Cycle 45

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FIGURE 3.7
Layout of ETR G-7 Loop Capsule Components

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FIGURE 3.8
Assembly of ETR G-7 Loop Capsule

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Weight Gain Measurements

Weight-gain measurements have been made on a series of Zircaloy-2 bend-test specimens representing 0, 10, 20, and 40% cold work and both the longitudinal and transverse directions with respect to rolling. Irradiations were conducted in the ETR, G-7, hot-water loop (controlled at 540 F) to estimated fast-neutron fluxes ranging from 2.1 to 3.7×10^{13} nv (> 1 Mev). The specimens were $3 \frac{3}{8} \times 1$ -inch plates, 0.05 inch in thickness, with a total surface area of 0.46 dm^2 (square decimeters).

Weighings were conducted on a Mettler, B-5 balance (200 gram range) to 0.1 mg, and corresponding specimens were measured before and after irradiation. Prior to irradiation, all of the specimens were autoclaved at 300 C for 48 hours. This treatment developed a black, coherent oxide layer resulting in an average weight gain/specimen of 7 mg/dm^2 . The appearance of the oxide layer did not change with reactor loop exposure. Demineralized water is used for loop makeup, and is adjusted to PH 10 with LiOH additions. Pressurization is done in a helium dome, and hydrogen additions are not made to control oxygen concentration. Total solids are negligible (about 0.2 ppm) and chloride ion concentration is less than 10 ppb.

Both net and specific weight-gain values for the specimens examined are given in Table 3.9. Each data entry represents the average of three specimens, so that the average value per quadrant of specimens corresponds to 12 determinations. Flux values are based on nickel, titanium, and iron flux-wire activations obtained during ETR Cycles 39, 40, and 41. Since the bend-test specimens were irradiated during ETR Cycles 42, 43, and 44, some small shift might be expected in the absolute flux density. However, changes in relative fluxes among the quadrants are expected to be small. The effective time given in Table 3.9 represents the accumulated time during which the specimens were exposed to water at 540 F. This time represents about 85% of the time that the specimens were exposed to pile neutrons at full reactor power.

Average weight-gains for the various quadrants are plotted as a function of fast neutron flux in Figure 3.9. The unirradiated values represent controlled, out-of-reactor loop exposures using identically-prepared specimens. The temperature history of this loop was programmed to duplicate that of the in-reactor loop, and water quality was duplicated as closely as possible. Consequently, the net difference in weight gain between the in-reactor and out-of-reactor loop specimens reflects flux effects only.

The plots of Figure 3.9 indicate a marked effect in the aqueous corrosion of Zircaloy-2. The curve representing 19.2 days exposure exhibits a leveling off beyond about 2.5×10^{13} nv and indicates that a power function may exist of the form

$$R(O)_t = A(t) O^n$$

where R = the corrosion rate

$A(t)$ = constant related to unirradiated value of weight gain for time t

O = neutron flux (nv)

n = constant

The 19.2 day curve breaks over too sharply at 3.8×10^{13} nv to give good fit to the above expression. However, forthcoming weight-gain data at higher fluxes may provide a better test of the flux relationship.

FIGURE 3.9

Weight Gain of Zircaloy-2 as a Function of Neutron Flux at 540 F

Reference to Figure 3.9 shows that the corrosion rate varies with flux. For example, the weight gain between 19.2 and 34.2 days in the out-of-reactor tests is only 2 to 3 mg/dm²; whereas, the corresponding weight gain for 2.1×10^{13} nv is 13 mg/dm². This is indicative that flux induced corrosion is in the breakaway region, which is consistent with the high specific weight gains observed. It is quite apparent in Table 3.9 that corrosion is independent of cold work. Although dissolved oxygen in the radiolytic water might have a significant effect on weight gain, the actual influence is as yet unknown.

TABLE 3.9

WEIGHT GAINS OF IRRADIATED ZIRCALOY-2

<u>Quadrant</u>	<u>% Cold Work</u>	<u>Flux nv, ($\times 10^{13}$)</u>	<u>Effective Time, Days</u>	<u>Net Weight Gain (Avg of 3), mg</u>	<u>Specific Weight Gain mg/dm²</u>
45	0	2.5	19.2	18.5	39.9
	10			19.5	42.0
	20			18.3	39.4
	40			19.0	40.9
					Avg 40.6
46	0	2.1	19.2	16.9	36.4
	10			18.6	40.1
	20			17.6	37.9
	40			17.6	37.9
					Avg 38.1
47	0	3.7	19.2	20.5	44.2
	10			20.6	44.4
	20			19.8	42.7
	40			19.7	42.4
					Avg 43.4
48	0	3.7	19.2	18.7	40.3
	10			20.3	43.8
	20			19.6	42.2
	40			19.6	42.2
					Avg 42.1
81	0	2.5	34.2	25.5	55.0
	10			24.8	53.4
	20			22.5	48.5
	40			25.6	55.2
					Avg 53.0
82	0	2.1	34.2	25.4	54.7
	10			25.3	54.5
	20			22.4	48.3
	40			22.7	48.9
					Avg 51.6

Mechanical Properties

Tensile specimens irradiated during ETR Cycles 39, 40, and 41 have been tested both at room temperature and at 300 C, and the results are listed in Table 3.10. Room temperature tests were conducted with an Instron Tester at a constant crosshead rate of 0.005 inch/min throughout the test. A Baldwin, B-3M, snap-on extensometer was used for strain measurement to the offset yield stress, at which point it was removed and the remainder of the deflection to fracture was measured by crosshead travel.

Tests at 300 C were conducted in a salt bath controlled within ± 3 F. The test arrangement is shown in Figure 3.10. The molten salt is contained in a stainless-steel pot which has a double wall for heat insulation. The pot rides up and down on the lower grip support by means of a multiple O-ring seal. Thus, the pot is lifted into place after the specimen is inserted in the grips such that the specimen, the lower grips, and about 2 inches of the upper grips are immersed in the liquid media throughout the test. The use of molten salt provides several advantages: the high conductivity provides for good temperature control, specimen temperature can be controlled accurately and without appreciable gradients, the salt protects the specimen surface and prevents oxidation, and radiation contamination due to the spread of particulate matter during deformation and fracture is confined to the bath. Upon completion of the test, the pot is merely lowered in order to remove the broken pieces from the grips. Nearly as many elevated temperature tests per day can be conducted with this arrangement as at room-temperature. Strain measurements for the elevated tests were made using crosshead travel throughout the test. Thus, even though the strain data have been corrected for thermal expansion and elastic takeup in the loading members, they should be considered as approximate. In order to measure strain within the specimen gage, a hermetically-sealed, immersible strain transducer is being developed.

The tensile stress and uniform strain for the various conditions of cold work, test temperature, and working direction represented in Table 3.10 are plotted as a function of fast neutron flux in Figures 3.11, 3.12, and 3.13. As noted in Table 3.10, integrated flux values for quadrants 1 through 8 are based on nickel dosimeter measurements in Table 3.8. Integrated flux values for quadrants 13 through 16 are based on titanium dosimeter measurements.

Referring to Figure 3.11, it is seen that the rate of damage accumulation to tensile strength is greatly decreased beyond 1.5×10^{20} nvt exposure. This appearance tends to be deceptive when strength is plotted as a linear function of neutron flux. For example, such a scale effect can be noted by reference to Figure 3.14, where the same data indicates saturation at about 2×10^{19} nvt. Hence, it must be understood that any reference to saturation depends on the range of the flux scale.

TABLE 3.10

TENSILE PROPERTIES OF IRRADIATED ZIRCALOY-2
(Average of two tests)

Average of the tests:							
Working Direction	% Cold Work	Test Temp C	Strength (10 ³ psi) 0.2% Yield Tensile		% Strain Uniform Necking		Plastic Work (ft-lbs)
<u>Quadrant 1 5.1 x 10¹⁹ nvt (Ni)</u>							
TD*	0	RT	71.0	71.8	1.1	10.6	27.7
		300	36.9	37.1		7.3	9.0
	20	RT	89.4	92.2	1.2	3.0	10.6
		300	52.6	52.7		3.4	7.9
	40	RT	96.2	97.9	1.5	3.0	12.1
		300	60.0	62.2		4.6	12.0
<u>Quadrant 2 5.3 x 10¹⁹ nvt (Ni)</u>							
RD**	0	RT	69.0	77.1	4.4	5.7	24.5
		300	36.5	37.3		10.2	16.4
	20	RT	91.4	93.2	3.7	3.7	16.3
		300	53.2	53.3		3.9	9.3
	40	RT	93.2	96.6	2.3	4.6	20.4
		300	57.6	58.2		4.4	10.7
<u>Quadrant 3 1.2 x 10²⁰ nvt (Ni)</u>							
TD	0	RT	80.3	80.5	0.8	5.4	14.5
		300	47.6	47.6		4.8	8.0
	20	RT	92.4	92.2	0.9	3.1	10.0
		300	55.1	56.2		3.3	8.4
	40	RT	103.2	105.4	1.2	2.5	10.2
		300	63.2	63.8		3.2	11.0
<u>Quadrant 4 1.1 x 10²⁰ nvt</u>							
RD	0	RT	70.8	78.5	4.2	7.0	28.6
		300	38.2	38.2		8.8	13.2
	20	RT	93.3	95.3	1.7	4.0	15.9
		300	54.2	54.2		3.0	7.6
	40	RT	92.2	98.8	2.5	3.8	18.7
		300	59.3	59.6		3.8	10.3
<u>Quadrant 5 1.3 x 10²⁰ nvt (Ni)</u>							
TD	0	RT	82.0	82.2	1.0	7.7	20.2
		300	38.4	38.7		6.1	8.2
	20	RT	91.8	92.6	2.7	3.6	11.9
		300	52.7	52.8		3.6	9.9
	40	RT	97.4	99.2	1.2	2.7	10.3
		300	61.7	62.2		3.8	11.4
<u>Quadrant 6 1.3 x 10²⁰ nvt (Ni)</u>							
RD	0	RT	72.5	79.0	4.2	6.2	26.2
		300	39.4	39.5		9.0	12.8
	20	RT	90.0	93.0	2.5	4.0	18.5
		300	53.1	53.2		3.8	8.4
	40	RT	97.2	100.3	2.6	4.1	20.2
		300	58.0	58.7		4.3	11.7

* Transverse Direction

** Rolling Direction

TABLE 3.10(Cont.)

Working Direction	% Cold Work	Test Temp C	Strength (10 ³ psi)		% Strain		Plastic Work (ft-lbs)	
			0.2% Yield Tensile		Uniform	Necking		
<u>Quadrant 7 3.8 x 10²⁰ nvt (Ni)</u>								
TD	0	RT	96.5	96.6	0.8	3.8	11.7	
		300	45.4	45.4		4.6	7.8	
	20	RT	94.7	97.7	0.9	2.7	9.2	
		300	57.8	58.1		3.3	8.4	
	40	RT	105.8	107.0	1.2	2.4	9.8	
		300	67.0	67.2		3.4	10.4	
<u>Quadrant 8 4.0 x 10²⁰ nvt (Ni)</u>								
RD	0	RT	77.8	82.0	3.2	6.0	23.6	
		300	44.6	44.7		5.1	8.6	
	20	RT	92.2	93.4	1.2	2.7	9.0	
		300	58.4	58.4		3.2	8.6	
	40	RT	96.8	99.2	2.2	3.7	17.6	
		300	62.2	62.4		2.7	10.2	
<u>Quadrant 13 2.0 x 10²⁰ nvt (Ti)</u>								
TD	0	RT	76.0	76.4	0.9	10.6	27.7	
		300	40.0	40.4		5.9	8.0	
	20	RT	88.4	88.8	1.0	5.0	14.8	
		300	52.3	52.4		4.0	8.7	
	40	RT	96.0	97.2	1.2	3.8	13.6	
		300	61.4	62.0		4.1	10.8	
<u>Quadrant 14 3.0 x 10²⁰ nvt (Ti)</u>								
RD	0	RT	68.4	76.7	5.0	11.8	42.7	
		300	39.2	39.3		14.2	8.8	
	20	RT	82.0	87.2	3.3	5.4	23.4	
		300	- -	- -		- -	- -	
	40	RT	88.6	93.8	3.1	4.3	21.5	
		300	54.0	54.9		6.1	14.7	
<u>Quadrant 15 5.9 x 10²⁰ nvt (Ti)</u>								
TD	0	RT	91.8	91.9	0.8	4.2	12.0	
		300	47.6	47.6		4.1	7.8	
	20	RT	94.2	94.7	1.1	0.8	2.8	
		300	58.2	58.2		3.4	8.8	
	40	RT						
<u>Control</u>								
TD	0	RT	54.9	63.3	11.9	20.5	69.5	
		300	17.8	76.5		19.2	31.7	
	20	RT	76.7	81.2	2.7	8.2	28.0	
		300	46.7	48.8		5.1	12.8	
	40	RT	81.3	86.6	1.9	6.7	23.8	
		300	52.2	55.2		5.6	13.1	
	RD	0	RT	47.4	66.5	14.2	14.5	71.3
			300	18.7	31.6		20.3	37.2
		20	RT	74.1	82.8	4.4	9.5	42.8
			300	48.8	49.6		4.2	11.9
		40	RT	78.6	89.4	4.7	9.1	45.9
			300	55.9	58.4		5.9	13.3

FIGURE 3.10

Salt Bath Fixture For Conducting Elevated Temperature Tensile Tests

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FIGURE 3. 11

Effect of Neutron Irradiation on Annealed Zircaloy-2

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FIGURE 3.12

Effect of Neutron Irradiation on 20% Cold-Worked Zircaloy-2

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FIGURE 3.13

Effect of Neutron Irradiation on 40% Cold-Worked Zircaloy-2

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FIGURE 3.14

Effect of Irradiation and Irradiation Temperature
on Tensile Stress of Zircaloy-2

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The net radiation hardening of annealed Zircaloy-2 is influenced by direction of straining at room temperature. Referring to Figure 3.11 the increase in tensile strength for the transverse direction is 33,000 psi with 4×10^{20} nvt exposure compared to only 15,500 psi for the longitudinal direction. It can also be noted that the uniform strain for the transverse direction is less than for the longitudinal direction and for both directions is nearly independent of neutron exposure beyond 5×10^{19} nvt. These directional effects are attributed to the difference in the mode of deformation between the longitudinal and transverse directions. As has been previously established, the crystallographic texture promotes slip in the longitudinal direction and twinning in the transverse direction. Since slip predominates over twinning as the temperature of straining is increased, directional effects on tensile properties should be eliminated at elevated testing temperatures. Hence, at 300 C the net increase in tensile strength with irradiation is about the same for both directions.

Reference to Figures 3.12 and 3.13 shows that the net increase in tensile stress with neutron irradiation is reduced with prior cold work. For an exposure of 4×10^{20} nvt, the net change in tensile stress is 15,000; 10,600; and 9800 psi with 0, 20, and 40% cold work, respectively. The effect of direction on postirradiation tensile strength is also reduced with cold work, and for the cold worked state the uniform strain at room temperature is somewhat less in the longitudinal direction and slightly greater in the transverse direction than for the annealed state.

The reduction in net radiation hardening with cold work is substantiated by hardness measurements obtained from several quadrants of bend-test specimens irradiated in the ETR, G-7 loop to estimated fast-neutron exposures ranging from about 4 to 9×10^{19} nvt. Measurements were made using the 30 T-scale of a Kentrall motorized hardness tester. The data are listed in Table 3.11. Unirradiated, control specimens ranged in hardness from 78.8 to 83.0 for 0 to 40% cold work (corresponding to Rockwell-B values of 92.4 to 99.0). The scatter ranges of hardness data, are plotted as a function of cold work in Figure 3.15. These plots show that for an exposure of 4.5×10^{19} and 8×10^{19} nvt is about the same for each cold work level.

In plotting the tensile data of Figures 3.11, 3.12, and 3.13, it was noted that specimens from quadrants 13 through 15 exhibited strength values falling consistently and significantly below the trend curves. Since these quadrants were irradiated in a substantially higher flux position than quadrants 1 through 8 (upon which the trend curves are based) the question of temperature gradients due to gamma heating arose. Calculations of temperature drops due both to convection across the water film and heat generation in the specimens gave the maximum specimen temperatures listed in Table 3.12 for the quadrants in question. The bases for these calculations are given elsewhere⁽⁶⁾ and assume a gradient in gamma heating with loop position corresponding to the fast neutron flux.

Effect of Irradiation on Hardness
as a Function of Cold Work for Zircaloy-2

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TABLE 3.11

INDENTATION HARDNESS MEASUREMENTS OF IRRADIATED ZIRCALLOY-2

Quadrant	Cold Work %	Fast Neutron Flux, nv	Total Exposure, nvt	(Avg of 3 Readings)	
45	0	2.5×10^{13}	5.3×10^{19}	81.1	
	10			82.0	
	20			82.0	
	40			83.0	
46	0	2.1×10^{13}	4.4×10^{19}	81.1	
	10			82.0	
	20			83.0	
	40			83.4	
47	0	3.7×10^{13}	7.8×10^{19}	81.9	
	10			82.4	
	20			83.2	
	40			83.9	
48	0	3.7×10^{13}	7.8×10^{19}	81.9	
	10			83.1	
	20			83.2	
	40			84.1	
81	0	2.5×10^{13}	8.9×10^{14}	81.9	
	10			82.5	
	20			83.7	
	40			84.0	
82	0	2.1×10^{13}	7.5×10^{19}	81.7	
	10			82.7	
	20			83.0	
	40			83.6	
Control (Avg of 10 Readings)				Rockwell-B	
	0	0	0	92.4	78.8
	10			96.0	81.3
	20			97.6	82.4
	40			99.0	83.0

TABLE 3.12

MAXIMUM SPECIMEN TEMPERATURES FOR G-7 LOOP POSITIONS A, B, and D

Position	Quadrants	Gamma Heat Watts/gram	Water Temp F	Film Drop F	Source Heating F	Specimen Temp F
A	1, 2, 3, 4	4.4	528	3.4	5.9	537
B	5, 6, 7, 8	9.4	532	7.4	12.7	552
D	13, 14, 15	18.2	539	14.3	24.5	578

The apparent temperature differences shown in Table 3.12 are best reflected in the change in necking strain with neutron exposure shown in Figure 3.15. Contrary to past reports, the necking strain of Zircaloy-2 is effectively reduced with irradiation, even at 280 C (540 F). The plots of Figure 3.15 show that quadrants 13 and 14 give substantially higher values of necking strain for 0 and 20% cold work than the trends established by quadrants 1 through 8. Exceptions are noted for quadrant 15, which for 20% cold work shows a low value indicating a trend toward further embrittlement at an exposure beyond 5×10^{20} nvt. Furthermore, values of necking strain for 40% cold work for quadrants 13 and 14 fall reasonably close to the trend lines.

Irradiation recovery has been observed to occur for Zircaloy-2 in the range 300 to 350 C and substantial recovery occurs above 350 C, consequently it is not reasonable that the specimens in quadrants 13, 14, and 15 have undergone recovery corresponding to an irradiation temperature above 300 C. The greater loss in tensile strength and increase in necking strain for the annealed compared to cold worked states indicates that recovery occurs more readily in a crystal lattice initially strain free.

It is also apparent from the curves of Figure 3.16 that irradiation at about 280 C, corresponding to quadrants 1 through 8, results in little recovery. These plots show a net increase in strength with 280 C irradiation approximately equal to that for irradiation at ambient ETR water temperature (100 C). It should be noted that specimens for the ambient irradiations represent the as-prepared condition; whereas, specimens for the loop irradiations were given a prior autoclave treatment at 400 C. This accounts for the lower strengths for the loop control specimens. If the temperature calculations of Table 3.12 are reasonably correct, then 300 C represents a critical temperature above which a retarding of radiation damage accumulation occurs due to continuous damage recovery. In these tests cold work recovery during irradiation is somewhat unlikely (but not excluded) due to the stabilizing anneal at 400 C prior to irradiation.

The nearly linear curve plotted in Figure 3.16 for the transverse, annealed condition irradiated at 280 C suggests a different damage mechanism associated with twinning as compared to slip. However, directional effects for the cold worked states are decreased at the higher irradiation temperature. Metallographic examination may aid in explaining these differences in directional effects.

Fracture Mechanisms

The effect of neutron irradiation on the shape of stress-strain curves for metals is very pronounced not only in the shifts in stresses and strains but also in the nature of the yielding and necking regions of the curve. A marked decrease in uniform strain is commonly observed in metals with low neutron exposures, and for Zircaloy-2 a decrease from about 12 to 1% in uniform strain occurs with only about 5×10^{19} nvt exposure. At higher exposures, deformation beyond yielding is unstable, and the remaining

FIGURE 3.16

Effect of Irradiation on Necking Strain

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ductility prior to fracture consists primarily of necking. In the present program, fracture mechanisms and the effects of irradiation on deformation leading to fracture are being studied by both mechanical testing and metallographic investigations.

Tensile fractures of unirradiated Zircaloy-2 range from predominately shear to predominately fibrous (ductile tear), and include combinations of the two, giving what would appear to be a cup-cone fracture in a round specimen. Shear lips are present also in irradiated specimens of Zircaloy-2 as exhibited by the profile and oblique views of Figure 3.17, which represent a 20% cold-worked longitudinal specimen irradiated to 3.8×10^{20} nvt and tested at room temperature. The necking strain of this specimen was 2.7% compared to 9.5% for the unirradiated state. Thus, even though necking strain has been markedly reduced, the actual fracture appears ductile.

The stress-strain curves for transverse specimens present a much different mode of deformation within the necking region. The curves of Figure 3.18 represent three levels of cold work and two test temperatures (room temperature and 300 C) for the unirradiated condition. Whereas the 20 and 40% cold-worked specimens exhibit somewhat rounded curves beyond the maximum load at room temperature, the same materials exhibit linear curves at 300 C. A similar series of specimens irradiated to about 5×10^{19} nvt exhibit similar but more accentuated effects in Figure 3.19. In this series, the 20 and 40% cold-worked specimens exhibit linear curves within the necking region at room temperature. At 300 C, these curves first exhibit a marked drop in load, then a gradual unloading with strain, and finally a linear drop in load to fracture. The extremely low load values at fracture for these curves are of particular interest.

The curves of Figure 3.19 represent a mode of deformation to fracture which is characteristic of transverse Zircaloy-2 specimens at this and higher exposure levels. Stress-strain curves for irradiated longitudinal specimens are generally more rounded, similar to the room temperature curves of Figure 3.18.

Differences in the fracture appearances of transverse and longitudinal specimens, can be related to their respective stress-strain curves. Typical examples of these fractures are illustrated in Figure 3.20. The fracture for the transverse specimen is distinguished by a cupped appearance, having a rather smooth fracture face near the edges and a fibrous region near the center. Note the linear neck profile on the one side and the near absence of contractile strain in the short transverse direction. This fracture starts near the center and grows in a diamond outline to the edges of the specimen. Thus, both halves of the broken specimen exhibit a cup. The longitudinal specimen, on the other hand exhibits a nearly straight-across, fibrous fracture, with both sides of the neck rounded.

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FIGURE 3.17

Fracture Appearance of Zircaloy-2 Irradiated at 300 C to 4×10^{20} nvt
Exhibiting Ductile Fracture with Shear Lip
Profile at left 5X, oblique at right 10X.

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FIGURE 3.18
Stress-Strain Curves for Unirradiated Zircaloy-2
(Transverse Direction)

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FIGURE 3.19

Stress Strain Curves for Irradiated ($\sim 10^{19}$ nvt) Zircaloy-2
(Transverse Direction)

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FIGURE 3.20

Fracture Appearances of Irradiated Transverse (left) and Longitudinal (right) Tensile Specimens Tested at 300 C (Oblique views at 10X)

The proposed mechanism for these fractures are drawn schematically in Figure 3.21. The transverse specimen, which initially undergoes plastic strain by twinning, becomes unstable with small amounts of plastic deformation. In the irradiated condition instability is probably associated with the onset of slipping and is characterized by the marked drop-in-load yield point shown in Figure 3.19. Nearly all of the slip is confined to a relatively few slip bands and cracking initiates at the zone of overlap. This initial fracture is represented by the fibrous zone of Figure 3.20 and by the gradual unloading region of the stress-strain curves of Figure 3.19. After initial growth, however, the crack enters a region of general shear. At this point the fracture trace is displaced in the direction of shear in proportion to the amount of crack growth normal to the tensile axis. Concurrently, the neck profile at the surface follows a similar development. That is, the contractile strain is along the slip trace and is inversely proportional to the distance away from the neck. The drop-in-load during this step of the fracture is proportional to strain, and is associated with the linear portion of the stress strain curves of Figures 3.18 and 3.19.

The longitudinal specimens, on the other hand, undergo substantially more plastic strain prior to necking, and several intersecting slip systems are developed along the gage section upon instability. Hence, both crack initiation and growth occur under a triaxial state of stress. Should uniform stress for longitudinal specimens decrease sufficiently with irradiation and should radiation hardening bring about a marked drop-in-load yield point, a stress-strain curve and fracture appearance similar to those described for transverse specimens could develop.

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FIGURE 3.21

Schematic Representation of Necking Processes for Transverse and Longitudinal Specimens of Zircaloy-2

It is stressed for emphasis that the fractures discussed above are entirely ductile and are accompanied with considerable plastic shear. The development of a brittle fracture upon neutron irradiation for a structural material which is inherently notch insensitive is of vital interest. The low value of necking strain plotted in Figure 3.15 for quadrant 15 specimens, representing an exposure of 6×10^{20} nvt, indicates the approach to a brittle exposure range.

In order to study fractures of broken tensile specimens adequately, it is necessary to preserve the fracture trace during metallographic preparation. With ordinary mounting and polishing techniques the fracture surface becomes unresolved due to rounding at the mount interface. A common technique for supporting and highlighting the fracture trace is to electroplate the fracture surface with a metal of comparable hardness.

Nickel plating of Zircaloy-2 has been generally successful using methods simple enough for remote operation. However, these procedures require that any oxide film be first removed in order to provide a coherent plate. In the current investigation the specimens are not only given a black protective oxide coat by autoclaving prior to irradiation, but they develop oxide layers when removed from the salt bath used for elevated-temperature testing. Since no means were found for stripping the oxide chemically without attacking the metal and disturbing the fracture outline, a plating process which gives a coherent layer of nickel on zirconium oxide was developed.

The starting solution is a Watt's bath of the following composition:

Ni $\text{SO}_4 - 7 \text{H}_2\text{O}$	1.1 - 1.3 M
Ni $\text{Cl}_2 - 4 \text{H}_2\text{O}$	0.12 - 2.5 M
H_3BO_4	0.5 - 0.75 M

To this standard solution is added 5 ml glycerin which enhances the formation of a uniform, coherent film. Satisfactory coverings are obtained with a bath temperature of from 45 to 50 C and with a current density of 50 to 70 amp/ft². An example of a plated specimen in the as-polished condition is shown in Figure 3.22.

Examination of current literature shows that mechanisms of fracture in many metals are associated with formation of voids after initiation of necking.⁽⁷⁾ An investigation has been made to determine the effects of strain-rate, rolling direction, and mode of fracture on the frequency and distribution of voids formed during the room-temperature tensile testing of Zircaloy-2.

Several broken tensile specimens were mounted and ground on the broad face corresponding to the rolling plane. Final polishing was done ultrasonically. The specimens represented three cold work levels (0, 20, and 40%), two strain rates (crosshead speeds of 0.005 and 20 inch/min), and both the longitudinal and transverse directions with respect to rolling.

FIGURE 3. 22

A Nickel-Plated Tensile Specimen of Zircaloy-2
Having an Oxide Film Highlighting Fracture Trace(100X)

Four annealed specimens representing the two strain rates and both working directions are illustrated in Figure 3. 23. These specimens were given a B etch (45 parts H_2O , 45 parts HNO_3 , and 10 parts HF) which not only accentuated the voids but also delineated second phase precipitates and inclusions as well. From these sections, the general impression is given that void formation is more extensive for the longitudinal direction and for the slower strain rate. Since testing was performed at room temperature, mass transfer by self diffusion (vacancy migration) should not play a part in void growth. This is confirmed by the presence of voids at the highest strain rate employed. Any rate effects may be associated with the time delay of yielding or with stress relaxation during void growth.

The next step of the investigation consisted of employing quantitative metallography to determine the volume fraction of voids as a function of distance from the fracture surface. It was of primary interest to determine if void nucleation occurred concurrently with the advent of tensile instability as has been suggested. The point of instability on the broken specimen is the point at which the parallel region of the gage section, corresponding to the zone of uniform strain, becomes tangent to the region of necking.

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VOID FORMATION IN TENSILE FRACTURE OF ANNEALED ZR-2

FIGURE 3.23

Void Formation in Tensile Fracture of Annealed Zircaloy-2
(250X-B etch)

A point-counting technique was used, by which the specimens were scanned at 0.1 mm increments (both horizontally and vertically) through a microscope fitted with a focusing eyepiece. The eyepiece reticle was inscribed with a 25-point grid. Intuitively it can be seen that the probability of a point falling in the area corresponding to a void is equal to the fraction of the total area covered by voids. This relationship has been proven mathematically, (8) and it has also been shown that area fraction is equal to volume fraction (for constant isotopic shape).

An analysis of this type gave plots of volume fraction of voids versus distance from the fracture surface such as the one shown in Figure 3.24. From this plot it was possible to extrapolate the curve to the probable point of zero volume fraction, corresponding to the point of nucleation. It was found that this point was always well beyond the point of instability on the stress-strain curve and not too far removed from the fracture trace. It was concluded that void nucleation occurs at an advanced stage of necking and is associated with the triaxial state of stress.

An analysis such as the one above lends itself to statistical analysis, and the standard deviation has been shown by Gladman and Woodhead⁽⁹⁾ to follow a binomial distribution given by the relationship

$$T = \left[\frac{P(1-P)}{n} \right]^{1/2}$$

where T is the standard deviation. P is the proportion of second phase (voids in this case), and n is the number of trials taken (points counted). The data in Table 3.13 demonstrates that the standard deviation approaches zero. As a result, the curve of Figure 3.24 given only semiquantitative values of actual volume fraction, and indicates a maximum of about 1.4 volume % voids at the fracture.

Metallographic examinations were made of a wide variety of specimens to achieve an understanding of the nature of void nucleation and growth. The following conclusions were reached:

1. Voids were observed under all conditions of cold-work, strain rate, and working direction.
2. Voids were less frequent with greater amounts of cold-work and for the transverse specimens.
3. Voids were found to be aligned axially for longitudinal specimens but did not exhibit any particular alignment for transverse specimens. This can be observed in Figure 3.25.
4. Figure 3.25 also shows that voids are generally absent behind the shear regions of the fracture trace.

The above conclusions indicate that void nucleation is influenced by mechanical anisotropy. While metallographic examination failed to confirm definitely whether grain boundaries or second phase particles were the sites of origin, grain boundaries are excluded for two reasons: (1) deformation at the grain boundaries is expected to be negligible at room temperature, and (2) void nucleation at the boundaries would not explain differences in void frequency or alignment with working direction. Conversely, both of

FIGURE 3.24
Volume Fraction of Voids Versus Distance from Fracture

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3.45

HW-74378

FIGURE 3.25

Void Distribution in Longitudinal (left) and Transverse Specimen (right) of Zircaloy-2
(50X as-polished)

ALC-GE RICHARD WASH

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of the latter points can be explained by assuming void formation at either stringers or particles of second phase mechanically strung out by rolling. In the case of longitudinal specimens, the second phase particles are strung out along the strain axis; hence, voids formed at the interface between these particles and the parent matrix will remain aligned with continued deformation. In the case of transverse specimens, second-phase particles are strung out normal to the tensile axis. Consequently, relative motion of these particles in the direction of tensile strain breaks up the initial alignment of voids formed at these particles into a random array. It is proposed, therefore, that voids nucleate at noncoherent particles under a triaxial state of stress with the initial crack oriented normal to the tensile axis.

TABLE 3. 13

STANDARD DEVIATION OF VOID FRACTION DETERMINED
BY THE POINT-COUNT METHOD

Points, VP	Total Points, T	Volume Fraction, V_p/T	Standard Deviation, σ	% σ of V_p/T
9	702	0.0128	0.0043	34
10	739	0.0135	0.0043	32
9	740	0.0122	0.0040	33
8	736	0.0109	0.0038	35
7	748	0.0094	0.0036	38
4	750	0.0053	0.0027	51
3	775	0.0039	0.0022	56
3	765	0.0039	0.0022	56
2	765	0.0026	0.0018	64
2	795	0.0025	0.0017	68
1	819	0.0012	0.0012	100

It remains to establish the nature of the growth of voids once nucleated. Vacancy migration and agglomeration, although a likely mechanism of void growth during high-temperature creep and stress-to-rupture tests, is an unlikely mechanism at room temperature where self diffusion is negligible in Zircaloy-2. It is proposed, rather, that a slip process exactly analogous with the fracture mechanism associated with Figure 3.21 is active. Thus, cracks initially formed normal to the tensile axis are drawn out by slip into diamond shapes nearly equiaxed in dimensions. This diamond appearance is commonly observed and is frequent among the voids of Figure 3.25.

Voids play a major role in the final fracture, the trace of the fracture actually forming a bridge from void to void during the tearing operation. Furthermore, the frequency of voids is dependent upon the strain prior to instability and the constriction of the neck. Longitudinal specimens which form gradual necks involving many intersecting slip bands have longer void gradients away from the fracture surface than transverse specimens which have rather localized necks.

Thus the above mechanisms proposed for the nucleation and growth of voids appear to be consistent with all the metallographic observations made to date. The importance of microcrack formation and growth on the neutron damage structure will be the goal for future investigation.

Damage Mechanisms - J. J. Laidler, H. E. Kissinger, F. A. Smidt, T. K. Bierlein and B. Mastel

The objective of this program is to establish the nature of the interaction between defects present prior to irradiation and those produced by irradiation, and to investigate the possibility of neutralizing the effects of impurity atoms by chemical stabilization. Alloys of high purity iron with small amounts of carbon and nitrogen and alloys to which a chemical stabilizer such as titanium have been added will be studied.

A preliminary investigation of impurity effects in irradiated iron is continuing. Armco ingot iron, henceforth referred to as iron A, was used as the starting material. This material was decarburized and denitrogenized in a wet hydrogen atmosphere, designated iron B, and then treated in an ammonia-hydrogen atmosphere calculated to add 100 ppm nitrogen to the sample, designated iron C. Tensile testing of irons B and C irradiated to fast neutron exposures of 1×10^{17} , 5×10^{17} , 1×10^{18} , and 5×10^{18} nvt has been completed and the data are shown in Figure 3. 26. Examination of these results and those on iron A irradiated under identical conditions show the following trends:

1. The ultimate strength and fracture strength increase uniformly with dose while the ductility uniformly decreases with dose.
2. Unirradiated iron A showed a large drop in load yield point, iron B showed none and addition of nitrogen caused a small one to reappear in iron C. This yield point, however, disappeared at a dose of 1×10^{17} nvt and did not reappear at the highest exposures studied.
3. The proportional limit, maximum load, and fracture strain in irons B and C were approximately the same for all doses and were markedly lower than in iron A.

The results indicate that the levels of carbon and nitrogen content as well as neutron irradiation strongly influence the mechanical properties of iron. The residual impurities, both metallic and nonmetallic, remaining in the iron after the hydrogen treatment are believed to have masked some of the effects of the nitrogen additions. For this reason samples of electron beam zone refined iron are being prepared from Ferro Vac E iron for use

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FIGURE 3.26
Stress-Strain Curves for Iron B (top) and Iron C (bottom)

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in perfecting the fabrication, carbon and nitrogen addition, and the various physical measurements techniques necessary before proceeding to experiments on the high purity material purchased from Materials Research Corporation and Battelle Memorial Institute.

A meaningful comparison of data for various samples is not possible unless all samples possess a fine uniform grain size (which is not present in the high purity iron purchased). The fabrication techniques necessary to realize this objective are under development. Swaging has been found to be unsatisfactory because of a swirled, spiral flow of metal produced by the operation. Dies have been obtained for tests on cold drawing methods.

The existing resistivity apparatus has been redesigned to permit measurements on 20 mil wire samples at helium temperatures. Measurements on a wire sample such as this will provide a larger emf for measurement as well as a more easily regulated current requirement. Measurements at 4.2 K will eliminate the resistivity from phonon scattering and measure only that due to imperfection in the lattice. Thus the change in resistivity with irradiation should be several hundred %, rather than the few % found in room temperature measurements.

Swelling of Irradiated Fissionable Materials - R. D. Leggett

One of the major deterrents to the use of uranium metal as a nuclear fuel at elevated temperatures is the tendency for uranium to increase in volume due to the formation of voids, cracks and pores within the metal. The majority of the information that exists concerning the swelling of uranium was obtained from the irradiation of actual fuel elements or their prototypes. While this information is highly important, it has not provided sufficient insight into the basic mechanisms involved to allow either extrapolations or predictions to be made concerning the fuel element behavior under conditions somewhat different than those for which the data was obtained. Consequently, the present investigation is directed toward establishing an improved empirical and fundamental understanding of swelling in irradiated uranium. The influence of irradiation variables such as temperature, burnup, burnup rate, and metallurgical variables such as structure, composition, and sample geometry are currently being studied. The present irradiations are being conducted under conditions of low restraint and low external pressure but plans for the near future call for studying pressure as a variable too.

Irradiation Program - T. K. Bierlein, B. S. Kosut, R. D. Leggett and B. Mastel

The evaluation of the variables indicated above has required the development, construction and irradiation of three different types of capsules. These are as follows:

1. A general swelling capsule that contains bare uranium specimens on which the temperature is kept constant regardless of reactor operation.

2. A capsule that permits the irradiation of metallographic uranium specimens which are maintained at a constant temperature only when the reactor is operating.
3. A capsule that does not contain thermocouples and in which uranium is irradiated at approximately ambient reactor temperature.

Two general swelling capsules that attained their goal exposures have been opened, their specimens recovered, and the majority of post-irradiation examinations completed. A detailed report on the results of the postirradiation examination of these specimens is contained in a succeeding section under "Post Irradiation Examination". Two capsules previously irradiated have been shipped to Radiometallurgy for disassembly and specimen recovery. In addition, one capsule is in the reactor discharge basin and one is still being irradiated at a constant temperature of 575 C. A general swelling capsule containing a single 1/16-inch electrical heater in lieu of the standard 1/8-inch heater is approximately 90% complete. Two 1/16-inch heaters were originally installed in this capsule but one heater was damaged during capsule assembly. This capsule will be charged to obtain needed data on the irradiation effects of a fissionable material that has been thermally cycled from 400 C to 575 C. The 400 C temperature will be maintained when the reactor is down and the 575 C will be maintained on the specimens when the reactor is up.

An order for 1/8-inch heaters that was placed in September, 1961 has been cancelled due to the failure of the vendor to deliver the heating elements. Another order has been placed to deliver the required heaters in approximately 1 month. Meanwhile, three 1/8-inch heaters have been located and these will be used in the assembly of additional swelling capsules.

Two tests have been completed on a prototype ETR-MTR metallographic uranium capsule. The first test involved the thermal cycling of a uranium cylinder with one surface that has been metallographically precharacterized. This specimen was cycled 100 times from approximately 70 to 600 C. Examination of this specimen is continuing to determine the thermal cycling effects on the polished surface. This same capsule from which the specimen was taken has been reassembled. The cycled rod specimen was replaced by a split, hollow cylinder of uranium machined from the same stock as the specimens contained in Capsule 11, which was irradiated. One half of the cylinder was beta heat-treated while the other half was made from the as-extruded material. These specimens were subjected to the same thermal history as the irradiated specimens. A second objective of tests with the ETR-MTR prototype capsule was the determination of the heat transfer characteristics of fins with different thicknesses. Figure 3.27 shows the power input versus capsule internal temperature with fins 0.041-inch thick compared to fins that were 0.100-inch thick. It is interesting to note that the thickness of fins does not affect the heat transfer up to approximately 300 C. Apparently the actual contact area between the fuel and tube is the same. However, as the capsule's internal temperature is raised, the internal

chamber expands and increases the contact area between each fin and the outer tube. This larger heat transfer area with the thicker fins results in a lower capsule internal temperature with the same power input above the 300 C threshold point. This heat transfer data is needed as a guide for this type of capsule for the ETR-MTR irradiations. The capsule will be reassembled to determine the maximum power input that the heater will withstand.

FIGURE 3.27

Electrical Power Input to Capsule Heater-Watts

The reason for the malfunctioning of the over-temperature galvanometer used on the general swelling capsules at the reactor has been discovered and corrected. It was necessary to provide a voltage regulating device on the power supply to this instrument to compensate for the supply voltage variations during reactor operation. The reworked Minneapolis Honeywell instrumentation has arrived and is being installed in the instrument bays. The digital readout system has also been shipped and new thermocouple extension wire has been ordered. It is hoped that this new and reworked instrumentation will be installed, tested, and in operation shortly.

Postirradiation Examination - T. K. Bierlein, B. S. Kosut, R. D. Leggett, and B. Mastel

Several attempts were made to polish and etch the specimens irradiated to 0.4 at. % burnup in capsules 7 and 8 (respective control temperatures of 525 and 575 C) but no insight into the microstructure beyond that reported last quarter⁽¹⁰⁾ has been obtained. The replicas stripped from the sample surface are too radioactive to be handled in the normal laboratory so that no electron microscope examination was possible. A sample was submitted for burnup analysis by both cesium determination and ratio determination of U^{238} to U^{235} . The cesium analysis indicated a burnup of 0.08 at. % which, of course, is ridiculously low. It is possible that cesium was leached from the samples prior to analysis. This could occur by action of the NaK during the elevated temperature operation of the capsules. Leaching effects would be expected to be more serious with these 0.030-inch thick samples than with specimens with larger cross sections. The analysis by mass ratio determination is not yet complete.

Two general swelling capsules, 11 and 12, were opened in Radio-metallurgy, the specimens recovered, and the visual examination completed. Each capsule contained three pairs of samples, from longitudinally sectioned hollow tubing of high purity uranium, 0.5-inch diameter with a 0.030-inch wall thickness. Two of the sample pairs (A and B) were in the as-extruded condition while the third pair (C) was vacuum annealed at 730 C for 15 minutes followed by a quench into oil at room temperature (standard beta treatment) prior to irradiation.

Capsule 18 operated at a control temperature of 625 C except for a 1 minute temperature excursion that went to 730 C followed by 2 days at 450 C. This excursion occurred at about 2/3 of goal exposure when a controller malfunctioned. An over-temperature device shut off the heater power allowing the temperature to drop to 450 C, which was the steady state operating temperature of the capsule due only to the fission and gamma heat generation. Capsule 11 was not controlled, it operated between 400 and 450 C when the reactor was up, and at ambient temperature when the reactor was down. These two capsules were irradiated in a tandem arrangement in the same test tube to an estimated exposure of 0.16 at. % burnup in the specimens.

The side views of the specimen pairs (A, B, and C) from capsules 11 and 12 are indicated in Figures 3.28 and 3.30, respectively. It is obvious that the samples from capsule 12 (625 C) are in much better condition than are the samples from capsule 11 (~400 C). There are also obvious differences between specimens irradiated in the same capsule. In each case the sample that had been beta treated prior to irradiation (sample C) exhibited more damage than did the samples that were irradiated in the as-extruded condition (samples A and B). This damage is thought to be primarily a grain size effect. The grain size of the as-extruded material was 20 to 50 μ .

whereas after the beta treatment the grain size was 200 to 500 μ . The poor condition of capsule 11 samples is believed to be due to irradiation growth. This is supported both by the fact that striations appeared in the extrusion direction (due to preferred orientation of the grains) and by the fact that the specimen with large grain size developed severe surface bumping. Thus growth effects are still quite pronounced in this material at temperatures as high as 400 C and the condition of the samples from capsule 12 suggests that these effects have not completely disappeared even at 600 C. The condition of capsule 11 specimens is not explained solely by the fact that the samples cycled to ambient temperature every time the reactor shutdown. This is shown by the condition of the samples from a laboratory capsule illustrated in Figure 3.29. These samples, one as-extruded and the other vacuum annealed at 730 C for 15 minutes and quenched to room temperature, were given the same number of thermal cycles as the in-reactor samples received. The temperatures were duplicated but the time at temperature was appreciably shorter. The heat transfer characteristics of the laboratory capsule were quite similar to the in-reactor capsule so that the heating and cooling rates were closely simulated. As can be seen, no distortion or surface roughening visible to the unaided eye was detected. Therefore, the dimensional instability observed in capsule 11 samples is attributed to irradiation effects rather than thermal cycling effects. The samples both irradiated and unirradiated will be processed for metallography.

Three of the samples (one piece from each sample pair) from capsule 12 have been completely processed through metallography. Figure 3.31 shows the specimens mechanically mounted in their holders after grinding through the 600 grit polishing wheel. These cross-sectional views clearly indicate the differences in the macrodamage suffered by the specimens. One corner of sample 12A was badly swollen and, as seen in Figure 3.31a, is very porous. Severe porosity extended almost 1/3 of the way down the specimen. It is possible that a gas bubble may have formed inside the capsule adjacent to the specimen. This region of the sample would be insulated and would reach a much higher temperature of operation than was recorded by the thermocouple located about 120 degrees from this region. The other half of sample 12A has not yet been polished but from the macrophotographs, it too appears more swollen in the region of the sample close to the bad area shown in Figure 3.31a. The view of samples 12B and 12C shown in Figure 3.30 is consistent with the view shown in Figure 3.31.

Light micrographs of samples 12A and 12B are shown in Figure 3.32 and 3.33 at 75X, 250X and 750X. These samples consist of large grains with cracks outlining many of the boundaries. In sample 12B (Figure 3.33) a second much smaller grain boundary network due to the aligning of large pores is clearly seen. This smaller network may also be present in sample a (Figure 3.32) but it is much less distinct. This

Specimen 11A
As-Extruded

Specimen 11B
As-Extruded

Specimen 11C
Beta Treated

FIGURE 3. 28

Macroappearance of High Purity Uranium Specimens Irradiated to
0.16 at. % Burnup at about 400 C in Noncontrolled Capsule 11

3X

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End View

Side View

Oblique View

FIGURE 3.29

Uranium Specimens Thermally Cycled in a NaK Filled Laboratory Capsule to Simulate the Thermal Cycles of In-Reactor Capsule Number 11 (The notched sample was beta treated prior to the cycling while the unnotched sample was as-extruded.)

Specimen 12A
As-Extruded

Specimen 12B
As-Extruded

Specimen 12C
Beta Treated

FIGURE 3.30

Macroappearance of High Purity Uranium Specimens Irradiated to
0.16 at. % Burnup at a Control Temperature of 625 C in Capsule 12

3X

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(a) Specimen 12A
(Upstream from control
thermocouple; operated
at 590 C with reactor up;
at 575 C with reactor down)

(b) Specimen 12B
(Adjacent to control
thermocouple maintained
at 625 C regardless of
reactor up or down.)

(c) Specimen 12C
(Downstream from control
thermocouple; operated at
610 C with reactor up;
620 C with reactor down.)

FIGURE 3. 31

Macroappearance of the Rough Polished (600 grit) Cross Sections of the
Thin Walled, Hollow, Cylindrical Uranium Samples Irradiated
to 0.16 at. % Burnup in Capsule 12 Controlled at 625 C

5X

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3.58

HW-74378

(C)

750X

same area as (b)

(B)

250X

(A)

75X

FIGURE 3.32

Microstructural Appearance of As-Extruded Uranium

Specimen 12A Irradiated to 0.16 at. % Burnup

(This Specimen operated at 590 C when the reactor was up and 575 C when the reactor was down. The surface was metallographically polished and lightly cathodically vacuum etched. Note the faint network of pores at the boundaries of small prior grains and the large holes and cracks at the boundaries of the existing grains.)

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FIGURE 3.33

Microstructural Appearance of As-Extruded Uranium Specimen 12B
Irradiated at a Controlled Temperature of 625 C to 0.16 at. % Burnup
(The surface was metallographically polished and lightly cathodically vacuum etched. Note the fine network of pores precipitated preferentially at prior grain boundaries and the large network of cracks outlining existing grain boundaries.)

network of pores is identical in size with the original as-extruded grain size. It would appear that the inert fission gas collects into pores at grain boundaries during the initial stages of the irradiation at 625 C. Electron micrographs of negative replica from sample 12B (Figure 3.34) shows the segregation of large pores to these old boundaries, a profusion of small pores within these old grains and a pronounced depletion of small pores in the vicinity of the segregated large pores. The etching characteristics of the uranium matrix shows a continuity of crystal structure across depleted areas which proves conclusively that the segregation of large pores has not occurred at now existing boundaries. Electron micrographs from sample 12A (Figure 3.35) confirm the suspicions derived from the light micrographs concerning the segregation of large pores to prior grain boundaries. The pores in this specimen are much larger than was the case from sample 12B. Their size makes the metallography more difficult and probably accounts for the indistinct nature of the segregation. The large grains now present in these samples are believed to have formed as a result of the 1 minute temperature excursion into the beta-phase that occurred at about 0.1 at. % burnup. It is not clear when the cracks formed at the boundaries of the large grains but they could have formed either during specimen handling or during cooling of the capsule. It is significant that these cracks follow the existing boundaries rather than the old ones. This would indicate that the cracks formed quite late in the irradiation (after the existing network of grains was established) and also that the segregation of large pores to prior grain boundaries does not introduce a path of weakness for crack propagation. It is also significant that no depletion in small pores exists adjacent to cracks (Figure 3.36). This fact supports the concept that the cracks formed late in the irradiation.

The light and electron micrographs of sample 12C are presented in Figures 3.37 and 3.38. The holes in this sample are enormous as well as numerous so that it is almost impossible to see grain structure. Twin bands could be seen in a small portion of the sample (Figure 3.37a and Figure 3.37b) but the majority of the sample was a mass of holes (Figure 3.37). The explanation for the poor behavior of sample 12C seems to lie in the fact that this sample was heated into the beta region prior to irradiation. Metallographic study of as-extruded and beta-heat treated specimens of this material have revealed no differences other than the large increase in grain size due to the beta treatment. It is possible, of course, that a very finely distributed, beneficial second phase was dissolved during the heating into the beta region but if so it was not detected. In so far as the irradiation conditions are concerned, the slight variations in the irradiation history of the specimens 12B and 12C can hardly account for the differences in swelling that were observed. The thermal cycling was about the same in both specimens due to the fact that a temperature drop of 30 to 50 C occurred on all of the samples every time the reactor scrambled. This would be expected to overshadow the extra temperature cycles experienced by sample 12C (610 C with the reactor up and 625 C with the reactor down during reactor cycles. At this point, it looks as though the large grain size of sample 12C

FIGURE 3. 34

The Segregation of Large Pores to Prior Grain Boundaries in
As Extruded Uranium Specimen 12B, Irradiated to 0.16 at. % Burnup
at a Control Temperature of 625 C

(Note that the crystallographic etching of the background uranium
shows that this entire picture is within what is now a single grain)

5700X

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FIGURE 3.35

Microappearance of As-Extruded Uranium Specimen 12A

Irradiated to 0.16 at. % Burnup

This sample operated at 590 C with the reactor up and 575 C with the
the reactor down. 5500X

Note the patch of small pores surrounded by very large pores probably
situated at prior grain boundaries. Note also the twin band crossing the
band of large pores.

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FIGURE 3. 36

A Large Crack in As-Extruded Uranium Specimen 12B
Irradiated to 0.16 at.% Burnup at a Control Temperature of 625 C
Note that the fine pores exist immediately adjacent to the crack as well as
in the general matrix.
5700X

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FIGURE 3.37

Microstructural Appearance of Beta-Treated Uranium Specimen 12C
Irradiated to 0.16 at. % Burnup

This specimen operated at 610 C when the reactor was up and at 620 C when it was down. The surface was metallographically polished and lightly cathodically vacuum etched.

FIGURE 3.38

Microappearance of Beta-Treated Uranium Specimen 12C
Irradiated to 0.16 at.% Burnup

This sample operated at 610 C when the reactor was up and at 620 C when the reactor was down. Note the extremely large size and irregular nature of the holes. This picture is at lower magnification than the previous three electron micrographs.

3400X

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was responsible for its behavior. From the appearance of specimen 12B, it appears that gas tends to collect preferentially at grain boundaries. Hence, a large grain size material would have fewer sites for pore growth and, hence, each pore will tend to be larger. The mechanism by which the gas gets to the grain boundaries is still largely unknown and requires clarification. Perhaps this process is intimately associated with growth which becomes less severe as temperature of irradiation is increased.

The condition of the samples from capsules 11 and 12 is consistent with the condition of the samples from capsules 7 and 8⁽¹⁰⁾ which were irradiated at 525 and 575 C, respectively, to a burnup of 0.4 at. %. In both instances, the damage was much greater than had been anticipated. It will be recalled that 1 centimeter diameter spheres irradiated in capsules 4 and 5^(11, 12) at 575 to 600 C (capsule 4 had an excursion into the beta-phase at about 0.2 at. % burnup similar to capsule 12) to a burnup of 0.3 and 0.05 at. %, respectively, were in excellent condition after irradiation. No distortion or surface bumping had occurred. Cracks were present and pores were observed with electron microscopy but the swelling was small.

Several differences other than geometry exist between the spheres and hollow cylinders that make a comprehensive understanding of the observations impossible at this time. These differences, however, suggest areas of study that will lead to a better insight into the fundamental mechanisms evident in swelling. The hollow cylinders were machined from high purity ingot uranium that had been electron beam remelted and extruded in the high alpha-phase. The spheres were machined from a small cylindrical casting prepared by vacuum melting electrolytically purified uranium fines and vacuum pouring into a cold copper mold. The materials differed a little in impurity content but not drastically. The ingot uranium contained 20 to 50 ppm Be, 30 ppm Si, and 115 ppm Fe compared to 18 ppm Si, 45 ppm Fe and no detectable Be in the electrolytic U. On the other hand, the Ni content of the ingot U was only 20 ppm compared with 50 ppm for the electrolytic U and the carbon plus nitrogen content only 30 ppm compared with the estimated 200 ppm for the electrolytic U.

Thus, there are at least three variables, composition, fabrication, and geometry, that might account for the observed differences in swelling. The effect of geometry will be evaluated by irradiating various shapes of specimen machined from the same rod of high purity ingot uranium. Evaluating the other effects will be scheduled into the program at a future date.

Extensive metallography has been conducted on Zircaloy-2 clad coaxial U (enriched) - U (depleted) diffusion couples. These couples were irradiated at <300 C to two burnup levels. They have been annealed for 100 hours at 650, 700, 800, and 950 C. The outer shell of enriched uranium had sustained a burnup of 0.2 and 0.4 at. % respectively, whereas, the burnup in the core of the specimens was lower by a factor of 20. As illustrated in Figures 3.15 and 3.16 of the previous quarterly report.⁽¹⁰⁾ Annealing causes considerable change in the Zr-U interface due to diffusion. After the 800 C

anneal a needle-like second phase has formed within the original Zr-2 clad at a considerable distance from the gap which exists between the Zr-2 and the enriched uranium (Figure 3.39 and 3.40). After the 950 C anneal the needle-like structure in the clad has almost completely penetrated the clad. Extensive porosity has now developed in the shell enriched with U²³⁵, and many of these pores appear to be interconnected. No evidence of cracking at the U-U interface was found.

Figures 3.41 and 3.42 show the U-U interface in couples with two burnups (0.2 and 0.4 at. % in the U²³⁵ enriched shell, respectively) after a 100 hour anneal at 800 C. Annealing at these high temperatures is sufficient to cause very extensive gas pore formation in the enriched layers. After a 950 C anneal, the porosity is so great that electron microscopy in this zone is impossible. Annealing at the high temperatures of 700, 800, and 950 C causes gas pore formation in the core which sustained burnups of only 0.02 and 0.01 at. % during the two irradiation exposures. Figure 3.43 shows the type and extent of porosity after the three annealing treatments in core regions which sustained 0.02 at. % burnup. It is quite evident that during the 950 C anneal, the larger pores are aligned, probably along gamma grain boundaries.

From the metallographic observations which have been made on this series of "diffusion couples", it is concluded that no gas migration has occurred from the shell enriched with U²³⁵ into the core depleted in U²³⁵. There are several explanations for the observations:

1. Once gas atoms have agglomerated into pores in the enriched zone, there is little driving force (activity gradient) for gas migration into the core. In other words the solubility of gas under the conditions studied here in both the enriched layer and depleted core contains the same concentration of gas.
2. Gas migration into the core has occurred but it was not possible to produce the supersaturated solution necessary to form gas pores so that no indication of the migration was observed. This argument is somewhat weakened by the fact that a few pores were observed in the depleted core after annealing at the higher temperatures but the proposed situation is still possible.
3. Migration of gas atoms from the shell to the core involved distances of the same order of magnitude as the uncertainty associated with the location of the U-U interface which is estimated to be about 0.5 μ . This places an upper limit on the diffusion coefficient, D, which can be calculated if the relationship $D = X^2/2t$ is assumed. In the equation, X is diffusion distance in a random walk process and t is the time. If $X = 0.5 \mu$ (the uncertainty of the technique) and $t = 100$ hours (the annealing time), D is a maximum of about 3×10^{-15} cm²/sec. Reported values of D, as calculated from fission gas release studies,⁽¹³⁾ vary from about 10^{-14} to 10^{-10} cm²/sec at 950 C. If the lower values are realistic, the uncertainty in the present technique precludes placing a definite value on D.

Annealed 100 hrs at 800 C
75X

Annealed 100 hrs at 950 C
75X

FIGURE 3.39

Microstructural Changes in a Zircaloy-2-Clad Uranium-to-Uranium Couple
as a Result of Postirradiation Annealing
(Burnup in U²³⁵ enriched layer is 0.2 at.%)

Annealed 100 hr at 800 C
75X

Annealed 100 hr at 950 C
75X

FIGURE 3.40

Microstructural Changes in a Zircaloy-2-Clad Uranium-to-Uranium Diffusion Couple
as a Result of Postirradiation Annealing
(Burnup in U^{235} enriched layer is 0.4 at.%.)

FIGURE 3.41

Interface in U²³⁵ Enriched - U²³⁵ Depleted Diffusion Couples
(Irradiated to 0.2 at. % Burnup in the Enriched Region after
Annealing for 100 Hours at 800 C.)

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FIGURE 3.42

Interface in U^{235} Enriched - U^{235} Depleted Couples
(Irradiated to 0.4 at. % burnup in the enriched region
after annealing for 100 hours at 800 C.)
3200X

FIGURE 3.43

Formation of Gas Pores in the Core of U-U Diffusion Couples
after Irradiation and Subsequent Annealing
(The core which is depleted in U^{235} had sustained a burnup of 0.02 at. %)
3100X

Several possibilities for continuing and extending studies on U-U couples are currently being considered:

1. preparation of new U-U couples with sharp interfaces,
2. utilization of such couples during controlled temperature irradiations in the range 550 to 650 C,
3. utilization of available couples irradiated at low temperatures for ultra-high pressure postirradiation annealing studies, and
4. conducting annealing treatments on nonirradiated Zircaloy-2 clad for comparison with results obtained on irradiated and annealed couples.

Quantitative Metallography - T. K. Bierlein, R. T. DeHoff, B. S. Kosut, R. D. Leggett, and B. Mastel

Three dimensional pore size and frequency information obtained from electron micrographs of uranium surfaces are needed for characterizing the state of swelling in specimens with specific histories. Unfortunately there is no simple measure of how accurately the two dimensional distribution data portrays the true three-dimensional case. Three approaches to this problem are currently under investigation. The first approach, being conducted by Applied Mathematics personnel, involves choice of a mathematical functional form of the true distribution, such as log-normal with adjustable parameters, and then a determination of the two dimensional analog and comparison with the experimentally determined two dimensional data. If no agreement is obtained, some other functional form must be assumed and the inversion and comparisons must be repeated until good agreement is obtained. A second approach for obtaining reliable three-dimensional information is subjective or intuitive. All previous distribution data has been obtained from classifying pores with respect to:

1. their maximum interface diameter, in cases where pores were cut below their centers, and
2. their maximum diameter, in cases where pores were cut above their centers.

If replication is faithful or can be corrected for distortion during stripping, it is obvious that case (2) will yield true three-dimensional information; whereas, case (1) will not yield three-dimensional data. If it is assumed that most pores of the case (1) type have interface diameters very close to their true diameters, the distribution data derived from the two dimensional measurements must be quite similar to that of the true three dimensional situation. If this is the case, then classifying pores into a smaller number of size classes, with each class having an associated range of pore diameters, should result in smoothing of the distribution curves with little change in shape, center of gravity, etc. If this condition does not prevail and the curves shift, then some other size range per cell must be investigated. All previous

distribution data on 0.29 and 0.41 at. % specimens annealed at various temperatures has therefore been replotted. Each cell now contains four of the adjacent cells of previous plots, and therefore involve one fourth as many cells. Changes in the shapes of these 'new' curves are being determined.

A third approach which has been considered is to unfold the distribution data directly into three dimensional plots. This unfolding requires knowledge of where pores are cut with respect to their centers. A typical replica of etched uranium containing pores (0.20 at. % burnup sample which had been annealed 100 hours at 400 C) was shadowed at a 15 degree angle of incidence and processed for electron microscopy. Prior to shadowing, however, the replica was sprayed with polystyrene spheres of known and constant size. Since shadows of pores revealed by replicas are a function of the angle for shadowing and the height of the pore protrusion, it should be possible to determine the "true" volume of a pore from its shadow and pertinent diameter (maximum diameter or interface diameter). Unfortunately uranium specimens must be etched to remove flow metal due to polishing, and during this process develop surface relief. The extent of this relief is indicated in Figure 3.44, one of thirty micrographs from which shadows from 240 polystyrene spheres were measured. The analysis shows that shadows can be forshortened or elongated as a result of localized tilt in the specimen surface. Tilting of the specimen can be as much as 25 degrees. Accordingly, whenever the specimen surface is nonplanar, which is generally the case, pore diameters can not be derived from shadow lengths. Thus with existing techniques for sample preparation the shadows can be used only to assist in the identification of pores and not in their measurement.

NEUTRON DAMAGE TO METALS

This study is concerned with establishing the combined effect of neutron irradiation and impurity atom concentrations on the properties of specific metals, and developing from damage recovery data a model describing the state of damage in the metal.

Molybdenum - T. K. Bierlein, H. E. Kissinger, J. J. Laidler, and B. Mastel

Three capsules containing 59 polycrystalline 0.180-inch diameter tensile and 27 hardness specimens prepared from rod stock containing 25 ppm C (total impurity level < 250 ppm) have been submitted for irradiation to exposure levels of 10^{18} , 10^{19} , and 10^{20} nvt ($E > 1$ Mev). The specimens can be classified into three groups according to their preirradiation annealing state: 16 hours anneal at (1) 1050 C, (2) 1300 C, and (3) 1500 C. One of these capsules has already been received after an exposure of $\sim 10^{18}$ nvt ($E > 1$ Mev). In addition, two capsules containing 26 single crystal tensile specimens, 6 X-ray diffraction specimens, 2-length change specimens, and 2 stored energy specimens have also been received after a similar exposure. These capsules will be opened by the remotized lathe which is expected to be in operation within a month. Additional wire electrical resistivity specimens will be prepared for future irradiations.

FIGURE 3.44

Variation in Shadow Lengths Due to Localized Surface Variations
(15,000X)

AIM

Association for Information and Image Management

1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

Centimeter

Inches

MANUFACTURED TO AIM STANDARDS
BY APPLIED IMAGE, INC.

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Capsules containing polycrystalline foil specimens (0.003-inch thick) of Johnson-Matthey stock, and foils of Materials Research Corporation stock with nominal carbon contents of 10 to 20 ppm, 100 to 200 ppm, and 400 to 500 ppm, have been received after an exposure of $\sim 10^{18}$ nvt and $\sim 10^{19}$ nvt ($E > 1$ Mev). These specimens had all been annealed at 1850 C for 1/2 hour prior to irradiation. They will be processed for X-ray diffraction and electron microscope studies in the near future.

One of the techniques to be used in studying damage in molybdenum is calorimetry. Energy released by test specimens at particular temperatures will be measured in a differential calorimeter which has been constructed. In order that energy releases associated with "cold-work" damage in polycrystalline specimens can be recognized, experiments have been performed to introduce sufficient cold-work in nonirradiated molybdenum for subsequent energy release measurements. Reduction of 0.5-inch diameter rod stock by swaging was performed at temperatures of 600, 400, 300, and 200 C. The higher temperatures yielded sound metal, but only the outer layer of the rods showed a worked microstructure. At lower working temperatures the rods could be reduced in cross section only a small amount before cracking occurred. A cold worked microstructure and increased microhardness was achieved in 0.5-inch diameter rods swaged to 0.257-inch diameter at 300 C. Additional reduction in area resulted in cracking. Upsetting of molybdenum rods 0.5-inch diameter by four stages to 0.795-inch at a temperature of 550 C followed by swaging resulted in failure of the metal; microhardness of the upset metal had increased. From these experiments it is concluded that "cold-work" as such is difficult to introduce in the annealed, fine grained molybdenum stock available for use. A combination of upsetting operations and swaging should provide worked rod stock of the proper diameter for use as unirradiated test specimens for energy release studies.

The stress-axis rotations of unirradiated single-crystal tensile specimens have been studied by Laue asterism methods. During the quarter four low-carbon (10 to 20 ppm) specimens and one medium carbon (100 to 200 ppm) specimen have been completely analyzed. Stress-axis rotations were determined by making a series of back-reflection Laue photographs at intervals during the deformation using the fixture and technique described previously.⁽¹¹⁾ Stereographic projections were then constructed to locate precisely the tensile axis. Where asterism was observed, the elongated spots were plotted similarly on the stereographic projection. The results of these studies are shown in Figure 3.45.

All low-carbon crystals deformed by slip on $\{112\}$ planes in $\langle 111 \rangle$ directions. The first crystal, 15s4, had an initial orientation near $[011]$. The stress-axis rotated initially toward $[\bar{1}11]$, then reversed direction toward $[111]$. This could be only slip on $(\bar{2}1\bar{1})$ and $(\bar{2}11)$ planes. Crystal 4s5 behaved similarly, differing in that as the asterism extended toward $[\bar{1}11]$ and $[111]$ with successive photographs, the stress-axis moved toward $[011]$. This would result from concurrent slip on $(\bar{2}1\bar{1})$ and $(\bar{2}11)$, as before. Crystals 1s2 and 1-s3 behaved in quite a similar fashion.

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FIGURE 3.45

Projections of Observed Stress-Axis Movements for Single-Crystals of Molybdenum
(Arrows indicate axis movements and solid lines show direction of Laue asterism)

The one medium-carbon crystal examined had an initial orientation very near [001]. Early deformation stages indicated that {112} planes were responsible; however, just prior to failure the asterism changed direction abruptly. The change in direction corresponded to slip on (001) planes in the [111] direction. No low-carbon crystals with this particular orientation have been tested as yet. Whether this behavior is a function of carbon content or of initial orientation remains undetermined.

Thin foils of molybdenum for study by electron microscopy and X-ray diffraction techniques have been and are being irradiated between aluminum spacers in small aluminum capsules. A recent test in such a capsule instrumented with a thermocouple indicated that the maximum temperature of the foil specimens did not exceed 39 C during irradiation in an evacuated capsule. If helium gas is introduced in the capsule, the specimen temperature falls to 30 C under normal reactor operation conditions (normal power and flow of water coolant over the capsule).

Polycrystalline foil specimens of Johnson-Matthey molybdenum irradiated in the as-rolled state and in a stress relieved state (1 hour anneal at 830 C) to an exposure of $\sim 1 \times 10^{18}$ nvt ($E > 1$ Mev) have been annealed for 2 hours at temperatures of 200, 300, 400, 500 and 600 C. Annealing at the lower temperatures produces no change in the as-irradiated microstructure revealed by transmission electron microscopy. After a 500 or 600 C anneal, however, numerous very fine spots and loops (25 to 150 Å in diameter) are detected. The irradiated as-rolled and the stress-relieved specimens show no difference and appear identical. In order to confirm that clustering has occurred, irradiated specimens and nonirradiated control specimens were annealed simultaneously at 600 C for 2 hours and then thinned for electron microscopy. Spots and loops as shown in Figure 3.46 had formed only in the irradiated specimens. If the thickness of the thinned foils is assumed to be 1000 Å, then the concentration of these clustered defects is $\sim 1 \times 10^{16}$ /cc in foils which were annealed at 600 C for 2 hours.

By analogy with work reported on metals bombarded with ions and neutrons, the defects observed are most probably due to clustering of vacancies, interstitials or both. The identity of the defects will be determined if possible in the future. That these defects are observed in molybdenum after irradiation and annealing is of considerable interest, because a recent article⁽¹⁴⁾ implies that no defects have been observed in molybdenum by similar techniques. Whether Johnson conducted annealing studies has not yet been ascertained. Annealing of irradiated foils at various additional temperatures is continuing.

Field ion microscope emitters have been prepared by Linfield Research Institute personnel under contract DDR-146. The first emitter, prepared from a 1/8-inch diameter single crystal containing 200 ppm C became contaminated with zinc during operation in the microscope. A second emitter prepared from a 1/16-inch diameter rod containing 20 ppm C produced a diffuse unresolved pattern. Experiments on specimens submitted to Linfield are continuing.

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Annealed Control
(nonirradiated)

Annealed Specimen
(irradiated to 10^{18} nvt ($E > 1\text{MeV}$))

FIGURE 3.46

Evidence of Clustering of Defects Formed in Neutron Bombarded Molybdenum
During Postirradiation Annealing (2 hours, 600 C)
150,000X

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An electron beam zone refining unit has been put into operation. A single-crystal molybdenum rod, 1/8-inch diameter by 13 inches long, has been prepared by the floating zone technique. Experiments are currently being planned to grow crystals with particular orientations by a seeding process. Single-crystal tensile specimens currently under irradiation cover a variety of axial orientations, but do not include any examples of [111] rod axis orientations. If such crystals can be grown with a minimum of effort, they will be submitted for irradiation.

An evacuable platinum wound furnace tube capable of operation at temperatures as high as 1600 C has been fabricated, tested, and put into use. Modifications in the shapes of foil tensile specimens for testing in the electron microscope have been made. A die and punch required for preparing specimens has been designed. In addition, an electrolytic jet polisher for thinning specimens has been designed.

SUPPORTING STUDIES

Notch Sensitivity of Zircaloy-2 - K. R. Wheeler and J. C. Tobin

Mechanical properties of Zircaloy-2 cladding are being investigated. In support of these studies, notched tensile samples, taken from Zircaloy-2 rolled sheet and specimens of coextruded cladding, are being tested in the irradiated and unirradiated condition. Forty eight sheet samples, notched to varying depths, plus 12 unnotched coextruded cladding specimens have been exposed to approximately 1×10^{20} nvt ($E > 1$ Mev) in the ETR. Specimen temperature during irradiation was that of the bulk water coolant (120 F). Postirradiation remote testing of these samples using an Instron Tensile Machine is in progress. Tensile testing temperature is 280 C at three different strain rates imposed by crosshead motions of 0.002, 0.02, 0.20 in/min. Load versus extension curves are being compared with similar curves from unirradiated controls. Metallography of fracture areas will be undertaken.

REFERENCES

1. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, October, November, December, 1961, HW-72346. January 15, 1962. (SECRET)
2. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, January, February, March, 1962, HW-72347. April 16, 1962. (SECRET)
3. Shoeck, G. "Influence of Irradiation on Creep," J. Appl. Phys. 29: 112. 1958.
4. Weertman, J. "Steady-State Creep Through Dislocation Climb," J. Appl. Phys. 28: 362-364. 1957.
5. H. H. Yoshikawa. Personal Communication. September, 1962.
6. Laidler, J. J. Proposed Structural Materials Irradiation Program HW-70052RD. June 21, 1961.
7. Averback, B. L. et al. "Fracture," Int'l Conf. on the Atomic Mechanism of Fracture, Swampscott, Massachusetts. New York: John Wiley, 1959.
8. Hilliard, J. E. and J. W. Cahn. "An Evaluation of Procedures in Quantitative Metallography for Volume-Fraction Analysis," Trans. AIME, 221: 344-352. 1961.
9. Gladman, T. and J. H. Woodhead. "The Accuracy of Point Counting in Metallographic Investigations," J Iron Steel Inst. 194: 189-193. 1960.
10. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, April, May, June, 1962, HW-74377. July 20, 1962. (SECRET)
11. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, April, May, June, 1961, HW-70557. July 15, 1961. (SECRET)
12. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, July, August, September, 1961, HW-71422. October 13, 1961. (SECRET)
13. Gray, D. L. Release of Inert Gases from Irradiated Uranium (A Review of the Literature), HW-62639. January, 1960.
14. Johnson, A. A. Phil. Mag. August, 1962.

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METALLIC FUELS DEVELOPMENT

NEW FUELS DESIGN

Shaped Single Tube Element - D. P. O'Keefe

Fabrication was started on an experimental Zircaloy-2-clad, single-component, N-reactor fuel element which is shaped to allow for uranium swelling without tensile elongation of the cladding. Further details of this fabrication step are described in the Fabrication Development section of this report. A cross section of the coextrusion is shown in Figure 4.1. A length of $10\frac{1}{2}$ inches was selected for fuel elements to be tested in the ETR. This short length will help circumvent the problem of warp that has been observed as a result of the severe radial flux gradient in the M-3 test facility in the ETR. Six $10\frac{1}{2}$ -inch sections were cut from the extrusion for the fabrication of test elements, and the preparation of caps for end closure was started. The end closure will be a Be-Zr brazed Zircaloy-2 cap approximately 0.5 inch in thickness. The cap-uranium joint will be of a V-groove geometry to provide a transition in the amount of swelling and cladding strain adjacent to the cap. The Zircaloy-2 stock from which the caps are to be made was extruded in the fluted shape of the uranium. The small amount of cleanup of this extrusion by machining and etching which was necessary for the desired fit in the cladding was started.

FIGURE 4.1

Cross Section of Shaped Fuel Element
1.75X

Figure without Title Unclassified

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Capsule Irradiations - J. W. Weber

Zircaloy-2-clad uranium rods with selected uranium and cladding temperatures, exposures, and cladding thicknesses are being irradiated in NaK-filled capsules to provide supplementary information on cladding and fuel performance.

Cladding Performance. Localized thinning or "necking" and ultimate fracture of Zircaloy-2 cladding has occurred on a rod from each of two 7-rod clusters irradiated in pressurized water, and on several rods irradiated under a variety of conditions in NaK-filled capsules. The characteristics of the failures are similar and have been described in previous quarterlies of this series.

To study the effects of cladding thickness variations on the susceptibility to failure, a program of irradiating Zircaloy-2-clad uranium rods in NaK capsules with intentional striations in the cladding is being conducted. Examination is complete on samples from four capsules recently irradiated in a Hanford reactor to an average exposure of 0.2 at. % burnup. Table 4.1 lists the conditions of the cladding before irradiation, the irradiation conditions, and the results of measurements made on each sample. The cladding strains are determined from the maximum change in diameter. There are no indications of necking or plastic instability of the cladding as seen in some previously irradiated Zircaloy-2-clad rods, even up to the 3.6 percent cladding strain of Sample 244. Figure 4.2 shows Samples 244, 154, and 1124 which experienced the greatest cladding strains of the 12 samples in this test. The grid system of equal sized squares, chemically etched in the cladding surface, was used to determine if there was any nonuniform cladding strain. There was no observable change in size or shape of the grid squares on any of the samples. Thus, the cladding strain was uniform over the circumference on the unstriated samples. In the striated samples, the uniformity of strain is not so clear since the total strain may be divided between the seven striations. Samples from capsules 3U2-3 and 3U2-5 show an effect of striations on the maximum strain, that is, the thinner the cladding at the striations, the larger the strain. This effect is not observed on the other samples.

Metallographic examination of Sample 1124 with approximately 3.5 percent total cladding strain shows that some localized deformation is occurring in the region of the striation, as evidenced by the fact that the uranium directly under the seven striations has deformed outward from its normal circular contour. A comparison of the cladding-uranium interface for Sample 1124 and the unstriated Sample 244 are shown in Figure 4.3. However, the structure of the Zircaloy-2 under the striations shows only a few twins and no elongation of grains. It is not apparent that all the deformation is taking place at the striations. The observed lack of localized deformation at high cladding strain is in agreement with observations on other test elements irradiated at cladding temperatures of 350 C and above as reported in the previous quarterly of this series.

244

154

1124

FIGURE 4.2

Samples from Capsule 3A2-2 After Irradiation
(Striations in Samples 154 and 1124 were machined in the cladding before irradiation to determine the effect of cladding thickness variations on cladding stability.)

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Sample 244
No Striation

Sample 1124
32% Striation

FIGURE 4.3

Comparison of Postirradiation Interface
of Striated and Unstriated Samples Showing the Movement
of the Uranium Outward from Its Normal Contour at the Striations

Unclassified Figure

Fabrication of components for a second irradiation test of Zircaloy-2 clad uranium rods with nonuniform cladding thickness has continued. All phases of assembly of the fuel elements is complete and they have been loaded into the capsules. Approximately 75 percent of the capsules have been filled with NaK. Assembly of several capsules to measure the uranium temperature is proceeding but problems on sealing thermocouples into the capsules are being encountered. A Zircaloy-stainless steel eutectic braze joint that was to be used has proven unsatisfactory in other recent applications of in-reactor temperature measurements. Another procedure using a mechanical seal is being considered.

Fuel Performance. Density decreases determined for the uranium in the four capsule tests are given in Table 4.1 along with the volume average uranium temperature and burnup. The swelling observed in the unalloyed fuel samples of this test is much lower than measured in previous uranium irradiations, and there is no apparent reason for this. The greater swelling of the uranium-2 wt % Zircaloy samples is consistent with other irradiation data.

Fuel-Fouling Detector - L. E. Kuhlken

A new design for the fuel-fouling detector (crud probe) for N-reactor is being developed. This design will still use a 1-inch-diameter, Zircaloy-2 clad, 1.6 percent enriched uranium rod with a 1/2-inch Zircaloy-2 core. The thermocouples, however, will be Zircaloy-2 sheathed and brazed into

TABLE 4.1

DENSITY DECREASES, VOLUME AVERAGE URANIUM TEMPERATURE, AND BURNUP

Capsule	Fuel Sample	Fuel Composition	Cladding Thickness, inch	Maximum Depth of Striation	Calculated Average Cladding Temperature, C	Total Cladding Strain, %	Calculated Volume Average Uranium Temperature, C	at. % Burnup	% Density Decrease
303-2	TD45	Unalloyed Uranium	0.025	0	438	1.36	580	0.18	2.7
	TD42			0.004	438	1.0	580	0.17	2.7
	TD36			0.008	438	1.3	580	0.18	3.3
302-3	BB212	Unalloyed Uranium	0.025	0	413	1.0	564	0.20	3.5
	BB31			0.004	413	1.45	564	0.20	2.3
	BB33			0.008	413	1.54	564	0.20	3.0
302-5	TC23	Unalloyed Uranium	0.025	0	423	1.0	578	0.20	2.3
	TC25			0.006	423	1.2	578	0.19	3.1
	TC27			0.010	423	1.53	578	0.21	3.3
3A2-2	244	U-2 wt % Zircaloy	0.025	0	402	3.6	518	0.20	7.7
	154			0.004	402	3.26	518	0.19	7.7
	1124			0.008	402	3.46	518	0.20	8.4

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4.6

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the assembly using beryllium-zirconium braze alloy, rather than stainless steel clad couples which were sealed into the assembly using a self-brazed stainless steel-to-Zircaloy-2 transition piece. Figure 4.4 is a section through a brazed assembly showing the excellent bonding of the Zircaloy-2 clad thermocouple to the cap and the Zircaloy-2 core of the test element.

FIGURE 4.4

Section Through a Thermocoupled Fuel Rod
Formed by Beryllium-Zircaloy Brazing
of the Zircaloy-2-Clad Thermocouple
to the Zircaloy-2 Cap and Core

Neg. No. 5W-65-1565

2X

Figure Without Title Unclassified

A low heat flux power probe is being developed for use in conjunction with the fuel-fouling detector. The initial design of the power probe utilized a 1-inch-diameter rod with a 1/2-inch core of natural uranium and

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0.25-inch-thick cladding. Test material has been successfully extruded and beta heat treated without failure of the uranium-Zircaloy-2 bond.

N-Outer Fuel Element Support - J. P. Pilger

Testing of the N-outer fuel element support-process tube compatibility has progressed through the following stages:

1. accumulated distance by oscillating one element back and forth over the same tube surface,⁽¹⁾
2. charging elements through short process tubes under static water condition, and
3. charging elements through long process tubes under static and flowing water conditions.

In the accumulated distance tests one fuel element was oscillated back and forth through a distance of 3 feet until a total wear distance of 1002 feet was accumulated. The element did not rotate; therefore, the supports traveled over the same tube surface during the entire test. Periodic and final examination of the tube surface indicated no damage to the tube. Support surfaces were in equally good condition. Water was static during the above tests.

Two tests comprise the second category of experiments. In the first test, 22 fuel elements were charged (singly) through a 5-foot-long section of process tubing until 135 charging passes had been completed. Examination of the tube and supports revealed no damage to either surface. The second test differed from the first in that the fuel elements were column charged in a short column of three elements. The aforementioned 22 fuel elements completed 92 charging passes without precipitating any damage to the process tube or the supports. The water was static during these tests, and the same section of process tubing was used for all tests described thus far.

A 17-foot-long section of process tubing was used to "shakedown" the facility and equipment for longer tube tests. Seven elements, five under static water conditions and two under a flow rate of 30 gpm, were charged through this tube with no evidence of damage. Cleanliness was considered a major factor, and becomes more difficult to maintain as tube lengths increase and facilities become more complex. A 52 foot 8 inch-long process tube and facility were cleaned in the same manner as the 17-foot tube (1-hour cold water detergent soak, flush rinse at 30 gpm, high velocity water jet impinging on tube wall full length), and set up to charge at a 30 gpm flow rate. The first fuel element scratched the tube on one side, i.e., one set of supports. A single scratch started approximately 33 feet 8 inches from the charge end of the tube. At about 35 feet, the scratch divided into two scratches. At this point the second support apparently crossed the original scratch. The two scratches continued in a narrow criss-cross pattern to the end of the tube. The scratches are about 0.0002 to 0.0006-inch deep, and 0.002 to 0.004-inch wide. The other set of supports passed directly

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over a projection on the tube wall (visible in the borescope) which severely gouged the supports. The projection remained in place and no scratching was initiated by the damaged supports. The tube was rotated approximately 90 degrees, recleaned with the jet, and testing resumed. Seventeen fuel elements were charged through the tube, and borescopic examination revealed no evidence of damage. The monofilament drawline broke after charging the 19th element approximately 10 to 15 feet. It was necessary to push the fuel element out of the process tube with the cleaning device. The cleaning probe had been idle for approximately 10 days prior to using it for this purpose. A probability exists that corrosion scale was flushed from the cleaning device during this operation. The element supports scratched the process tube from 19 feet to the discharge end.

The tube was recleaned and rotated to expose unscratched areas to the fuel supports. Music wire, 0.030-inch-diameter, was substituted for the monofilament line. Element 21 induced a very light scratch in the last 5 feet of the tube. Element 22 induced a scratch in a butt weld joint 18 feet from the charge end of the tube. Testing was suspended at this point. No definite mechanism has been established for initiation of scratching. It appears, however, that a minute particle (sand, oxide flake, etc.) imbedded in the relatively soft support surface would produce the type of behavior witnessed in these tests. Cleanliness was relaxed as the tests were progressing satisfactorily. This factor may account for the scratching produced in the last two tests.

Fuel Element Support Evaluation - K. R. Merckx and R. G. Wheeler

The N-fuel element supports must withstand high loads and impact, have flexibility, and limit fuel element vibration. To develop a support capable of satisfying these requirements in the limited space of a fuel element coolant channel, mechanical tests, stress analysis and vibration analysis studies of various support geometries have been undertaken. To determine the desired flexibility requirements needed to limit fuel vibrations, a program was written for a two degree-of-freedom, damped-mass system subjected to ground vibrations. The system was formulated to represent tube-in-tube fuel elements supported by flexible supports in a process tube undergoing vibrations. The schematic diagram for the fuel element vibration system and the differential equations governing the system are shown in Figure 4.5. The effects of various spring constants and fluid damping have been investigated. The change of natural frequencies for a representative outer support flexibility and various inner supports is shown in Figure 4.6. The maximum amplitude magnification for various dampings for a representative support flexibility is shown in Figure 4.7. Shaker table tests on the inner fuel tube support system produced an amplification factor of 7 at the resonant frequency of supports having a spring constant of 15,000 pounds per inch. The addition of water to the system produced no significant change in the damping. On the basis of available data, the damping is estimated to be 3 lb-sec/in. Damping present when the whole assembly is submerged in water will be measured when the new

OUTER FUEL TUBE EQUILIBRIUM

$$\begin{array}{l}
 \text{Left side forces (pointing left):} \\
 DX * (\dot{x} - \dot{x}_g) \\
 WX * \ddot{x} \\
 SX * (x - x_g) \\
 \text{Right side forces (pointing right):} \\
 DY * (\dot{y} - \dot{x}) \\
 SY * (y - x)
 \end{array}$$

$$WX * \ddot{x} + DX * (\dot{x} - \dot{x}_g) + SX * (x - x_g) = DY * (\dot{y} - \dot{x}) + SY * (y - x)$$

INNER FUEL TUBE EQUILIBRIUM

$$\begin{array}{l}
 \text{Left side forces (pointing left):} \\
 DY * (\dot{y} - \dot{x}) \\
 WY * \ddot{y} \\
 SY * (y - x) \\
 \text{Equation:} \\
 WY * \ddot{y} + DY * (\dot{y} - \dot{x}) + SY * (y - x) = 0
 \end{array}$$

FIGURE 4.5

Schematic Diagram for Fuel Element Vibrational System

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FIGURE 4.6

Calculated Natural Frequencies of Fuel Assembly
As a Function of Spring Constant of Inner Fuel Tube Supports
(From the above plot it is apparent that the lower natural frequency can not be increased appreciably by increasing the spring constant of the inner supports alone.)

FIGURE 4.7

Vibration Amplitude of Inner Supports
Expressed As a Factor
of the Ground (Process Tube) Amplitude

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design (stronger) supports are available. If the vibrations are due to limited acceleration or displacements of the piping, high natural frequencies of the damped systems will yield low maximum amplitudes.

Collapse strengths higher than existing in present support systems is desired. Maximum-energy absorption in the supports is also desired. These requirements suggested that mechanical analyses of the load-deflection and load-stress behavior of various geometries be studied. The actual geometries of supports require small radii-to-thickness ratios; hence, thin curved beam theory is not appropriate. A computer program has been written that accounts for normal, shear, and bending stresses (thick beam behavior). The shape of the support is described in terms of equal angle increments but various external radii. Bending stresses, forces, shears, displacements, and moments are found for each radial increment. The program is based on a complementary strain energy approach which produces influence functions for the support structure. Calculations using this program are not yet available.

FABRICATION DEVELOPMENT

Self-Brazing Closure for Zircaloy-Clad Fuel Elements - E. A. Smith

Capitalizing on the developments described in the last period report, several closures were fabricated employing the familiar V-groove contour cap, with Cu-Ni-Zr "tinned" faying surface. These closures were found to be uniformly sound metallurgically. They were, however, contaminated with uranium which was extruded during processing into the annulus between cap and jacket wall. This, of course, is undesirable from a corrosion standpoint, and efforts were turned to developing methods for preventing the flow of uranium into this annulus.

Zircaloy caps were prepared having a 0.075-inch tapered skirt on the faying face around the central hole and around the periphery (Figure 4.8).

Faces "tinned"
with Zr-Cu-Ni Alloy

FIGURE 4.8

Longitudinal Section of Zircaloy Cap

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These caps were plated with 1/2 mil of copper and overlain with 1/4 inch of nickel on the surfaces of the faying face, the skirts and to within 1/16 inch of the rims of the cap, after which they were "tinned" by induction heating in vacuo. These caps were then electron beam welded into recesses of matching contour at the ends of the fuel elements. Following this, they were pressed with resistance-generated heat in the Sciaky heavy-duty spot welder. Ultrasonic testing showed a high incidence of sound bonding at the Zircaloy/uranium interfaces, but somewhat incomplete bonding on some of the sidewalls. On the basis of previous success, this condition presumably may be corrected by better adjustment of pressing conditions.

One of the completely sound closures fabricated by this procedure was sectioned longitudinally to permit examination of the metal structures. This study indicates that the heat-affected zone brought about by the Sciaky pressing extends only a short distance ($\sim 1/16$ inch) into the uranium below the braze layer (Figure 4.9). This is significant in that it presumably

FIGURE 4.9

Zone at Interface Between Uranium and Zircaloy
on "Self-brazed" Closure Showing the Brevity
of the Heat-Affected Zone

(Recrystallized area in uranium extends less than
1/16 inch below interface.) 50X

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would permit β -heat-treating the fuel element stock before beginning the closure processing. Thus, structure quality screening tests may be applied to the bare uranium at the element ends before recessing. In addition, the processing operation would be somewhat simplified by eliminating one or more vapor-blasting and bright etching steps.

Brazed Closure Development - R. N. Johnson

Two N-inner fuel elements have been braze-closed by placing a 0.010-inch copper wafer under each end cap and forming the Cu-Zr eutectic braze by diffusion at 1000 C using induction heating. One of the elements was left in the as-brazed condition and one was electron-beam welded over the brazed closure. The elements were then autoclaved for 100 hours in 300 C water to test the corrosion resistance of the braze and of the braze-contaminated weld. The braze-contaminated welds showed some discoloration but no severe corrosion. The as-brazed closures were corroded down about 0.020 inch, showing that the braze alloy will corrode but has sufficient resistance to corrosion to limit the penetration in case of a primary weld fracture.

Melting point studies on a series of arc-melted alloys have shown the existence of a ternary eutectic at approximately 80 wt % Cu-10 wt % Ni which melts at about 850 C. Successful brazes have been made on N-inner elements using this alloy, but the alloy appears to be very brittle and may be too brittle for reactor conditions.

Projection-Welded Brazed Closure - W. I. Steinkamp

The projection-welded brazed closure has been pursued as an alternate closure method for NPR fuel. It consists of projection welding a cap to the cladding of an element and then, by a combination of heat, preplaced brazing alloy, cap shape, and force, brazing the cap to the uranium core of the element. The fuel element is contained during the process by a ceramic cylinder and a paper-clad steel center plug. The paper-clad steel plug has apparently eliminated the previous problem of arc burns on the ID cladding. The concave-shaped cap section has prevented burning of the ID welds.

Hot-Headed Closure Studies - W. F. Brown

A projection-welded and bonded end closure currently under study is an attempt to take the "Hot-Headed" Projection-Welded End Closure a step further. This latter method consisted of projection welding a Zircaloy-2 end cap with "ring" projections to the partially clad end of a full element. The partial cladding of the ends was accomplished by back extruding the fuel element so that the cladding flowed over the major portion of the fuel ends. Although this method provided a strong projection-welded end closure, there were areas between the end cap and the fuel element which were not bonded.

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The method under study involves the bonding of the unwelded areas by preplacement of a suitable material within the ring projection on the cap. The cap is projection welded to the end of the fuel element, thus encapsulating the bonding material. Additional heat is applied to the end of the element to effect bonding in the unwelded areas. Resistance welding equipment is being used to supply heat and pressure to accomplish this duplex joining method.

To date, experimental work indicates that preplaced materials can be encapsulated by projection welding. Although the conditions have not been determined for making a successful duplex joint, the projection welds appear to be satisfactory and some samples show that bonding can be achieved at the uranium and Zircaloy-2 cap interface in the areas between the projection welds.

Some of the problems currently being studied in an attempt to obtain a successful duplex joining method are

1. welding projection design and number of projection rings,
2. thickness of interface bonding materials,
3. methods of preplacing bonding materials,
4. surface conditions at the joint interface, and
5. projection welding and postheating condition to effect bonding.

Fluted Single Tube Element - G. S. Allison

Coextrusion of the eight-fluted single tube element material appears completely successful. The cross-sectional dimensions vary within less than 5 percent of the desired, and the cladding is bonded to the core with a smooth interface. As reported last quarter, the die cone was not modified to feed the high points of the extrusion; however, the extrusion filled the die properly. The cap stock extrusion discussed below did not fill the die at the high points by about 0.010 inch on the diameter. This is due to the increased stiffness of the cap stock material, but indicates that the situation is borderline and the die should be modified by addition of feeder grooves to the cone.

Before extruding the fuel, a composite billet of the same design was extruded with Zircaloy-2 substituted for the uranium core. The Zircaloy-2 core was oxidized chemically to prevent bonding to the Zircaloy-2 cladding. It was intended that the cladding be stripped from Zircaloy-2 core after extrusion to furnish cap stock for the fuel elements. This failed because the cladding bonded to the core sufficiently to prevent stripping; in fact, the bond could be termed fair to good. Subsequent treatment of Zircaloy-2 specimens, oxidized both chemically and by autoclaving in vacuum at the temperature of billet preheat (1250 F), indicated that the disappearance of the oxide film permitted the bonding. The cap stock required for the fuel element is being extruded to shape and size.

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Fabrication of Zircaloy-2 Strip for Fuel Tube Support Applications -
D. P. O'Keefe, R. G. Wheeler, and F. B. Quinlan

The performance capability of inner fuel tube supports is currently limited by the bend ductility of available Zircaloy-2 strip. All attempts to alter the support design and still meet design criteria have failed to circumvent the need for improved bend ductility. To meet this need, a program was initiated to develop a process for making Zircaloy-2 strip supports under conditions that would produce sharp bends without damage to the material and to develop a test procedure for evaluating Zircaloy-2 strip quality and finished support quality. A discussion of the progress made in all three phases of this program follows.

Phase I - Material Development. Although the outer fiber strain produced in sheet bent to a radius of $1.7 T$ is considerably less than the tensile elongation exhibited by conventionally rolled Zircaloy-2 sheet, it is not possible to bend such sheet to this radius without fracture. Presumably, the biaxial stress condition that develops as the bend is formed limits the amount of longitudinal strain. The degree of biaxiality that is produced in a sharp bend is evidenced by the extent of anticlastic bending, which varies with the crystallographic texture of the sheet. Measurements of the extent of anticlastic bending were found to be related to the ratios of principle strains formed in tensile specimens from sheet with various fabrication histories. In general, sheet material which exhibits a large strain in the short transverse direction of a tensile specimen will also exhibit a high bend ductility and a comparatively small amount of anticlastic bending. By examining the parameters of strain ratios and extent of anticlastic bending along with the minimum achievable bend radius, it was possible to establish the dependence of bend ductility on crystallographic texture.

The most favorable textures for high bend ductility were found to exist in extruded sheet and in extruded and drawn material. Strip cut normal to the rolling plane in both the transverse and longitudinal direction of rolled plate also were textured favorably for good bend ductility. It, therefore, appeared to be possible to base a fabrication method for ductile sheet on schedules involving extrusion for primary reduction of the ingot, or possibly rolling employing properly sequenced cross-rolling procedures.

In the process of evaluating numerous fabrication schedules based on the above concepts, there was developed empirical evidence that control of annealing temperatures and working sequence would provide some control over sheet texture. Hot rolling appeared to alter the favorable texture of extruded material; whereas, extruded tubes could be taken through a considerable reduction by cold rolling with 600 C intermediate anneals to produce a ductile sheet product. Consequently, the procedure adopted in the most recently evaluated schedules was to keep the working and annealing temperatures low for the reduction stages from about 0.5 inch in thickness to the final 0.038-inch sheet. All annealing was done in air. The final cleanup of the sheet was done by grit blasting to remove the oxide and etching to the desired thickness of 0.035 to 0.036 inch.

Three fabrication procedures were found which would produce sheet or strip of sufficient ductility to make the inner supports. This was demonstrated by fabricating several hundred supports from material produced by the following schedules:

- Schedule 18 - Extrude to 0.4-inch wall tube, then cold roll to 0.038-inch with intermediate and final anneals at 600 C.
- Schedule 22 - Extrude to 0.350-inch-diameter rod, then draw cold through a roller Turkshead to 0.257 x 0.038-inch strip with intermediate and final anneals at 600 C.
- Schedule 21 - Hot roll (8 phase) to 2-inch plate which is β -quenched, followed by hot rolling in the direction transverse to the ingot axis in the high alpha temperature region (750 C) to 0.5-inch plate, then cold roll to 0.038 inch with intermediate and final anneals at 600 C.

Of the three fabrication procedures, the strip produced by Turkshead rolling had the best bend properties. No further development work is planned for this schedule. The other two schedules, which allow for the production of wider sheet, will be investigated further, particularly in regard to optimum working and annealing temperatures. The effect of working temperature on texture and the effect of final grain size on bend ductility are to be determined.

Phase II - Support Forming Equipment. Equipment for forming demonstration quantities of supports was made and is now ready for testing. One of the two machines provides for momentary resistance heating of the strip during the forming in order to take advantage of the higher bend ductility of Zircaloy-2 strip at elevated temperatures. The other machine is a two-step die that crowns and cuts the strip to length in the first step then simultaneously forms all four bends around pins in the second step.

Phase III - Strip and Support Quality Tests. To evaluate the bend ductility of strip produced by the numerous experimental fabrication schedules, a rapid bend ductility test was needed. The following test was the most satisfactory of all the tests tried: Fold the strip into a "U" shape and place the strip between the smooth jaws of a vise to complete the bending. Limit the amount of bend given the strip by placing a shim of steel inside the "U" bend. Examine the bend at 20X for cracks. If none appear, continue the bending using a progressively thinner shim until cracks appear. One half the thickness of the thinnest shim the strip will bend over without showing cracks is taken as the minimum bend radii of the strip. The shim thickness is expressed in sheet thickness units. Some of the best available commercially rolled strip that had produced better than a 50 percent yield of usable supports had a minimum bend radii of 1.70 T. Minimum bend radii of some of the experimental strip fabrication schedules are listed in Table 4.2.

TABLE 4.2

ANTICLASTIC BENDING
(See Figure 4.10)

<u>Schedule</u>	<u>Material</u>	<u>Forming Method</u>	<u>θ, Degrees</u>	<u>Δ, Inches</u>	<u>Minimum Bend Radii Thickness Units</u>	<u>Tensile Tests Width Strain/ Thickness Strain</u>
	Mild Steel		43	0.0075	<0.5	0.95
	Zircaloy	Extruded and Drawn Tubing	35	0.0100	<0.5	1.0
22	Zircaloy	Extruded and Turkshead	62	0.0112	1.08	
19	Zircaloy	Extruded and Rolled	66	0.013	1.72	
21	Zircaloy	Rolled	68	0.0131	1.49	
18	Zircaloy	Extruded and Rolled	69	0.0150	1.56	
20	Zircaloy	Extruded and Rolled	70	0.0150	1.76	1.95
	Zircaloy	Commercially Rolled	79	0.0150	1.94	2.3

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The mechanical anisotropy of Zircaloy-2 sheet material is well documented, ⁽²⁾ and it was shown that conventional rolling practice produces a material that does not thin much in the sheet thickness direction when it is pulled in tension in either the rolling or transverse directions. Geometry effects on an isotropic material such as steel causes a greater strain to occur in the thickness direction of a flat tensile specimen than in the width direction. The ratio of natural thickness strain-to-width strain shown in Table 4.2 illustrates the mechanical anisotropy of Zircaloy-2; whereas, this ratio for steel is less than 1.0, all the values for Zircaloy are greater than 1. This characteristic of rolled Zircaloy-2 strip, a high flow stress in the thickness direction with respect to the width and length direction, increases the amount of anticlastic curvature present when the strip is bent. When the angle θ (Figure 4.10 and Table 4.2) is used as a measure of anticlastic curvature it is apparent that the larger the anticlastic curvature the greater is the minimum bend radius.

The final test of strip quality is the yield of usable supports obtainable from the strip. Over 100 support blanks were cut from each of Schedules 18, 19, 20, 21, and 22. There were no rejects from any of these schedules because of cracking in the bends.

FIGURE 4.10

Schematic Illustration of Anticlastic Curvature in Bent Strip
(The angle " θ " was used as a measure of the relative amount of anticlastic curvature of samples from different fabrication schedules. These values are listed in Table 4.1)

Figure

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The final test of finished support quality is a fatigue test. The support is preloaded so that a 25-pound oscillating load will produce a deflection of 0.030 inch. If samples from a lot of material consistently endure 25,000 cycles of this testing, the lot is considered acceptable.

Rolling Cerium - F. B. Quinlan

Cerium metal has been successfully rolled from a cast billet to a 1/6-inch sheet. The metal was cold rolled to a 20 percent reduction, taking approximately 5 percent per pass. It was then annealed in an evacuated quartz tube at 500 C for 1 hour. This procedure was repeated until 3/4-inch thickness was reduced to 1/16 inch. Severe edge cracking of the cerium will occur if reductions of more than 20 percent are attempted. Chamfering of the edges of the billet prior to rolling reduces the tendency to edge crack. No special precautions against oxidation were used during the rolling operation.

FUEL EVALUATION

Dual-Enriched Single Tube N-Reactor Fuel Element - D. P. O'Keefe

The experimental Zircaloy-2-clad single-tube dual-enriched metallic uranium fuel element, described in previous quarterly reports, has completed the third cycle of high-temperature irradiation in the M-3 pressurized water loop in the ETR. The element has accumulated a maximum exposure of approximately 700 Mwd/ton and is currently undergoing the fourth cycle of reactor operation. The element was examined after the first and second cycles of operation. After the first cycle, the element had warped 100 mils toward the reactor core. It was reinserted with the direction of warp away from the core, but after the second cycle, was again found to be warped 100 mils toward the core. Since the M-3 test facility is in the reactor shield, an element irradiated in this facility is subjected to a pronounced radial flux gradient. The resulting diametral temperature gradient in the test element is believed to be the cause of the warp. No corrosion effects or other damage have been observed on the element.

Initially in this experiment, the element operated with the maximum uranium temperature of 570 C. In subsequent reactor cycles, the decreases in the general power level of the reactor have resulted in progressively lower power generation rates for this experiment. The element currently is operating with a maximum metal temperature of about 450 C.

Irradiation of Fuel Elements with Brazed Closures - R. N. Johnson

Radiometallurgical examination is nearly complete on the variable braze-thickness irradiation test, GEH-4-68, 69, and 70. All end closures have been sectioned and have been found to be free of any cracks in the braze or other signs of incipient failure, regardless of the thickness of the braze layer. Several of the sections, however, have shown extensive cracking in the uranium in the braze-heat-affected zone. In one section,

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the lower end of GEH-4-70, the cracking appears to originate in a cluster of voids. These voids may have been caused by the brazing process prior to irradiation, or may have occurred by grain boundary tearing during irradiation. Figure 4.11 shows a cluster of these voids with the cracks radiating out from them. Figure 4.12 gives a closer look at one of these void areas. The entire length of GEH-4-70 is being sectioned to be sure that the cracking is confined only to the braze-heat-affected zones of the uranium.

FIGURE 4.11

Voids in Irradiated Uranium
250X, Bright Field

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FIGURE 4.12

Cracks Originating in Void Clusters
25X Composite, Polarized Light

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Photograph Without Titles Unclassified
ARCEC RICHMOND, WASH.

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Fuel Element Swelling Model - K. R. Merckx

Swelling of metallic uranium during high temperature irradiations is a major factor influencing in-reactor performance of metallic fuel elements. An empirical model for swelling based on reactor fuel performance is needed to predict the behavior of swelling for various metallic uranium fuel systems. Since the temperature and burn-up dependence of swelling is nonlinear, a method is needed to estimate the incremental swelling from the gross volume changes observed in test fuel elements and capsules. An empirical model for the incremental swelling behavior has been developed using seven parameters. The incremental model for swelling is integrated over the volume of the fuel material to form an estimate of the gross fuel element swelling for each test irradiation. The values of the parameters are adjusted to minimize the total variance between the estimated and measured gross fuel element swelling for any series of test data. The relationships required to make an evaluation of parameters in such a swelling model were programed and used on the experimental results from 12 test fuel elements. To make adequate estimates of this swelling data, parameters had to be included that were equivalent to a burn-up dependent cutoff temperature below which only the theoretically minimum swelling occurs. The incremental volume swelling model, capable of representing the experimental data, separates the total swelling into a theoretical minimum swelling component and that component of swelling attributed to fission gases as restrained by coolant, cladding, and a fuel restraint pressure. This incremental volume swelling model has the form

$$v = A*B + (G*B*T)/(P_c + P_r + \theta_7*T_r);$$

where

v = incremental swelling,

B = burnup,

T = absolute temperature to standard temperature ratio,
(Temp. °K)/273,

P_c = coolant pressure,

T_r = (outer cladding radius)/(cladding thickness),

P_r = fuel restraint pressure,

$$= \theta_1*B^{\theta_2}*T^{\theta_3}[1 + \exp(-\theta_4*(T + \theta_5*B - \theta_6))],$$

$\theta_1, \dots, \theta_7$ = parameters adjusted by model,

A = theoretical minimum swelling rate, and

G = gas generation rate.

To effectively evaluate data from various test irradiations, all the numerical relationships are programed. The computer program allows any of the parameters to be fixed at prescribed values in order to evaluate the effect of the variation of different parameters. In the initial evaluations, the effect of cladding restraint has been neglected by fixing θ_7 at zero. An additional program was written to evaluate and plot the results of the incremental volume swelling model for five increments of burnup

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and up to one hundred temperature increments. A sample output of this program is given in Figure 4.13. A report⁽³⁾ describing the model, method of evaluation, and how to use the computer program has been published.

Tubular Test Element Fuel Swelling - J. W. Goffard

Previous analysis of 1.6 percent enriched KSE-3 uranium fuel element (1.75-inch OD x 1.06-inch ID x 0.025-inch inner and outer Zircaloy-2 cladding) swelling data yielded an empirical expression which relates the total fuel swelling to the 1.65 power of burnup and mean fuel temperature in the temperature range 400 to 470 C.⁽⁴⁾ The expression is

$$\% V = 3.0 (\text{at. \% B.U.})^{0.0145} (T_m - 365)(\text{at. \% B.U.})^{1.65};$$

where T_m = volume mean fuel temperature, °C, and
B.U. = burnup of fuel.

This expression describes the fuel behavior during the transition from the theoretical minimum to the theoretical maximum swelling. However, the use of a mean fuel temperature as a parameter in a temperature range where swelling is a highly temperature dependent phenomenon greatly restricts the use of this expression.

A more complete empirical swelling model developed by K. R. Merckx is based upon the temperature, burnup, and pressure dependent swelling behavior of an incremental volume of fuel. (See Fuel Element Swelling Model, this report.) The model describes the swelling behavior of an incremental volume of the uranium fuel and can be readily used to describe the swelling of a fuel element by summing of the individual incremental changes in the fuel volumes chosen to represent the element. The model is expressed as

$$v = AB + \frac{GBT}{PC + \theta_1 B^{\theta_2} T^{\theta_3} [1 + e^{-\theta_4 (T^{\theta_5} B - \theta_6)}]};$$

where v = vol% increase of a volume element,
 A = constant = 3.0,
 B = atom percent burnup,
 G = constant = 550,
 T = $K^\circ/273$,
 PC = coolant pressure, atmospheres, and
 θ_1 to θ_6 = parameters, statistically evaluated.

The values of the parameters have been evaluated to be

$$\begin{aligned}\theta_1 &= 19.5067, \\ \theta_2 &= -1.1470, \\ \theta_3 &= -1.0739, \\ \theta_4 &= 195.00, \\ \theta_5 &= 0.11204, \text{ and} \\ \theta_6 &= 2.50385.\end{aligned}$$

FIGURE 4.13

Fuel Swelling Versus Temperature in Transition Region

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The six parameters of interest have been statistically evaluated using a computer program to obtain the best agreement between the model and the experimentally determined KSE-3 fuel element swelling data. Figure 4.13 is a plot of volume increase versus fuel temperature in the transition range at a coolant pressure of 1600 psi and for burnups to 0.5 percent in 0.1 percent increments. The temperature sensitivity of the transition in swelling from the theoretical minimum to the limiting maximum is very striking. As burnup is accumulated in the fuel, the temperature at which transition occurs is decreased. It is to be noted that the figure illustrates the swelling behavior of an incremental volume of uranium and not of a specific fuel element.

The theoretical maximum swelling, the plateaus on the right side of Figure 4.13 by which the transition curves are limited, have not been established experimentally. To obtain data in the temperature region above the transition region, additional fuel element irradiations are planned. Single tube coextruded fuel material of a slightly modified KSE-3 geometry has been extruded and is being fabricated into fuel elements for irradiation. This irradiation will furnish fuel swelling data for fuel temperatures higher than obtained with the KSE-3 elements and will hopefully verify the maximum swelling limits defined by the fuel element swelling model.

METALLIC FUEL ELEMENT DEVELOPMENT

Metallic Fuel Element Development - G. A. Last and J. E. Minor

Thorium is a ductile, readily fabricable, high-melting, cubic metal that is more conductive, corrosion resistant, and resistant to irradiation damage than uranium. As a fertile material in a reactor fuel system, it yields highly fissionable U^{233} . For these reasons, thorium metal or thorium alloys containing fissionable isotopes have great potential as economic nuclear fuel materials capable of being tailored to a wide variety of thermal reactor applications.

A program has been initiated which will develop the techniques required to fabricate coextruded, Zircaloy-clad, fully bonded thorium-uranium fuel elements suitable for irradiation in pressurized high temperature water environments. The irradiation behavior of these elements in that in-reactor environment will be evaluated, and additional elements and samples will be subjected to a battery of out-of-reactor destructive and nondestructive evaluation tests.

After study of the history of thorium materials available at Fernald, a stock of arc melted material in slug form was selected and shipped to Hanford. Fully enriched uranium fabricated into sheared strip has been obtained for the required enrichment. The facilities required for the various cleaning, welding, melting, extrusion, and chemical milling operations have been provided with suitable ventilation and filter protection to permit the safe handling of these materials.

Electrode Fabrication and Arc Melting Development - R. G. Nelson

The development of the fabrication procedures for these elements is being performed on 24-pound pilot billets, 2.7 inches in diameter, using natural uranium. Selected thorium slugs, 1.34 inches in diameter by 6 inches long, were drilled to provide a 1/4-inch longitudinal hole for the insertion of the uranium additions. Eleven of these slugs were electron beam welded end to end to form first melt electrodes.

The second pilot electrode included sufficient zirconium strip welded to the side of each slug to form a Th-2.5 wt% U-1 wt% Zr alloy as the final product. Figure 4.14 shows the first melt electrode stub, uranium wire, zirconium strips and the first melt ingot. Since the zirconium addition to thorium will have the effect of sequestering and tying up carbon originally present in the thorium, effect of the alloy addition on coextrusion properties, bond properties, and irradiation performance will be a matter of major interest.

The first melt ingots were then fabricated into second melt electrodes by quartering the first melt ingot longitudinally and electron beam welding the quartered sections end to end. The pattern of placement of the quarter sections within the electrode was chosen to promote homogeneity of the final ingot. Observation of melting during initial heat (thorium-uranium with no zirconium addition) led to modification of the melting practice for the second ingot. The thorium-uranium ingot first melted showed sidewall porosity, and complete cleanup was not obtained at the 2.696-inch finished diameter. The 1 wt% zirconium ingot produced during the second melt machined to finished size without visible defects. Figure 4.15 shows the pronounced difference in the as-cast appearance of the 1 wt% zirconium ingot, Heat #119.

To further confirm the effect of the zirconium addition on ingot quality, a third ingot was melted (Heat #121). This ingot had no zirconium addition and melting conditions were identical to those used on the 1 wt% zirconium ingot, Heat #119. While the quality of Heat #121 was improved over the first ingot (Heat #118), its surface condition was rougher and more porous than Heat #119.

The 1 wt% zirconium addition definitely improves the as-cast surface condition of the ingot. It apparently increases the fluidity of the melt, scavenges the gases, and lowers the melting point slightly. This addition also lowers the room temperature hardness from 94 to 72 BHN.

FIGURE 4.14
Primary Electrode and Ingot of Pilot Heat #117
(Th-2.5 wt % U-1 wt % Zr)

FIGURE 4.15

As-Cast, Double Vacuum Arc-Cast Ingots, Heat #118 (Th-2.5 wt% U)
and Heat #119 (Th-2.5 wt% U-1 wt% Zr)

(Note the improved sidewall quality of the 1 wt% zirconium alloy)

Neg. No. 0621215-3

AEC-62 RICHLAND, WASH.

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Chemical analysis for the three ingots melted and the extrusions produced are shown in Table 4.3. Uranium and zirconium values are the average of center and edge locations. No logical explanation for the low hardness in 1 wt % zirconium ingot is revealed by these analyses. Oxygen levels of all three ingots are in the 1000 to 1100 ppm range; nitrogen, 100 to 200 ppm; and carbon, 150 to 300 ppm. It appears from the table that somewhat better homogeneity of uranium is shown in the 1 wt % zirconium alloy.

TABLE 4.3

CHEMICAL ANALYSES

Ingot and Extrusion No.	Weight Percent					
	U	Zr	O	N	C	H
118 Top	2.58		0.1090	0.0187	0.0205	0.0001
Bottom	2.38				0.0202	
Front	2.7					
Middle	2.54					
Rear	2.74					
119 Top	2.47	1.08	0.1100	0.0188	0.0145	
Bottom	2.50	1.08			0.0300	
Front	2.38					
Middle	2.48					
Rear	2.54					
121 Top	2.35		0.1153	0.0075	0.0152	
Bottom	2.20				0.0160	
Front	2.9					
Middle	2.71					
Rear	2.8					

Metallic impurities all less than 50 ppm except for Fe at 100 ppm and Ni at 50 to 100 ppm.

Electron beam welding of pure thorium has shown tendency toward cracking the metal. This was noticeable in the pilot electrode welding. While the joints had a rather low strength, the strength requirement of the small electrode (24 pounds) was not large enough to cause concern.

The 75-pound weight of the enriched electrode necessitated better quality weld joints. It was found that zirconium added to the weld joint produced a good strong crack-free weld.

The work of these pilot ingots, to date, has shown that the zirconium addition

1. lowers room temperature hardness,
2. improves ingot sidewall quality resulting in higher metal yields,

3. indicates an improvement in uranium distribution, and
4. adds to strength of the weld joint in the primary electrode.

For these reasons and the fact that the zirconium addition does not show any harmful effects on bond strength or structure, the decision to make the enriched material with the 1 wt % zirconium addition has been made.

Coextrusion Development - P. A. Ard

The three pilot thorium alloy ingots were successfully coextruded into Zircaloy-2-clad, 0.875-inch OD by 0.495-inch ID tubes at an extrusion ratio of 17.4 to 1 based on the tooling dimensions. The billets were designed to simulate the extrusion of the final 1.75-inch OD, Zircaloy-2-clad, thorium-enriched uranium alloy tubes as to extrusion ratio, preheat temperature, and the alloy to Zircaloy-2 ratio.

The bare thorium billets machined from the as-cast billets are shown in Figure 4.16. Figure 4.17 shows the billet assembly prior to sealing.

The thorium alloy cores were vacuum sealed within the Zircaloy-2 cladding sleeves and the composite billet was vacuum sealed in copper and lubricated with Fisks 604 and graphite (suspension of aluminum fines) for extrusion. The 3-inch OD billets were preheated with the graphite cutoff blocks in closed preheat containers for approximately 4 hours at 760 C. The extrusion die and billet arrangement can be seen in Figure 4.18.

The difference in the extrusion force between the thorium alloy billets was not large. Running extrusion constants (K) are tabulated below.

<u>Billet</u>	<u>Alloy</u>	<u>K, (Tons/in.²)</u>
118	Th-2.5 wt % U	18.7
119	Th-2.5 wt % U-1 wt % Zr	18.7
121	Th-2.5 wt % U	18.0

K is derived from the extrusion expression,

$$F = KA \ln R,$$

where F is the total extrusion force in tons,

A is the area of the billet to which the force is applied, and

R is the reduction ratio.

Billet 121 showed a $3\frac{3}{4}$ percent decrease in force from the other two billets. This billet had the better quality core of the two Th-2.5 wt % U billets. The addition of the 1 wt % zirconium does not show any significant effect on the extrusion force at 760 C preheat temperatures.

FIGURE 4. 16
Bare Th-2.5 wt% U Extrusion Billets

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4.33

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611

FIGURE 4.17

Billet Assembly for Pilot Th-2.5 wt % U Extrusion

Neg. No. 0621397
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FIGURE 4.18

Extrusion Die and Billet Arrangement Showing the Billet
Coated with Lubricant for Extrusion

Copper Stripping of Thorium Coextrusions - R. V. Bowersock and
G. S. Allison

Difficulties were encountered in bright etching of the Zircaloy-2 cladding on the thorium coextrusions. The difficulty manifested itself as small "islands" of material being left in relief as the bulk of the surface was etched away. The islands were oriented in longitudinal bands that varied in width from 1/64 to 1/4 inch. Close examination of the islands revealed a thin cap of copper-colored material. The bumping was associated with areas of surface tearing of the Zircaloy-2 cladding exposed when the copper was removed by stripping in warm nitric acid. It has been found that by use of a more stringent copper stripping procedure the surface will etch uniformly in the standard HF-HNO₃ Zircaloy etch. This procedure involves stripping in hot, 60 C or greater, nitric acid (50 percent); followed by a thirty-second dip in 40 C HCl (37 percent), then back into the hot nitric acid for approximately two minutes.

A possible explanation of the observed behavior is that a brittle Zr-Cu alloy or compound is formed at the higher extrusion temperature of 760 C required for coextrusion of thorium. This brittle material consequently broke up during the extrusion process. Assuming the material to be more difficult to strip it would provide protection for the underlying Zircaloy while in the bright etch resulting in the formation of "islands"

Extrusion of copper-clad, Zircaloy-2 billets to 0.300-inch diameter rods indicates that excessive diffusion may be the cause of the difficulties noted. Two billets were extruded, one at 800 C and one at 850 C preheat. The higher preheat temperatures were used to compensate for increased chilling of the smaller billets and thereby approximate the extrusion temperatures of the thorium fuel coextrusions.

The resulting rods were cut into 3-inch lengths, samples of which were copper stripped on half their length. Those of the 800 C preheat exhibited very little of the abnormal etching behavior. Those of the 850 C preheat exhibited a large amount of the abnormal etching behavior as seen to a lesser degree on the thorium fuel pieces.

Two of these pieces, one 800 C and one 850 C preheat, were sectioned through the unstripped end and polished to show the Cu-Zr compound layer. As can be seen in Figure 4.19, the rod of higher preheat shows intermittent excessive diffusion at the interface, while the lower preheat shows little or none. The excessive diffusion seen on the 850 C preheat sample is identical in appearance to that seen on some cross sections of the thorium fuel pieces.

Since the thorium fuel for irradiation will be coextruded in a 3 percent Si-Cu can, trials of this material will be made. It is possible that, since the excessive diffusion experienced seems to be borderline, the different material may inhibit diffusion sufficiently to eliminate the problem.

Zircaloy

Copper

850 C

Zircaloy

Copper

800 C

FIGURE 4.19

Diffusion Zone of Copper-Clad, Zircaloy-2 Rods
Extruded at Temperatures Shown
250X

Neg. No. 4W-62-2241; 4W-62-2240
AEC-GE RICHLAND, WASH.

Coextrusion Evaluation - R. S. Kemper

Two Th-2.5 wt% U and one Th-2.5 wt% U-1 wt% Zr alloy, Zircaloy-clad pilot coextrusions have been examined for clad-core bonding, clad uniformity and core structure. Clad uniformity is extremely good in both alloys with both inner and outer clad thickness variation being approximately ± 0.0005 inch, indicating that as-cast, machined billets may be used without primary working. A typical cross section of a coextruded tube is shown in Figure 4.20. Metallographic examination was made of samples from the front, middle and rear of each of the coextrusions. The clad-core interfaces were clean with no evidence of unbonding. The thorium alloy cores were only partially recrystallized at this extrusion temperature. Recrystallization increased progressively toward the rear as indicated in Figures 4.21 and 4.22. The finer grain size of the alloy with zirconium addition is evident in these figures. As-extruded hardness shown in Table 4.4 also indicates increasing recrystallization and the softening effect of the zirconium addition. The location or amount of any uranium not in solution in either alloy has not been identified metallographically. The primary inclusions shown in Figures 4.21 and 4.22 are thorium oxide. Sections of both alloys have been treated for 1/2 to 3 hours at 700, 800 and 900 C. to observe the effect on clad-core diffusion and core structure. Examination is not completed but uniform growth of the Zr-Th alloy diffusion zone was observed in 1/2 hour at 800 C with complete recrystallization in the Th-2.5 wt% U-1 wt% Zr alloy.

TABLE 4.4HARDNESS OF AS-EXTRUDED ALLOY

<u>Billet No.</u>	<u>Alloy</u>	<u>Hardness, R_F*</u>		
		<u>Front</u>	<u>Middle</u>	<u>Rear</u>
118	Th-2.5 wt % U	90	90	89
121	Th-2.5 wt % U	89	85	83
119	Th-2.5 wt % U-1 wt % Zr	81	72	70

* R_F - 1/16-inch ball - 60 kg load

Acid Milling of Thorium Alloys - R. V. Bowersock

One step in the fabrication of fuel elements from cut lengths of Zircaloy-2-clad thorium alloy fuel requires that the fissionable material be removed from the ends to permit the insertion of Zircaloy-2 caps. The criteria governing the recessing process are that the fissionable material shall be recessed up to 0.3 inch, the exposed cladding must be completely free of all fissionable material which could contaminate the end closure, the cladding shall not be adversely affected by the recessing process, the resulting surface of the core material should be relatively flat and perpendicular to the cladding, the metallurgical bond between the remaining core material and the cladding shall not be adversely affected, and the recessing process shall be amenable to manufacturing standards. Recessing by selective chemical dissolution is being investigated to see if such a process can be developed to meet the foregoing criteria.

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FIGURE 4.20
Cross Section of Zircaloy-Clad
Th-2.5 wt % U Alloy
Coextruded in Copper
3X

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4. 39

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Middle

Rear

FIGURE 4. 21

Extrusion 119 - Th-2.5 wt % U-1 wt % Zr, Longitudinal Sections
500X

Front

Neg. No. 4W-62-1830B; 4W-62-1832
4W-62-1834B
RECEIVED, WASH

UNCLASSIFIED

UNCLASSIFIED

4.40

HW-74378

FIGURE 4.22

Extrusion 121 - Th-2.5 wt % U, Longitudinal Sections
500X

Neg. No. 4W-62-1964B; 4W-62-1966B;
4W-62-1968B
AEC-RICHMOND, WASH.

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To date, two milling solutions have been investigated. They are the standard milling solution for metallic uranium and hydrochloric acid. The standard uranium milling bath consists of a mixture of HNO_3 , H_2SO_4 , and CuSO_4 to give the following bath composition:

Cu	0.175 lb/gal
NO_3^-	1.1 lb/gal
SO_4^{--}	4.8 lb/gal

No apparent attack of either thorium alloy was observed in the 75 to 95 C temperature range. Various concentrations of HCl have been investigated and the results are summarized in Table 4.5.

TABLE 4.5

ACID MILLING RATES
OF THORIUM-URANIUM ALLOYS IN HCl

HCl, %	Temperature, C	Th-2.5 wt % U, mils/min	Th-2.5 wt % U -1 wt % Zr, mils/min	Remarks
5	30	0.2	0.4	
10	24	0.4	0.4	
	31	0.9	1.0	
	40	1.6	1.7	
	50	2.6	3.0	
	31	1.9	2.5	Surface uneven with deep pockets
20	24	0.2	0.2	Bond line under-cutting
	34	0.7 (avg)	0.4 (avg)	Initial rates were 1.3 and 1.2 mils per minute, respectively, but dropped off rapidly
37 ¹ / ₂	23	0.15	0.2	Bond line under-cutting

Generalization from the work to date indicates that 10 percent HCl could be used as a satisfactory milling bath and that the addition of 1 percent zirconium to the Th-U alloy appears to enhance both surface characteristics and etch rate. Further investigation of HCl is scheduled with emphasis on surface characteristics.

Irradiation Testing of Thorium Alloy Fuel Elements - J. W. Goffard

To evaluate the high burnup performance of thorium base fuel elements in high temperature water coolant, Zircaloy-2-clad Th-1 wt% Zr -2.5 wt% Orallo (93.5 percent enriched uranium) fuel elements will be irradiated in the ETR. The 8-inch-long test elements will be fabricated from coextruded tubular material 1.75-inch OD x 1.05-inch ID x 0.025-inch clad. The initial peak operating conditions for the foregoing test elements in the ETR P-7 test facility are calculated to be:

Maximum specific power	223 kw/foot
Maximum fuel temperature	~620 C
Maximum inner heat flux	1.11×10^6 Btu/(hr)(ft ²)
Maximum outer heat flux	1.01×10^6 Btu/(hr)(ft ²)

The specific power is expected to decrease with fuel burnup, e. g. ,

223 kw/foot maximum at 0.0 at % burnup
181 kw/foot maximum at 0.5 at % burnup
156 kw/foot maximum at 1.0 at % burnup

The surface heat fluxes are reduced in proportion to the power reduction. The average specific power of the 2-foot column of fuel material (three 8-inch-long elements) will be 122 kw/foot with a wide range of fuel operating conditions prevailing within the irradiation test. Dimensional stability and fuel swelling will be determined during the course of irradiation by interim measurement in the ETR canal.

The design of an experimental test assembly and the experimental test proposal for irradiating three test assemblies in the P-7 loop facility at the ETR are essentially complete.

REFERENCES

1. Cadwell, J. J. Quarterly Progress Report, Fuels Development Operation, April, May, June, 1962, HW-74377, p. 4.6. July 20, 1962. (SECRET).
2. Rittenhouse, P. L. and T. A. L. Picklesimer. Metallurgy of Zircaloy-2 Part II, ORNL-2948. February 1, 1961.
3. Merckx, K. R. Statistical Evaluation of an Empirical Fuel Element Swelling Model, HW-74919. September 11, 1962.
4. Cadwell, J. J. Quarterly Progress Report, Fuels Development Operation, January, February, March, 1962, HW-72347, p. 4.35. April 16, 1962. (SECRET).

CERAMIC FUELS DEVELOPMENT

CERAMIC (URANIUM) FUELS RESEARCH

Explosive Melting of UO_2 and ThO_2 - J. A. Christensen

Direct resistance heating of UO_2 and ThO_2 was used as means of generating high temperature fuel states typical of what might be expected in the cores of high-rated fuel elements, or during severe reactor power transients. A further goal is development of techniques for resistance heating semiconductors for examination at high temperature in oxidizing atmospheres without the presence of foreign materials. Short, high current pulses were used to cycle samples from 1000 C to above the melting temperatures, to below 1000 C in less than three seconds. Energy input to a $1/4 \times 1/10 \times 1$ inch long specimen of sintered UO_2 was varied between 5 and 30 kw. Heating time was controlled by varying the voltage drop across the specimen. High speed motion pictures (to 16,000 pictures/second) of the melting sequence that were made to study the melting cycle also provided data of more fundamental nature, such as the surface tension of $\text{UO}_2(\text{liq})$ (Figure 5.1).

The O/U ratio remained constant at 2.001 during fusion cycles lasting between 1.5 and 3.5 seconds. Molten drops which fell from specimens during heating were quenched very rapidly by impinging on a cold tungsten plate. The microstructure of these quenched drops was typical of fused UO_2 except for the presence of many metallic appearing inclusions which were etched preferentially in $\text{HNO}_3\text{-H}_2\text{O}_2$ solutions (Figure 5.2). The identity of these inclusions is yet to be established. The material showed only a UO_2 X-ray diffraction pattern. An X-ray diffraction pattern of the surface of a quenched drop showed almost no crystallinity, indicating a highly strained structure. The interiors of specimens were 100 percent crystalline, as were ground samples.

The method of heating, the high melting point, and low thermal conductivity of UO_2 result in an increasing thermal gradient from the surface to the center of the specimen. Melting of the UO_2 initiates at the center of the sample, often resulting in a molten core surrounded by a shell of solid, sintered UO_2 --a structure frequently realized in fuel cores during irradiation. Figure 5.3 illustrates this situation with a vertical section of a heated UO_2 rod in which the molten center was partially expelled through a gap in the solid surrounding crust. The sharp discontinuity between fused and sintered UO_2 is very similar to that seen in irradiated UO_2 .

FIGURE 5.1

High Speed Photographs Showing Stages in Development
of Drops of Liquid Uranium Dioxide, 1 X
Temperature: 2800 C; Time Lapse Between Photographs: 5×10^{-3} sec.

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5.3

HW-74378

FIGURE 5.2

Microstructure of Explosively Melted UO_2 : Note Preferentially Etched White Inclusions

Left: 4J1340A; Right: 4J1340D

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Negative 5J1338

10 X

FIGURE 5.3

Explosively Fused UO_2 ; Center of Bar Melted While Outer Crust Remained Solid

Surface Tension of Molten UO_2 - J. A. Christensen

Surface tension of $\text{UO}_2(\text{liq})$ was calculated from drop profiles recorded on high speed motion pictures of explosively melting UO_2 (Figure 5.1). Analyses of three pendant drop profiles by two independent methods yielded values between 340 and 760 dyne cm^{-1} with a mean of 490 dyne cm^{-1} (roughly the same as mercury or molten lead). The temperature of the liquid was assumed to be constant at 2800 C, the melting point of UO_2 .

Specific Volume of Molten UO_2 - J. A. Christensen

The specific volume of UO_2 was measured in the temperature range 1200 to 3100 C (Figure 5.4) by high temperature Co^{66} radiography of single crystals in sealed tungsten vessels. Excellent reproductibility was achieved at all temperatures. The change in volume on melting was 9.65 percent. Specific volume of the liquid at the melting point was 1.25 times that of the solid at room temperature. The coefficient of expansion of the liquid between 2800 C and 3100 C was 3.7×10^{-5} in/in C. Lower temperature results (less than 2500 C) agree well with those of other investigators.

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FIGURE 5.4

 UO_2 Specific Volume Versus Temperature

Heat of Fusion of UO_2 - J. A. Christensen

Measurements of the change in volume on melting and the surface tension of UO_2 have been used to calculate the heat of fusion. A value of 3300 cal/mole was obtained.

These data also were used with the Clapeyron equation to calculate the dependence of melting temperature upon pressure. The results indicate an 8 C increase in melting temperature should occur under 100 atmospheres pressure, which is the maximum pressure achievable with the present equipment. Since the precision in measuring temperature at 2800 C is $\pm 20^\circ$, this 8° change was not detected.

A knowledge of the heat of fusion of UO_2 may also be of value in investigating the kinetics of reactor transients.

Plastic Deformation of UO_2 - J. A. Christensen

A single crystal of UO_2 was ductile under torsional stress at 2100 C. The crystal, 1/8-inch thick by 1/4-inch wide by 1-inch long, was twisted 120 degrees at approximately 10 degrees/second. It was deformed plastically much like the polycrystalline UO_2 tested previously.

The O/U ratio of polycrystalline UO_2 remained constant at 2.001 ± 0.0002 during torsional tests at 2300 C. The microstructures of deformed specimens showed some separation at the grain boundaries and little grain growth.

Melting Temperature of Irradiated UO_2 - J. A. Christensen

The melting temperature of fused UO_2 was not altered by 300 MWD/T irradiation at temperatures between 400 and 2750 C. Fifty melting point measurements of specimens selected from known locations in each microstructural region of an irradiated 1.44-inch OD fuel rod cross-section gave values within 50 degrees of the melting point of nonirradiated UO_2 . The melting point of sintered UO_2 was previously observed to increase approximately 100 degrees during irradiation to 300 MWD/T. Sintered UO_2 irradiated to between 40 and 7000 MWD/T has consistently demonstrated higher melting temperatures than nonirradiated oxide. Fused UO_2 , however, does not become more refractory during irradiation--presumably, because it contains no excess oxygen which seems to be required to stabilize higher melting compositions.

Microhardness of Single Crystal UO_2 - J. L. Bates

Microindentation hardness measurements are useful in characterizing the plastic properties of UO_2 and evaluating lattice alterations after irradiation. Recent data indicate that hardness increases with increasing neutron exposure. The hardness of a UO_2 single crystal⁽¹⁾ (A-7), irradiated to 1.44×10^{21} fissions per cubic centimeter (f/cc), increased 18-22 percent (Figure 5.5) that of a second single crystal (G-7), irradiated to only approximately 1×10^{14} f/cc, increased approximately 8 percent, Figure 5.6. The hardness of each crystal remained dependent upon crystal orientation after irradiation.

Irradiations and measurements on single crystal UO_2 are continuing.

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FIGURE 5.5

Microindentation Hardness of UO_2 Single Crystal (A-7) as a Function of Crystal Surface-Indenter Orientation Before and After Irradiation to 1.44×10^{21} f/cc

FIGURE 5.6

Microindentation Hardness of UO_2 Single Crystal (G-7) as a Function of Crystal Surface-Indenter Orientation Before and After Irradiation to Approximately 1×10^{14} f/cc

Pressurized Melting of UO₂ - J. A. Christensen

High temperature microscopy of UO₂ heated to its melting point (2800 C) under argon pressures between 0 and 1500 psi continued to show marked inhibition of vaporization by increased gas pressures. Crystal growth from the vapor above melting UO₂ was entirely absent at 1500 psi and typically profuse below 100 psi. As expected, pressures in this range (0 -1500 psi) had no effect on the melting point of UO₂. Specimen temperature at a given heat input rate was, however, an inverse function of pressure--presumably because of more efficient heat transfer by convection at higher pressures. For example, the UO₂ temperature increased from 2800 to 2920 C when the argon pressure dropped from 1500 psi to 15 psi. This temperature change enhances the apparent effect of pressure on vaporization rate but does not alter the observation of decreased vaporization rate at increased pressures.

A satisfactory theoretical explanation of the inhibition of vaporization by increased over-pressure of gas was postulated assuming diffusion of vaporizing molecules through a stagnant surface film as the rate limiting step. Film thickness increases with pressure; thus, the higher the pressure, the longer the time required for a UO₂ molecule to diffuse to the film surface.

Thermal Testing of Molten UO₂ Fuel Pins - J. A. Christensen

Tungsten clad single crystals of UO₂ are scheduled for irradiation to determine the in-reactor behavior of molten fuel cores. Specimens were heated to 3100 C, 300 above the melting point, with no indication of capsule failure. Capsules were fabricated by grinding and spark drilling of swaged tungsten rod. Each capsule was filled with a 1/8-inch diameter single crystal of UO₂ and was closed by either electron beam welding or tungsten inert gas welding (Figure 5.7). Six capsules (three closed by electron beam and three by TIG welding) were tested by resistance heating them to 3100 C (or to the temperature at which the solid tungsten stem melted--if this occurred first). Of the six, one leaked through a small crack initially present in the tungsten capsule. The other five capsules were sound to at least 2900 C, and three of them remained intact to 3100 C. Time at temperature was eight minutes at 2800 C, 2900 C, 3000 C, and 3100 C for each capsule.

Neg. 29801-1

Before Welding

Neg. 0621788

Electron Beam Welded
FIGURE 5.7
Tungsten Clad UO_2 Pins

UO₂-ThO₂ Equilibrium Studies - J. A. Christensen

The liquidus for the UO₂-ThO₂ system was determined (Figure 5.8). Principal features are total miscibility and melting minimum of 2760 C at 2 wt % ThO₂. Solid solutions were prepared by electrolysis from fused salts. A UO₂ 7.5 wt % ThO₂ solution was obtained by in-reactor homogenization of a heterogeneously mixed fuel core. Samples with 95 wt % ThO₂ were prepared at ORNL by the Sol-Gel process. All melting points were measured under one atmosphere of argon in a tungsten filament furnace.

FIGURE 5.8
UO₂-ThO₂ Liquidus

Gross fractionation of uranium to the vapor phase was observed from each specimen studied, reflecting the high vapor pressure of UO_2 relative to that of ThO_2 . X-ray fluorescence analyses of the fused residue and the sublimate from specimens having initial compositions of UO_2 - 2 wt % ThO_2 , UO_2 - 12 wt % ThO_2 and UO_2 - 41 wt % ThO_2 showed the following compositional changes during melting:

<u>Initial ThO_2 Concentration (wt %)</u>	<u>ThO_2 Concentration in Sublimate (wt %)</u>	<u>ThO_2 Concentration Residue (wt %)</u>
2	0	4.3
12	8	28
41	17	51

The continuously changing composition introduces uncertainty into phase equilibrium studies. These results suggest a possible approach to U-Th separation, and are valuable in interpreting the structures of previously irradiated UO_2 - ThO_2 mixtures.

Electron Microscopy of UO_2 Single Crystals - J. L. Daniel and J. O. McPartland

Earlier reports of this series discussed changes of surface structure that occurred during heating of an irradiated UO_2 crystal. Additional surface changes, apparently brought about by in-microscope heating to 980 C, were detected during studies of photomontages of reflection electron micrographs of the UO_2 single crystal irradiated to 1.44×10^{20} f/cc. Numerous additional surface protrusions were apparent after heating, as well as modifications of some original features (Figure 5.9). The added small particles may have resulted from an explosive disintegration of part of the nearby surface structure during the heating period.

Thermal Conductivity of Single Crystal UO_2 - J. L. Bates

An increase in the thermal conductivity of a nonirradiated UO_2 single crystal above 700 C has been clearly demonstrated by Daniel, et al., in measurements to 1200 C.^(2,3) This increase at high temperatures was initially explained by assuming the conductivity to be the sum of two heat transfer components: (1) a phonon component proportional to $1/T$, and (2) a radiation component proportional to T^3 . Curves a and b in Figure 5.10 show the upper and lower limits of the data extrapolated on a T^3 basis. However, it has been found that the observed increase in thermal conductivity above 700 C is better described by a $T^{4.2}$ relationship (Curve c in Figure 5.10) in the two-component equation.

As-Irradiated

After In-Microscope Heating,
980 C, One Hour

FIGURE 5.9

Irradiated UO_2 Single Crystal (1.44×10^{20} f/cc)
(Reflection and electron micrograph montage of polished and etched surface)

FIGURE 5.10

Thermal Conductivity of UO_2

In order to explain the greater than T^3 dependent increase in thermal conductivity, other processes of heat transfer were considered. The observed thermal conductivity, k_{obs} , can be written

$$K_{\text{obs}} = k_p + K_r + k_e + k_x \quad (1)$$

where k_p is the phonon component, k_r is the radiative component, k_e is the electronic component, and k_x is the excitation process component.

The phonon component contribution to the thermal conductivity, $k_p = 0.03202 + \frac{17.9}{T}$ watt $\text{cm}^{-1} \text{K}^{-1}$, was obtained from the linear portion of a plot of k_{obs} versus $1/T$. The radiation component, $k_r = 4.1 \times 10^{-12} T^3$ watt $\text{cm}^{-1} \text{K}^{-1}$, was calculated using Plank's radiation equation.^{(4)*} The k_p and k_r components are subtracted from k_{obs} to yield a third component, k_{ex} that perhaps represents combined electronic and excitation processes.

*Note: The use of $4.1 \times 10^{-12} T^3$ watt $\text{cm}^{-1} \text{K}^{-1}$ instead of

$2.55 \times 10^{-12} T^3$ watt $\text{cm}^{-1} \text{K}^{-1}$ originally used⁽³⁾ for the radiation component provides a better fit to the data. This is equivalent to a change in the value of the extinction coefficient from 5×10^{-4} to 3×10^{-4} , well within the error of extrapolation.

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A plot of $\ln k_{ex}$ versus $1/T$ is linear. From it an activation energy of 0.83 eV (Figure 5.11) is computed. For single crystal UO_2 the observed thermal conductivity can be expressed as:

(2)

$$k_{obs} = \left(0.03203 + \frac{17.9}{T}\right) + (4.1 \times 10^{-12} T^3) + (7.5 \times \exp(-9580/T)) \text{ watt cm}^{-1} \text{ K}^{-1}$$

as illustrated in Figure 5.12.

FIGURE 5.11

Temperature Dependence of the Excitation Component of the Thermal Conductivity of UO_2 Single Crystal

FIGURE 5. 12

Thermal Conductivity of Single Crystal UO_2 Showing Phonon, Radiative and Excitation Components

It is not possible on the basis of the present evaluation to designate a mechanism for K_{ex} . The activation energy appears large for an exciton diffusion and small for electronic heat transfer.

Thermal Conductivity of Irradiated UO₂ - J. L. Daniel and J. O. McPartland

Thermal conductivity measurements were resumed on a single crystal UO₂ specimen, following the second irradiation to about 10¹⁴ fissions/cc. Study of the data collected after both the first and second irradiations suggests that the substantial conductivity decrease (10 to 20 percent) reported earlier may have resulted in part from physical damage to the crystal sustained during irradiation or handling. One end of the specimen was accidentally broken off prior to the second irradiation. However, data indicate uniform conductivity over the entire length of the specimen prior to breakage, and optical micrographs show no evidence of microcracking either before or after the first irradiation. Measurements and evaluation of the data are continuing.

Irradiation of UO₂ Single Crystals - G. R. Horn

A capsule (GEH-14-360) containing single crystal and bi-crystal cylinders (1/2-inch diameter) of fused UO₂ was fabricated and delivered to the MTR to be irradiated to produce a surface heat flux of approximately 1,000,000 Btu/hr-ft². The test will generate data pertinent to thermal conductivity of single crystals, fission fragment retention and mobility, and recrystallization phenomena (or lack thereof).

Irradiation of Swaged UO₂ to High Exposure - G. R. Horn and J. L. Bates

Preliminary examination of two swaged UO₂ capsules (GEH-14-178 and 179) irradiated to high exposure revealed microstructural features closely resembling those observed previously in similarly exposed capsules. Photographs of as-ground cross sections from one of these capsules (GEH-14-178) are shown in Figure 5.13. The long, thin columnar grains are very similar to those observed in GEH-14-177.⁽⁵⁾ These grains were apparently formed by slow void migration during the more than two years that the capsules were in-reactor. The continued migration of voids has obliterated microstructural features that are presumed originally present and that are normally observed in capsules irradiated for relatively short terms.

It is anticipated that the exposures of GEH-14-178 and 179 are approximately the same as that of GEH-14-177 (~14,200 MWD/T_U). The maximum surface heat fluxes were estimated to be 0.34 x 10⁶ and 0.29 x 10⁶ Btu/hr-ft², respectively. This estimation will be checked during extensive postirradiation examination.

The capsules (GEH-14-178 and 179) are the second and third in a series irradiated to high exposure. (GEH-14-177 was the first.) One additional capsule (GEH-14-180) remains in-reactor. Goal exposure for each capsule is the same (15,000 MWD/T) but the exposure rates differ. Exposure rate (if exposure is sufficiently high) apparently has no significant effect on microstructure.

B-1986

B-1987

B-1988

1/2 Inch
Below Midplane

1 Inch
Below Midplane

1-1/2 Inch
Below Midplane

FIGURE 5.13

Cross Sections of Swaged UO₂ Irradiated to
High Exposure (GEH-14-178)

It was previously reported⁽⁵⁾ that two swaged UO₂ fuel capsules, GEH-14-177 and GEH-3-52, were discharged from the reactor after attaining exposures of 37,000 and 53,000 MWD/T_U, respectively. Mass spectrometric burnup analyses indicate that the original exposure estimates were in error. Revised performance data for these capsules are shown in Table 5.1.

TABLE 5.1
REVISED IRRADIATION DATA FOR HIGH EXPOSURE
SWAGED UO₂ CAPSULES

	<u>GEH-14-177</u>	<u>GEH-3-52</u>
Cladding OD, inch	0.565	0.770
Cladding Thickness, inch	0.030	0.050
Maximum Surface Heat Flux, Btu/hr-ft ²	0.4 x 10 ⁶	0.4 x 10 ⁶
Exposure, MWD/T _U	14,200	23,200
Fission Gas Release, Percent of Total Formed	32.9	81.5
Xe/Kr Ratio	9.95	9.57

A "three dimensional" photomosaic (170X) of GEH-14-177 was constructed to study spatial relationships of microstructural features. The three surfaces comprising the mosaic are reproduced in Figure 5.14. As previously reported,⁽⁵⁾ the two ends of this short (1/2-inch long) sample have significantly different microstructural features. The bottom end exhibits the long, thin grains typical of high exposure UO₂, but the columnar grains in the top end are shorter. Apparently, UO₂ in the central region melted during initial startup and ran down into the bottom half of the capsule. Since a significant portion of fuel was thus removed from the top portion of the capsule, continued irradiation produced temperatures and temperature gradients in that region that resulted in void migration rates insufficient to form the characteristic long thin grains.

UNCLASSIFIED

5. 20

HW-74378

TOP END

LONGITUDINAL SECTION

BOTTOM END

FIGURE 5. 14

"Three-Dimensional" Photomosaic of Swaged UO_2 Rod (0. 56-Inch OD)
Irradiated to High Exposure (GEH-14-177)

Negative 0621852
AEC-GE RICHLAND, WASH

UNCLASSIFIED

Fission Fragment Migration in UO_2 - J. L. Bates, J. A. Christensen and W. E. Roake

The migration of fission fragments and plutonium in UO_2 fuel elements during irradiation has significant implications in burnup analyses, optimum fuel life, and fuel reactivity. Earlier reports (5, 6) of this series discussed experiments that revealed extensive relocation of fission product and plutonium atoms from the points at which they came to rest after the fission or absorption events. UO_2 specimens irradiated to relatively low exposures (0.08×10^{20} fissions per cubic centimeter) show a depletion of fission fragments and plutonium in the high density, nonporous columnar grains. Higher concentrations were observed nearer the cladding and the thermal center. New data from swaged UO_2 irradiated to longer exposures (6.6×10^{20} and 4×10^{20} f/cc) show relatively less segregation of Zr^{95} - Nb^{95} , Ru^{106} , Ce^{144} and plutonium, but more extensive relocation of Cs^{137} to the cooler portions of the fuel near the cladding (Figures 5.15 and 5.16).

The marked differences between the distributions observed in the high and low exposure elements are attributed to the continual change of radial temperature profile during irradiation that modifies the relative effectiveness of the vapor phase zone refining process, diffusion in the thermal gradient, and diffusion to grain boundaries.

TABLE 5.2IRRADIATION DATA FOR HIGH EXPOSURE, UO_2 FUEL CAPSULES

<u>Test Designation</u>	<u>Fuel Rod Dia., cm</u>	<u>Exposures</u>		<u>Heat Rating, watt/cm</u>	
		<u>f/cc</u>	<u>Days</u>	<u>at Startup</u>	<u>at Discharge</u>
GEH-3-52	1.96	6.6×10^{20}	828	775	230
GEH-14-177	1.43	4×10^{20}	435	575	230

FIGURE 5.15

Fission Fragment Distribution in Swaged UO_2
Irradiated to 4×10^{20} Fissions/cc

FIGURE 5.16

Fission Fragment Distribution in Swaged UO₂
Irradiated to 6.6×10^{20} Fissions/cc

UNCLASSIFIED

Identification of the Molten Zone in UO_2 (UO_2 Relocation Experiment) -
G. R. Horn, J. A. Christensen and D. R. deHallas

The microstructural features characteristic of once molten UO_2 were identified by examining irradiated UO_2 pellets that originally contained tungsten marker wires. During initial reactor startup, UO_2 in the central region of the capsule (GEH-4-72) melted and the marker wires in that region sank to the bottom of the melted zone.

Figure 5.17 shows a photograph of one of the pellet faces before irradiation and a cross-section of the same pellet after irradiation. The 0.014-inch diameter by 1/4-inch long marker wires were originally positioned in a spiral array. The absence of wires in the central region after irradiation is clearly evident. The region of the fuel capsule that contains the large tubular voids is presumed to have been quenched from the melt at the time the reactor shut down. Calculations indicate that the fuel was initially melted to a radius beyond the tubular voids. Columnar grain formation in the solid UO_2 then caused a slight improvement in thermal conductivity and therefore a slight decrease in central temperature and some solidification of UO_2 . This process would probably have continued until the entire core was solid had not the reactor been scrammed and all the remaining liquid UO_2 rapidly solidified. A photomosaic of that segment of the cross section that contains the innermost remaining wire is shown in Figure 5.18. The original position of the next wire in the sequence is marked with an "X". Obviously, the microstructure of the UO_2 adjacent to the innermost wire still in place is characteristic of material that was not molten during irradiation, whereas the microstructure of the material in the region of the "X" is characteristic of material that was molten during irradiation.

An enlarged view of the region adjacent to the innermost wire is shown in Figure 5.19. A similar area from a second pellet in the same capsule is shown in Figure 5.20. These figures confirm the previously reported hypothesis⁽⁷⁾ that the molten zone did not include all of the large columnar grain region; but as shown in Figure 5.18, the molten zone did at one time extend beyond the region of large tubular voids. Therefore, the radial limit of melting is hypothesized to correspond to the radial limit of observable sub-grain structure because that is the only structural discontinuity that appears between the innermost wire and the original position of the next wire in the sequence. The initially molten material is characterized by the appearance of sub-grain structure or collections of voids along sub-grain boundaries. The solid material adjacent to the initially molten zone is generally very dense and relatively free of voids because high temperatures and steep temperature gradients in this solid UO_2 cause very rapid void migration rates⁽⁷⁾ during the early part of the irradiation.

Irradiation of the test element was discontinued immediately after reaching full power and there was insufficient time for all the small voids to migrate to sub-grain boundaries; therefore, only a semblance of the sub-grain structure is observable in this UO_2 . In several other irradiated UO_2 specimens (reported previously), in which sufficient irradiation time allowed definition of the sub-grain structure, the position of the initial solid-liquid interface is easily identified. Some typical examples are shown in Figure 5.21.

UNCLASSIFIED

5. 25

HW-74378

INNERMOST WIRE
STILL IN PLACE

BEFORE IRRADIATION

AFTER IRRADIATION

FIGURE 5.17

Cross Sections of Marker Wire Capsule (GEH-4-72)

Negative 0621681
ARCES NICHILAND, WASH.

UNCLASSIFIED

FIGURE 5.18

Photomosaic of Marker Wire Capsule in Region of Innermost Remaining Wire

UNCLASSIFIED

5. 27

HW-74378

FIGURE 5.19

75 X

Microstructure of UO_2 Adjacent to Innermost Remaining Wire
(Pellet #1, GEH-4-72)

UNCLASSIFIED

75 X

FIGURE 5. 20

Microstructure of UO_2 Adjacent to Innermost Remaining Wire
(Pellet #2, GEH-4-72)

UNCLASSIFIED

(GEH-14-194)

(GEH-14-189)

Sintered
 UO_2 Pellets Swaged
(GEH-14-231) Vibrationally Compacted
(GEH-14-233)Fused and
Crushed UO_2 14 wt% UO_2 + 86 wt % ThO_2
(GEH-4-65) 90 wt % UO_2 + 10 wt % ThO_2
(GEH-4-61) UO_2 + ThO_2 FIGURE 5. 21Typical Examples of Microstructure at Initial Solid-Liquid
Interfaces in Irradiated Ceramic Fuels (75 X)UNCLASSIFIED

The radial limit of initial melting generally can be identified by ceramographic examination of a polished and etched surface; it can not, in general, be identified on as-cut or as-ground sections.

A split in the cladding of this fuel element was attributed to thermal expansion of the UO_2 . Experiments at Hanford have shown that UO_2 expands about 9 percent on melting. A diametral clearance of only 0.004 inch (cold) was provided between the 1.25-inch OD pellets and the aluminum cladding (0.095 inch thick). Examination of the cladding shows considerable necking adjacent to the split. There was no apparent bulge or other evidence of excessive internal gas pressure.

Photomosaics of Fuel Elements - J. J. Hauth

Interpretation of phenomena such as void migration, fuel relocation and microstructural changes in irradiated UO_2 fuels has been greatly facilitated by detailed examinations of large photomosaic reproductions of sections of irradiated fuel elements.⁽⁸⁾ Similar representations of non-irradiated fuel elements are being prepared to assist in correlating particle size distribution and fuel quality with effects observed in irradiated specimens.

Sections of vibrationally compacted, fused UO_2 before and after irradiation are shown in Figure 5.22.

High-Rate Densification of UO_2 - D. W. Brite, L. P. Murphy and K. R. Sump

The effects of pressure, temperature, UO_2 surface area, and O/U ratio on the density obtained by high rate densification (Dynapak) of UO_2 were studied. Impact pressures of 200,000 to 400,000 psi were obtained using tungsten carbide die sets, and pressures greater than 400,000 psi were produced with modified Bridgman anvils. Pressures were measured with strain gages, and recorded with a Visicorder.

The effects of impact pressure and temperature on the compacted density of high-rate-densification (HRD) UO_2 are shown in Figures 5.23 and 5.24. Greater densities are achieved with higher pressures and temperatures. By sintering HRD UO_2 for 12 hours at 1700 C, densities of approximately 99 percent TD are obtained with specimens compacted to a wide range of densities, regardless of whether compacted densities were varied by changing pressure or temperature. This effect is shown in Figure 5.25.

An initial UO_2 specific surface area of at least one square meter per gram is required in order that HRD UO_2 compacted at 1200 C and 400,000 psi can be sintered to 99 percent TD (Figure 5.26). Initial surface areas considerably greater than one square meter per gram are needed to achieve densities >99 percent without sintering. The UO_2 powder used in these experiments was prepared by crushing and ball milling sintered UO_2 . High densities were obtained using electrodeposited UO_2 of relatively large particle size. Minus 20 mesh, electrodeposited UO_2 was compacted at 1200 C to 10.84 g/cc by 330,000 psi and to 10.79 g/cc by 250,000 psi impact pressure. A solid pellet was produced in each case.

As-Compacted
Bulk Fuel Density, 90% TD
Particle Size Distribution:
55% 6 + 10
12.5% 10 + 20
12.5% 35 + 65
20% 200

After Irradiation
Bulk Fuel Density, 89% TD
at Assembly
Exposure, 1000 MWD/T_U
(2.5×10^{19} fissions/cm³)
Maximum Surface Heat Flux,
920,000 Btu/hr-ft²
Maximum Heat Rating, 39 kw/ft

FIGURE 5.22

Cross Sections of Vibrationally Compacted Fused UO₂ Fuel Rods

FIGURE 5.23

Effect of Impact Pressure on "As-Compacted" Density of Micronized UO_2
Preheated to 1200 C

FIGURE 5.24

Effect of Preheat Temperature on
"As-Compacted" Density of Micronized UO_2

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FIGURE 5.25

Effect of "As-Compacted" Density on Sintered Density of High Energy Impact Formed Micronized UO_2

FIGURE 5.26

Effect of Surface Area of $\text{UO}_{2.01}$ on Density After High Energy Impact at 1200°C and 400,000 psi

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Introduction of excess oxygen into UO_2 is known to cause a contraction in the lattice, resulting in an increase of density. Excess oxygen also has been shown to decrease, by as much as 900 C, the temperature above which UO_2 can be deformed plastically. Varying the O/U ratio of HRD UO_2 causes the changes of density shown in Figure 5.27. Approximately 99 percent TD was obtained by hydrogen reduction (1200 C) of UO_2 having initial O/U ratios between 2.03 and 2.16. Final densities were approximately one percent lower for HRD UO_2 having an initial O/U ratio of 2.24, probably because of a change in phase from U_4O_9 to UO_2 during reduction. As O/U ratios were decreased below 2.03, compacted densities fell off sharply, because of decreased plasticity in the UO_2 . At least 99 percent TD was obtained by sintering in hydrogen (1700 C) where the original O/U ratios was 2.16 or less.

FIGURE 5. 27

Effect of O/U Ratio on Density of High Energy Impact Formed UO_2

Pulverized (minus 20 mesh) sintered UO_2 scrap that had been oxidized at 140 C to $\text{UO}_{2.01}$ and compacted at 900 C and 200,000 psi to 10.17 g/cc was sintered at 10.84 g/cc during 12 hours in 1700 C hydrogen.

The data presented in Figure 5.24 indicate that a gain in density should result if preheat temperatures greater than 1200 C are used. However, lower densities were obtained because of reduction of the UO_{2+x} by the can material. Densities in the range 90 to 95 percent TD were obtained at 1100 to 1600 C, under conditions in which the O/U ratio was decreased to approximately 2.01. Type 304 stainless steel reduced UO_{2+x} to an O/U ratio less than 2.03 at temperatures below 1200 C.

A sintering study was conducted on HRD UO_2 having a density of 10.17 g/cc. Temperature, heating rate, and soaking time were varied. Sintered densities increased (a) as the heating rates were decreased from 225 to 75 C per hour, (b) as the soaking temperatures were increased over the range of 1400 to 1750 C, and (c) as the soaking time was increased over the range of 1 to 12 hours. Sintering rate was greater during the first few hours of soaking time. The effects of varying these sintering conditions on the density of HRD UO_2 is shown in Figure 5.28.

FIGURE 5.28

Effect of Sintering Conditions on Density
of High Energy Impact Formed and Sintered UO_2

A good sintering program for this material, with consideration given to time and cost as well as density, would be a heating rate of 100 C/hr to 1750 C, followed by 4 hours soaking time.

High-Rate-Densification of Uranium Mononitride -D. W. Brite and L. E. Kuhlken

Uranium mononitride powder was compacted by high rate densification (Dynapak) at 1100 C and 550,000 psi to a density of 14.25 g/cc (99.5 percent TD), using a modified Bridgman anvil technique. Microstructure of the compacted UN is shown in Figure 5.29. Plastic behavior during compaction is indicated. The second phase inclusions visible in the UN have not been identified.

Polished

100 X

FIGURE 5.29

Microstructure of Uranium Mononitride Compacted by High Rate
Densification to 14.25 g/cc (99.5 Percent TD)

High-Rate-Densification of Cermets - D. W. Brite and K. R. Sump

A 50 wt % tungsten - UO_2 mixture was compacted by high rate densification to produce a solid with density of 13.44 g/cc (96 percent TD). Blended minus 65 mesh fused UO_2 and minus 325 mesh tungsten powders, evacuated while being heated to 1100 C in a stainless steel container, were

compacted at 250,000 psi impact pressure. Metallographic examination revealed a uniform dispersion of UO_2 particles in a continuous tungsten matrix. Sintering for 12 hours in 1750 C hydrogen had no measureable effect on density. The microstructure of the cermet is shown in Figure 5.30. Grain growth in the tungsten matrix after sintering is apparent.

15X

100X

500X

500X - Sintered

FIGURE 5.30

Microstructure of High Rate Densified UO_2 -W Cermet

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Graphite Welding - L. E. Mills

A graphite cap was welded onto the end of a 1/2-inch diameter by 1/32-inch wall thickness graphite tube, without the use of intermediate material at the weld interface. Ceramographic examination of the weld revealed a recrystallized weld zone which exhibited a finer crystal size and higher density than the base material. Two graphite tubes, each having an end plug welded on one end are illustrated in Figure 5.31. The third tube and end plug in 5.31 illustrates the shape of the parts before welding. Figure 5.32 is a macrograph of a graphite weld section at 7 X magnification and Figure 5.33 is a graphite weld micrograph at 100 X magnification.

FIGURE 5.31

Graphite Closure Caps Welded to Graphite Tubes

Negative 0621723

7X

FIGURE 5.32
Macrograph of Graphite Tube Closure Weld

Negative 5J1377C

100 X

FIGURE 5. 33

Micrograph of Graphite Tube Closure Weld

The graphite cap and tube are brought together with an initial force of 250 pounds. A 1/60 second alternating current pulse of 16,000 amperes at 18.3 volts is employed to produce the weld. The high current density literally explodes the graphite at the interface between the parts to produce a temporary gap between the parts. An arc exists for 8 to 12/milliseconds. The parts are rapidly brought together, extinguishing the arc, and a pressure of approximately 15,000 psi is developed at the interface to complete the weld.

AEC-GE RICHLAND WASH.

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High-Power Laser Welding and Cutting - L. E. Mills

An evaluation of a high power laser for welding and cutting was performed for Hanford by the General Engineering Laboratory. Individual pulses of the laser beam with a maximum output of 20 joules and with time duration of 2 to 12 milliseconds were impinged on metal surfaces to evaluate penetration characteristics. The large peak power, short time duration laser beam produced cavitation and vaporization of the metal with little or no molten metal remaining to effect a weld. This short pulse laser beam has application for drilling small holes but has little value for welding. A network storage system provided a laser beam pulse of 12 milliseconds with lower peak energy values. Spot welds were produced on the metal surface that were similar in shape to that of a small arc spot weld. Present application of laser beam welding is limited to spot welding with a maximum energy of 50 joules. The development of a continuously operating, high power laser beam is required before the laser can be employed other than spot welding. Present difficulties confronting the development of continuously operating high power lasers are; (1) relatively long power supply charging time, (2) degradation of flash lamps, and (3) heat accumulation in the laser.

Postirradiation Examination of Swaged UO₂ PRTR Fuel Rods - W. J. Flaherty and W. E. Roake

An increase of the thermal conductivity of swaged UO₂ at unexpectedly low bulk fuel temperatures (300 to 500 C) was observed during irradiation of a thermocoupled, 19-rod fuel element in PRTR. Bonding of point contacts between particles because of localized, transient high temperature in the regions of the individual fission events is believed to be the cause of the enhanced heat transfer rate. Normal ceramographic examination of fuel cross sections from this element revealed no changes in microstructure from that of nonirradiated, cold swaged UO₂. Further examination by high magnification optical microscopy and electron microscopy is being conducted to verify the hypothesis stated earlier. Photomicrographs (250X) of cold-swaged UO₂, irradiated to 1200 MWD/T_U in another PRTR fuel element, reveal bonding of particles in the cooler region of the fuel cross-sections (Figure 5.34). The microstructure of nonirradiated cold-swaged UO₂ is shown in Figure 5.35.

Chemical analyses of cladding samples from two PRTR fuel rods irradiated to 200 MWD/T_U revealed 37 to 55 ppm hydrogen. Analyses of cladding from three fuel rods irradiated to 1200 MWD/T_U in the PRTR showed a maximum of 100 ppm hydrogen. Nonirradiated Zircaloy-2 having the same fabrication history contains approximately 35 ppm hydrogen. However, photomicrographs of the Zircaloy-2 cladding from both groups of rods revealed an anomaly; i. e., what appeared to be hydriding approximately three times as severe as the chemical analyses indicated (Figure 5.36). This has not been resolved; however, we tend to believe the results of chemical analyses. Perhaps the appearance of the hydride platelets is "anisotropic", as suggested by studies at SRL, because of the nature of the working of the metal during fabrication.

250 X
Polished and Etched

FIGURE 5. 34
Low-Temperature Bonding of UO_2 Particles

250 X

FIGURE 5.35

Nonirradiated Cold-Swaged UO_2

250 X

Zircaloy-2 from 1200 MWD/ T_U
Exposure
PRTR Fuel Element

250 X

Zircaloy-2 from 200 MWD/ T_U
Exposure
PRTR Fuel Element

FIGURE 5.36

Hydriding of PRTR Fuel Cladding

Negatives 5J1469B, B826, B818

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Hydriding of Zircaloy - G. R. Horn

A purposely defected four rod cluster fuel element was irradiated 120 days at low heat generation rates to study the hydriding characteristics of Zircaloy.

The 12-inch long fuel element contained two Zircaloy-2 clad rods and two low-nickel Zircaloy-2 clad rods. One each of the Zircaloy-2 and low-nickel Zircaloy-2 clad rods was deliberately defected by drilling a 0.006-inch diameter hole through the cladding. The element was irradiated with a surface heat flux less than 200,000 Btu/hr-ft².

The results of metallographic examination at the defect are summarized in Figure 5.37. There appeared to be no significant difference between the defected and nondefected rods. Oddly enough, the defected Zircaloy-2 became hydrided to a lesser extent than did the defected low-nickel Zircaloy-2. In each case the hydrides were concentrated at the outer surface of the cladding. Hydride concentration decreased across the cladding thickness to essentially zero at the inner surface.

Examination of the two defected rods at a point 4 inches from the defect revealed considerably less hydriding than that indicated in Figure 5.37.

Irradiation of AISI-406 Stainless Steel Clad Fuel Rods - G. R. Horn

Examination of an AISI-406 stainless steel clad (0.009-inch thick) fuel element⁽⁹⁾ which reportedly failed after 20 hours of irradiation at a maximum surface heat flux of 520,000 Btu/hr-ft² revealed no evidence of failure. The element was leak tested by immersing it first in liquid nitrogen and then in alcohol. No bubbles were observed during immersion in alcohol and no fission products were detected in a sample of the alcohol. Each rod of the cluster was punctured for fission gas collection and the rate of change in system pressure after puncturing indicated no leaks. The anomalous radioactivity reported in the loop coolant has not been explained, unless it is assumed that the defects are small enough to permit the surface tension of water and alcohol to seal the leak against nitrogen pressure.

A photograph of the element before irradiation and a cross-section after irradiation are shown in Figure 5.38.

Evaluation of the AISI-406 alloy as a candidate fuel cladding material is continuing.

Defected Low-Nickel Zircaloy-2

Outer
Surface

Inner
Surface

Defected Zircaloy-2

Nondefected Low-Nickel
Zircaloy-4

Outer
Surface

Inner
Surface

Nondefected Zircaloy-2

FIGURE 5.37

Hydrides in Zircaloy Cladding (250 X)

UNCLASSIFIED

5.46

HW-74378

As-Fabricated

Cross Section After Irradiation

FIGURE 5.38

AISI-406 Stainless Steel Clad Fuel Element

Negatives B-2388, 062065-6
AEC-GE RICHLAND WASH

UNCLASSIFIED

Irradiation of a Large Diameter UO_2 Rod - G. R. Horn, M. K. Millhollen,
D. R. deHalas and J. J. Hauth

Three 2.33-inch OD UO_2 fuel rods were fabricated for irradiation in a rupture-test loop to study the effect of variations in start-up rate on the fuel structural changes. Irradiation of the first of these (GEH-12-29) was terminated when the element developed a cladding split during the initial reactor start-up.

Postirradiation examination revealed a bulge and the split in the cladding. A photograph of the element after irradiation is shown in Figure 5.39. As indicated, the diameter of the element increased significantly. Circumferential ridges or ripples were observed on the Zircaloy-2 cladding of the irradiated specimen but are not in evidence on two as yet nonirradiated elements. The height of the ridges (0.003-inch maximum) was greatest near the midplane of the element. The ripples (Figure 5.40), thought to reflect cyclic variations in UO_2 bulk density, may have resulted from thermal expansion of the UO_2 , or from an extended series of severe pressure cycles to which the element was subjected during its residence in-reactor prior to start-up. The series of pressure cycles will be reproduced in an autoclave containing a similar element to determine the effect of pressure cycling on the cladding.

Examination of a cross section through the split (Figure 5.41) revealed a central void surrounded by large columnar grains. The appearance indicates that the central UO_2 reached a much higher temperature than expected from the neutron flux previously available in that portion of the loop. The asymmetry of the structural features indicates that a severe diametral variation in neutron flux existed during the short irradiation. Bowing of the element that reduced the coolant annulus on the convex side may have caused a hot spot near the midplane that subsequently overheated the fuel and cladding to cause the bulge and split. Addition of ribs or wire wrap spacers to the large diameter fuel elements will alleviate such hot spot problems.

Fabrication and irradiation data for GEH-12-29 are shown in Table 5.3.

TABLE 5.3
FABRICATION AND IRRADIATION DATA
FOR LARGE DIAMETER UO_2 FUEL ROD (GEH-12-29)

Cladding OD, inches	2.33
Cladding Thickness, inches	0.060
Cladding Material	Zircaloy-2
Fuel	High Energy Impact Formed Natural UO_2
Fabrication Method	Vibrational Compaction
Particle Density, percent TD	98
Bulk Density, percent TD	87.5
Maximum Surface Heat Flux, * Btu/hr-ft ²	~90,000

*At full reactor power (175 Mw) the element was calculated to have generated 320,000 Btu/hr-ft². The element failed at a reactor power level of ~50 Mw. It was not possible to make accurate calorimetric measurements at this power level.

AIM

Association for Information and Image Management

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Silver Spring, Maryland 20910
301/587-8202

Centimeter

Inches

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BY APPLIED IMAGE, INC.

3 of 3

FIGURE 5.39
Large Diameter UO₂ Fuel Rod (GEH-12-29) After Irradiation

FIGURE 5. 40

Oblique View of Large Diameter Fuel Rod (GEH-12-29)
Showing Ripples and Bulge in Cladding

Negative RM-B-1939

AEC-GE RICHLAND, WASH.

UNCLASSIFIED

FIGURE 5. 41

Cross Section of Irradiated Large Diameter
UO₂ Fuel Rod (GEH-12-29)

Negative B-2314
AEC-GE RICHLAND, WASH.

UNCLASSIFIED

Prototypic Advanced PRTR Fuel Element Cladding - L. E. Mills and
M. K. Millhollen

Large diameter, single rod ceramic fuel elements may have potential advantages in an application such as fueling the PRTR. However, Zircaloy cladding tubes of large diameter (for example the 2.328-inches OD by 0.060-inch wall tubing) will collapse when subjected to 1100 psig external pressure at 400 C. Cladding of the same dimensions will not collapse under these conditions when connected by five equally spaced ribs to a 3.068-inches OD by 0.060-inch wall Zircaloy outer support tube (Figure 5.42). The latter arrangement did not collapse at pressures below 1500 psig at 400 C.

FIGURE 5.42

Samples of Reinforced and Normal Zircaloy-2
Tubing After Autoclaving at 1100 psig and 400 C

The cladding assembly fabricated from the two pieces of tubing plus spacer ribs lends itself to the design of an integral fuel element-process tube combination. In this arrangement, the inner tube would contain the fuel while the channel between the two tubes would contain flowing coolant water. A short sample of such a fuel element-process tube combination is shown in Figure 5.43.

FIGURE 5.43

Prototype One-Piece Fuel Element and Process Tube

This cladding is a welded fabrication employing the high voltage electron beam welding process. The spacer ribs are welded to the inner tube fuel cladding by impinging the electron stream tangent to the tube at the base of the rib. The spacer ribs are welded to the outer support tube by completely penetrating the tube from the outside with a radial electron stream.

Remote Fabrication Studies - R. C. Smith and R. F. Klein

The initial phase of the remote fuel fabrication study is complete. Several new developments and concepts for remote fuel element fabrication were evolved. More important were the development of UO_2 densification by vibrational compaction via energy transmission through a steel beam and the successful, remotely controlled welding of Zircaloy clad fuel rods containing gamma active "recycled" UO_2 .

Probably the more important concepts evolved during these studies were preassembling and pretesting of fuel element components. The objective was to reduce to a minimum the equipment and operations in the contaminated or "hot" work zone. Under the system evolved all of the vibrational compaction equipment except a small portion of the steel beam is maintained in the "cold" operating area where contact maintenance is used. Of approximately 25 welds necessary in fabricating Mark II-C fuel elements, all but four are made and tested before the "hot" material is added; most of the welding equipment is kept outside the contaminated zone. Autoclaving and the major portion of quality control testing is also accomplished before the recycle fuel is added. The amount of equipment necessary in the "hot" zone is therefore greatly reduced, as are the complexity and number of remote operations.

During these studies a semi-remote fabrication pit was constructed and used for most of the development work accomplished.

Construction of a top extension over one-half of the cell shown in Figure 5.44 is underway and should be completed in November. Manipulators are ordered and delivery is expected by the end of the year. TV cameras are now being used in lighting experiments and for remote "seeing" practice, camera orientation, and equipment placement.

Figure 5.44 shows the equipment which has been assembled. The vibrational compaction exciter and control console are located outside the cell wall and the vibrational energy is transmitted via a steel beam to the hanging fuel element. The two inserts show the remotely controlled welding turntable and the inert gas welding chamber with a weld-test cladding segment in welding position. The turntable is installed and the welding chamber is ready for installation when the cell enclosure is complete.

Figure 5.45 shows the final closure sequence for a Mark II-C fuel element, as accomplished by remote handling. These pictures are of a dummy element used for weld test purposes; the prototype element is approximately 8 feet long. The Zircaloy end caps will have been inserted immediately after fuel compaction and welded by TIG, time-sequenced,

FIGURE 5. 44
Remote Controlled Fuel Fabrication Operations

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5.55

HW-74378

A. Separated

B. Inserted

C. Completed

FIGURE 5.45
Closure Welding Sequence

Negative O621804-1
ATCCE NICHLAND, WASH

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remote controlled welding in the atmosphere control chamber shown in Figure 5.44. Figure 5.45a shows the two fuel components. The larger component consists of the preassembled inner rod, outer tube and bottom end hanger. The smaller component is the preassembled inner tube and top end hanger. After completing the three end cap closure welds, these two components are slipped together as shown in Figure 5.45b and the final end hanger weld made in the position shown in Figure 5.45c. With the illustrated end hanger modification the final TIG, time-sequenced, fusion weld can be made by rotating the element on the same turntable used for the end closures.

REFERENCES

1. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, April, May, June, 1962, HW-74377, p. 5.1. 1962 (Secret)
2. Daniel, J. L., J. Matolich and H. Deem. Thermal Conductivity of UO_2 , HW-69945. 1962.
3. Cadwell, J. J. Fuels Development Operation Quarterly Progress Report, April, May, June, 1962, HW-74377, p. 5.5. July 20, 1962. (Secret)
4. Bates, J. L. "Thermal Conductivity of UO_2 Improves at High Temperatures", Nucleonics 19, No. 6: 83-87. 1961.
5. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, January, February, March, 1962, HW-72347. April 16, 1962. (Secret)
6. Bates, J. L., J. A. Christensen and W. E. Roake. "Fission Products and Plutonium Migrate in Uranium-Dioxide Fuel", Nucleonics 20, No. 3: 88-90. 1962.
7. Cadwell, J. J. Fuels Development Operation, Quarterly Progress Report, April, May, June, 1962, HW-74377. July 20, 1962. (Secret)
8. Cadwell, J. J. Fuels Development Operation Quarterly Progress Report, April, May, June 1962, HW-74377. p. 5.30-5.33. July 20, 1962. (Secret)
9. Cadwell, J. J. Fuels Development Operation Quarterly Progress Report, April, May, June, 1962, HW-74377. July 20, 1962. (Secret)

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