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HW-69085

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**QUARTERLY PROGRESS REPORT
FUELS DEVELOPMENT OPERATION
JULY, AUGUST, SEPTEMBER, 1960**

OCTOBER 15, 1960

HANFORD LABORATORIES

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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This document consists
of 148 pages.

QUARTERLY PROGRESS REPORT
FUELS DEVELOPMENT OPERATION
JULY, AUGUST, SEPTEMBER, 1960

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October 15, 1960

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

Work performed under Contract No. AT(45-1)-1350 between the
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The present Quarterly Report is the continuation of a series issued by the new Fuels Development Operation. Reports in this series combine portions of the quarterly reports by the former Metallurgy Research and Fuel Technology Sub-Sections. Work reported includes research conducted by the Physical Metallurgy Operation, and research and development conducted by Fuel Design, Fuels Fabrication Development and Ceramic Fuels Development Operations. Studies formerly reported by the Radiometallurgy, Metallography, and Welding and Corrosion Units, in addition to portions of the Fuels Technology work, are reported elsewhere.

Reports previously issued as Metallurgy Research and Fuel Technology Quarterlies are listed in the first report of the present series, HW-46091.

QUARTERLY PROGRESS REPORTS - FUELS DEVELOPMENT

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HW-62656	July, August, September, 1959.
HW-64863	October, November, December, 1959.
HW-66094	January, February, March, 1960.
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PHYSICAL METALLURGYMECHANICAL AND PHYSICAL PROPERTIES OF IRRADIATED MATERIALS

Isochronal annealing of irradiated iron, copper, and molybdenum has been extended to temperatures of 550, 650, and 875 C. The resistivity of irradiated copper did not completely recover even with annealing temperatures of 450 C. This effect is thought to be due to the relatively large amount of transmutation products present in copper after irradiation.

Irradiation of a number of nickel base alloys at 650 C in a Hanford graphite channel resulted in very little obvious deterioration of the specimens. Weight gain measurements show some differences amongst the materials examined.

A prototype capsule for the measurement of creep in-reactor is still operating satisfactorily after more than 9,000 hours. The instrument facility for in-reactor testing has reached the stage of acceptance testing.

MECHANICAL AND PHYSICAL PROPERTIES OF UNIRRADIATED MATERIALS

A precision pole figure goniometer which uses spherical specimens has been calibrated and put into operation. This unit is being used to study textures in alpha uranium.

Quenching of a Zr - 2Ni - 2Sn alloy from above 900 C results in the formation of α' by a shear transformation process. Strengthening of this alloy is found to be independent of the presence of the α' phase.

FUNDAMENTAL STUDIES

A collector irradiated in close proximity, but not in contact with a uranium dioxide film was found to have U_3O_8 deposited on it. Repetition of the same process in an evacuated capsule resulted in the deposition of UO_2 on the collector.

Two uranium swelling capsules continued to operate satisfactorily during the quarter. A third capsule having a modified capsule geometry was prepared for insertion into the reactor. Inspection of irradiated uranium after annealing to as high as 700 C has revealed a close correlation between the void fraction and immersion density measurements.

Differences have been observed in the recovery of X-ray lattice parameters for molybdenum specimens irradiated to 5×10^{19} nvt and 6.5×10^{18} nvt. Observed annealing stages have been interpreted in terms of trapped interstitials and interstitial clusters which are produced in the molybdenum by neutron irradiation.

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METALLIC FUELS DEVELOPMENT

NEW FUELS DEVELOPMENT

Nine Zircaloy-2 clad rods irradiated in NaK capsules were recently examined. Five were found to have cladding failures. The failures occurring in the 0.020-inch clad specimens were ductile, while the one found in a 30-mil clad specimen was of a brittle nature. Metallographic comparison of irradiated and unirradiated clad rods did not show a correlation of clad thickness with failure. The metallographic sections examined, however, did not necessarily represent sections through the points of failure initiation.

Post-irradiation examination of two thermocoupled Zircaloy-2 clad rods showed evidence of only one having failed. No evidence was found on either rod which indicated that failure had initiated in the stainless steel to Zircaloy-2 braze joints.

Wear testing of 1008 steel support material against 321 stainless steel, resulted in severe scratching of both the stainless and 1008 surfaces. The use of water soluble charging oil proved to be of some value in reducing galling tendencies.

Tests conducted on unirradiated coextruded Zircaloy-2 cladding show that the state of stress significantly effects the mode of fracture. The average circumferential strain decreased from 16 per cent to 5 per cent in changing the technique of loading from internal pressurization to compression of silicon rubber inside the cladding tube.

A mathematical model of a plastically deformed fuel element has been developed to aid in establishing the effects of fuel geometries and clad thicknesses on directing the fuel swelling in Zircaloy clad uranium fuel elements.

Constant load thermal cycling tests have resulted in strain rates comparable to the isothermal strain rates found at stresses an order of magnitude higher. Test results for a specimen cycled at 750 psi load are reported.

FABRICATION DEVELOPMENT

An "extrusion closure" is being developed. A Zircaloy ring is placed in a recessed groove at the end of a coextruded tube and welded in place by electron beam. This ring is subsequently back extruded to bond the cap and clad. This process must be extended by finding a bonding medium which will permit bonding of the cap to the substrate uranium. Attempts to complete the bond with various metallic brazes are reported.

In another approach, a cap is prepared with projecting rings on the OD and ID base and with brazing material placed between those rings. Such a "projection cap" has been successfully bonded to the fuel cladding by projection welding. Attempts to complete the closure by further heating are continuing.

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The attachment of all-steel supports to Zircaloy-clad fuel elements has been achieved by means of "riveted" Zircaloy studs.

In the development of a heat treatment for the KER single tube element, quench rates varying from 7700 C/min to 3700 C/min have been studied. The best macro and micro structures obtained were observed at the 3700 C/min quench rate.

The arc melting facility recently installed is being utilized to prepare a series of 13 zirconium base alloys.

Coextruded fuel rods have been fabricated into shapes with increased surface areas by means of a specially designed "turks head".

FUEL EVALUATION

Examination of an inner component of a KER tube/tube fuel element which failed at 3200 MWD/T is under way. The fuel element failure was a result of a longitudinal clad split. Striations extended longitudinally from each end of the split. Examination results indicate that the failure was the result of localized plastic deformation of the clad. Post-failure corrosion along internal cracks within the fuel resulted in propagation of cracks into the clad in areas adjacent to the clad failure. Evidence was also found that the inner bore in the nonfailed portion of the fuel element has distorted appreciably from its original circular cross-section.

The first attempt to irradiate fuel elements with brazed end closures was terminated by premature failure indications. Subsequent examination reveals no evidence of a failure in the brazed elements.

Work is continuing on alloys other than the 5 w/o Be-95 w/o Zircaloy-2. A composition consisting of 12 w/o Fe plus 4 w/o Be plus 84 w/o Zircaloy-2 appears promising and will be further investigated.

Examination of the fourth in-reactor rupture test element has been completed. This previously unirradiated tubular fuel element operated in-reactor in a high temperature water environment for a period of 30 minutes after failure was initiated. Examination results indicate that the in-reactor corrosion rate was approximately four times faster than in a comparable ex-reactor experiment. Corrosion appears to have been accelerated along the fuel cladding interface.

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CERAMIC FUELS DEVELOPMENT

FABRICATION DEVELOPMENT

Special Fuel Elements and Components for PRTR Start-Up Tests. Three PRTR 19-rod fuel elements containing lead, one PRTR 19-rod cluster containing lead-cadmium, and one PRTR swaged UO_2 19-rod cluster having detachable fittings were fabricated for PRTR design tests and critical tests.

Hot Swaging of UO_2 . Electrodeposited UO_2 crystals in Zircaloy cladding were cold swaged to 88 per cent TD and hot swaged to 91 per cent TD. Analyses of hot-swaged (850 C) Zircaloy cladding have revealed an increase in hydrogen and oxygen content. No protective atmosphere was used during swaging of these rods. Fuel rods for a 19-rod cluster PRTR fuel element were hot-swaged (850 C) to a bulk UO_2 density of 93 per cent TD.

Vibrational Compaction Studies. A mixture of fused oxide grain consisting of 62.5 w/o (-6 +12 mesh), 17.5 w/o (-35 +65 mesh) and 20 w/o (-200 mesh) was vibrationally compacted in Zircaloy tubes to a bulk density of 90.5 per cent TD. Uranium carbide grain was vibrationally compacted into stainless steel tubes to a bulk density of 85.5 per cent.

High-Energy Impact Forming. Sealed capsules of UO_2 07 were high energy impacted at 1300 C and at 800 C. The UO_2 bulk densities were 94.2 per cent TD and >99 per cent TD, respectively. The lower density apparently resulted from the release of oxygen at 1300 from the nonstoichiometric UO_2 and the resulting microporosity, revealed by electron micrographs.

Welding Fabrication Development. A tungsten arc inert gas welding process was developed for remote welding of closures on Zircaloy tubes filled with recycled UO_2 fuel.

Resistance seam welding of spacer ribs to Zircaloy tubes resulted in excessive crevice formation on both sides of the weld, between tube and rib.

High frequency resistance welding of spacer ribs to Zircaloy-tubes showed a definite improvement in weld quality as the welding rate was increased.

FUEL EVALUATION

Prototypic PRTR Fuel Element Irradiation. GEH-12-4. A prototypic, three-foot-long, swaged 19-rod, Zircaloy clad fuel assembly was irradiated in the ETR. The test was terminated after 24 days because of fission product activity in the coolant.

Irradiation of Vibrationally Compacted Nonhomogeneously Enriched UO_2 Fuel Cluster. GEH-4-44. A 4-rod cluster fuel element was irradiated in the MTR. Each rod contained UO_2 fuel of the same average U^{235} content, but differed in the method of enrichment.

Swaged UO₂ Capsule Irradiation. Swaged UO₂ capsules have attained an estimated exposure of 17,500 MWD/T.

Thin-Wall Stainless Steel Clad Fuel Rods - VBWR-2. Uranium dioxide fuel rods vibrationally compacted in thin-wall (0.015-inch, 0.010-inch, 0.008-inch) stainless steel tubes were prepared for irradiation in the VBWR.

Irradiation of 7-Rod Cluster, Purposely Defected, Swaged UO₂ Fuel Element - GEH-12-20. A 7-rod cluster consisting of swaged UO₂ rods clad in Zircaloy-2 and Zircaloy-4 was irradiated for 18 days. Two each of the Zircaloy-2 and Zircaloy-4 fuel rods were defected with 0.006-inch holes.

MTR Zircaloy-2, Zircaloy-4 Defect Rod Test - GEH-4-54. Uranium dioxide was vibrationally compacted in a tube consisting of two butt welded sections of Zircaloy-2 and Zircaloy-4. The fuel rod was defected at the center of the weld with a 0.006-inch hole. The rod will be irradiated for direct comparison of hydride formation in Zircaloy-2 and Zircaloy-4.

UO₂ Fuel Element Defect Tests - HD-1, HD-2, HD-3. Four-rod cluster, UO₂ fuel elements, purposely defected, were prepared for long term irradiation in a Hanford reactor.

Irradiation of Purposely Defected, Swaged UO₂ Elements - GEH-4-39, 4-45, 4-52, 4-53. Irradiation and examination of four purposely defected, swaged UO₂ fuel rods was completed.

BASIC STUDIES

High Temperature Behavior of Irradiated UO₂. Studies were conducted of the high temperature behavior of UO₂ irradiated to various exposures. The melting point increased with exposure to a maximum of 2915 C at about 1.4×10^{19} fissions/cc.

Thermal Conductivity. The thermal conductivity curve of a cold pressed and sintered specimen was intermediate between the two curves established for the isostatically pressed specimens and for the extruded specimens.

Electron Microscopy. The JEM 5Y/6A electron microscope was received and installation started.

PHYSICAL METALLURGYMECHANICAL AND PHYSICAL PROPERTIES OF IRRADIATED MATERIALSEffects of Irradiation on Thorium and Thorium Alloys - R. D. Leggett

No new information has been obtained during this quarter.

Effects of Irradiation on Nonfissionable Materials - J. J. Laidler and K. R. Wheeler

Neutron Damage to Metals. The purpose of this program is to advance the theory of irradiation damage to metals by neutrons. Copper, nickel, titanium, zirconium, iron molybdenum, and type 347 stainless steel specimens were irradiated at Brookhaven, Hanford, and the MTR under various exposure and temperature conditions. Post-irradiation measurements of mechanical and physical properties which were initiated at KAPL will be completed at HAPO.

Isochronal annealing treatments of iron, copper and molybdenum have been extended to 550, 650, and 875 C, respectively. Annealing of titanium was terminated at 575 C. A two-stage recovery over a limited temperature range has been observed in irradiated copper. Significant microhardness decreases in copper occurred at 375 and 425 C. There has been no appreciable decrease in the electrical resistance of irradiated copper after annealing at 450 C, and none is expected to occur at higher temperatures since the added resistance is largely due to transmutation products. Therefore, the microhardness recovery is thought to occur by a complex dislocation reaction, and no definite mechanism will be postulated before isothermal recovery is studied.

Recovery of the initial hardness increase in molybdenum was complete after isochronal annealing at 825 C for specimens with exposures of 4.4×10^{18} and 1.5×10^{20} nvt. In both cases microhardness has recovered to a value lower than the as-irradiated microhardness. It is possible, however, that the recovery reaction at 825 C involves carbon atoms. If the hardening were due to an aging process followed by over-aging, the recovery stage at 825 C may be an effect of accelerated agglomeration of carbon atoms.

Post-annealing X-ray studies have been completed on isochronally-annealed zirconium and titanium specimens. Although microhardness and electrical resistance measurements indicated that damage recovery was complete in both irradiated zirconium specimens, a small amount of lattice distortion remained in the form of a shrinkage in the c_0 lattice parameter. The a_0 parameter for both specimens recovered completely. Consequently, the line width of the (220) reflection in both specimens recovered to the unirradiated value, but the line width of the (302) reflection showed a retained broadening of 22 and 10 per cent, respectively, at exposures of 4.7×10^{18} and 1.5×10^{20} nvt.

Isochronal annealing of irradiated titanium has graphically illustrated the pronounced effect of interstitial impurities. Titanium specimens used in this program were cut from three ingots varying widely in nitrogen and oxygen content; carbon and iron contents were nearly constant. Since nitrogen and oxygen have the greater effect on mechanical properties, anomalous behavior is to be expected among specimens cut from different ingots. Due to limitations in specimen availability, specimens used for isochronal annealing varied in composition. The unirradiated control specimen and the specimen irradiated to 1.5×10^{20} nvt contained 0.0014 and 0.087 weight per cent nitrogen and oxygen, respectively. Upon annealing, the irradiated specimen exhibited complete recovery in lattice parameters, microhardness, and electrical resistance. The specimen irradiated to 1.1×10^{19} nvt contained 0.004 and 0.021 weight per cent nitrogen and oxygen, and naturally recovered to lower values of microhardness and electrical resistance. Specimens to be used for the isothermal annealing have been selected so that there is no effect of varying impurity content.

A considerable effort has been directed toward construction of an isothermal annealing fixture. Since two of the independent variables in kinetics studies are temperature and time, it is imperative that the time involved in heating to temperature and cooling be minimized; for an anneal of five or ten minutes duration, the effect is significant. The fixture which is to be used involves an evacuated tube which is immersed in molten lead for heating and flowing water at 20 C for cooling. The furnace may be operated under dynamic vacuum or static helium pressure. Best results have been obtained by heating to temperature under 20 psi helium pressure, pumping off the helium, and conducting the remainder of the anneal in a dynamic vacuum of 10^{-5} mm Hg. Immediately prior to cooling, the system is isolated, and helium is again introduced into the tube. Heating times to 500 C average 40 seconds; cooling to 50 C requires about 30 seconds.

Strain aging studies of irradiated iron reveal that solute redistribution occurs concurrently with the accumulation of radiation damage. This is manifested by a loss in aging response due to nitrogen after moderate exposures. The process is thought to involve a breakup of nitrides by spikes and the trapping of nitrogen atoms, as well as other solute atoms at complex dislocation defects formed by irradiation. This mechanism is supported by tempering curves performed on specimens having differing degrees of irradiation. The tempering zones associated with the formation of coherent and incoherent precipitates from supersaturated solution are present after low exposures but are absent after exposures in the range of 10^{20} nvt.

Nickel Base Alloys Irradiations. From thermodynamic considerations advanced reactors must operate at higher temperatures. The combined effects of temperature and reactor environment impose rather severe restrictions on the materials available for structural use. Ex-reactor properties such as strength at temperature, corrosion resistance, and fabricability indicate that various stainless steels and nickel base alloys meet general requirements. Final selection of the material then depends upon its performance under conditions peculiar to the operating reactor.

In support of in-service studies of candidate materials, a group of nickel base alloys, iron-chromium-aluminum alloys and several stainless steels are being irradiated in ventilated graphite capsules in a dry hole of a Hanford Reactor. The first short exposure capsule, charged to determine any gross difference in reaction rates of the various alloys with the gaseous reactor atmosphere at near 650 C has been discharged and the washer shaped samples examined. Visual inspection revealed no excessive reactions. A possible criterion for evaluation might be a weight gain per unit area of washer surface. Table 3.1 gives such a comparison for several of the more interesting alloys. Values represent averages of 2 to 3 samples.

TABLE 3.1

TYPICAL SURFACE REACTIONS OF SELECTED ALLOYS

<u>Alloys</u>	<u>Surface Area in²</u>	<u>Avg Wt Gain (gms)</u>	<u>Surface Reaction gms Gained/in²</u>
AISI 406 SS	0.868	0.0035	0.0040
304L SS	0.939	0.00056	0.00060
F-Cr-Al	0.868	0.00075	0.00085
Hastelloy X	0.939	0.00096	0.0010
Inconel X	0.939	0.0008	0.00089

A second capsule containing tensile specimens of the nickel base alloys, Inconel, Inconel 702, Hastelloy X, and Hastelloy R-235, has been discharged after an exposure of approximately 3×10^{20} nvt thermal. Tensile data on these alloys is now being obtained in a shielded facility in Building 326.

A third capsule containing tensile specimens of the nickel alloys mentioned above continues to accumulate exposure time.

In addition, a fourth capsule containing tensile specimens of Ferral (Fe-Cr-Al) alloy, and AISI 316L, 347, and 406 stainless steel alloys has been charged into the reactor.

In-Reactor Measurements - L. J. Chockie, J. J. Holmes, and J. C. Tobin

The physical and mechanical properties of structural materials are significantly altered upon exposure to a fast neutron flux. The objective of the in-reactor measurements program is to determine the physical and mechanical properties of structural materials during fast neutron irradiation. Currently, a study of the in-reactor creep properties of Zircaloy-2 is being conducted. A prototype creep test capsule has been constructed and used to determine creep rates in a Hanford reactor.

Creep Measurements. As reported previously, Zircaloy-2 creeps faster in-reactor than ex-reactor. The ratio of the radiation enhanced in-reactor creep rate to the ex-reactor rate has been shown to increase with temperature at 30,000 psi stress in the range 500 F to 550 F. In order to determine the effect of stress on the radiation enhanced creep rate the stress on both the in-reactor and ex-reactor specimen was raised from 30,000 to 33,000 psi at 550 F. Nine hundred hours are required to establish a reliable creep rate with the first generation capsule. Shortly after the stress was raised extensive construction work was initiated in the area of the prototype instrumentation. The vibration from air hammers and other heavy equipment, together with a series of helium pressure losses has made the accumulation of nine hundred hours of uninterrupted testing impossible, therefore, no new creep rate can be reported at this time.

The prototype capsule appears to be operating satisfactorily after more than 9,000 hours of elevated temperature in reactor use. The capsule was examined for leaks by checking the exhaust line for helium flow with a mass spectrometer helium leak detector. No flow was detected, which indicates the numerous radiation resistant ceramic seals in the capsule are still helium tight and no leak has developed in the loading bellows. All heaters and thermocouples are still operating.

The operation of the prototype capsule for more than 9,000 hours proves the durability of this instrument and its potential for determining long-time in-reactor creep data. The basic design concepts of the prototype capsule will be retained in future HAPO creep capsules. Improvements in the strain measuring apparatus and heat transfer characteristics must be made. The weak point of the prototype capsule is its inability to measure creep rates that are rapidly changing. Transient creep occurring with flux changes, stress changes or temperature changes may well be the limiting factor in determining the suitability of a structural material for reactor core applications.

Capsule and Instrument Development. Acceptance test procedures are being written and initial acceptance tests conducted on an instrumentation facility at the reactor. The facility is designed to provide the major part of the instrumentation, electrical input and helium supply required for in-reactor mechanical property test capsules. Included in the facility are sixteen identical channels of pressure and temperature controlling, and recording instrumentation, cooling apparatus, and direct current resistance bridges for strain measurement. These instruments are shown in Figure 3.1. Also included is an ex-reactor capsule test rack which is capable of simulating water flow and temperature of any test hole in which a capsule is inserted. The ex-reactor capsule rack will be used for both comparing capsule operation with standard testing equipment and to simulate in-reactor conditions so a direct comparison of in-reactor and ex-reactor material behavior can be made.

A 48-channel digital data recording system capable of recording test data from the variable permeance transducers is being manufactured. The digital system features a charge printer and tape punch which can record data

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FIGURE 3.1

A Partial View of the Instrumentation Facility at the K-West Reactor

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from each channel at a maximum rate of four points per hour and minimum rate of one point per day. The data punched in digital form will be analyzed with an IBM 709 computer. Automatic data analysis will greatly simplify the work involved in establishing confidence limits, evaluating activation energies, determining creep rates and evaluating other creep parameters of interest. The digital data processing system will also incorporate control and readout equipment for mechanical extensometers; this equipment, however, will not be capable of automatic operation in the 48 channel system. A picture of the data processing system is shown in Figure 3.2.

In-Reactor Creep Theory. Creep of metals above one-half the absolute melting point is generally considered to be the result of diffusion controlled dislocation climb. Steady state creep rates in the dislocation climb or rapid recovery range can be expressed as:

$$\dot{E}_s = ACov$$

where

$\dot{E}_s$ = steady state creep rate

A = constant

C_o = vacancy concentration

v = jump frequency.

Gunther Shoeck has modified the above equation to apply to the case where excess vacancies created by neutron atom collisions participate in deformation processes. Shoeck derives the expression

$$\dot{E}_i = \dot{E}_s \left(1 + \frac{a^2 \alpha f}{Dp} \right) \quad (1)$$

where

$\dot{E}_i$ = in-reactor creep rate

$\dot{E}_s$ = ex-reactor creep rate

a = atom spacing

α = atomic fraction of vacancies produced per unit flux

p = atomic fraction of vacancy annihilation sites

f = flux

D = diffusion coefficient

Equation (1) is valid at temperatures where creep is a consequence of dislocation climb and the concentration of vacancies produced by neutron-atom collisions is comparable to that produced by the combined effects of thermodynamics and plastic flow. The metallurgical structure of the ex-reactor and in-reactor specimens must also be identical in order for Equation (1) to be valid. Figure 3.3 is a graphical representation of Shoeck's equations for Zircaloy-2. Ex-reactor creep data has shown dislocation climb governs the creep of Zircaloy at temperatures as low as 288 C. The limiting climb temperature has not been established but recovery studies lead one to believe that 280 C is close to that limit. If climb cannot occur below 280 C

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FIGURE 3.2
Forty-Eight Channel Data Processor

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then portions of the curves of Figure 3.3 falling below 280 C are meaningless and measurable irradiation induced effects will be observed only at flux levels of greater than 5×10^{12} nv. Radiation induced creep rates have been observed in Zircaloy-2, however, at temperatures as low as 260 C which increased with a temperature increase to 288 C. This could be explained by assuming climb mechanisms do not abruptly cease at low temperatures, but their participation in creep gradually diminishes with decreasing temperatures. The effect of irradiation on the climb contribution to creep at 260 C is then sufficient to increase the over-all creep rate; at 288 C, the increased participation of climb results in an increased radiation enhanced creep rate. Observed radiation induced creep rates are considerably greater in magnitude than indicated in Figure 3.3; but values of α , p , D , and the neutron flux for the in-reactor creep experiment could be assumed which would correct the curves to fit the observed data.

MECHANICAL AND PHYSICAL PROPERTIES OF UNIRRADIATED MATERIALS

Preferred Orientation in Uranium - D. L. Gray

Orientation of extruded uranium rods and tubes with various fabrication and heat treatment histories is being determined by several methods. Growth index values, "inverse" pole figures and pole figures are being obtained. A method for determining pole figures from a spherical specimen was developed this quarter. Use of a spherical sample has the advantage that pole density variations with pole orientation for a given set of hkl planes can be measured with one sample.

A spherical specimen with a stem on it is machined from the rod, tube or sheet for which orientation is to be examined, as illustrated in Figure 3.4. The extrusion axis, radial directions or tangential directions (EA, R_1 , and R_2 , T, D) are marked on the stem in order to refer it to the material from which it was machined. The sphere is mounted in the pole figure goniometer (pfg) shown in Figure 3.5a. The pfg has three axes of rotation and is capable of rotating through angles, α , ϕ , or δ as illustrated in Figure 3.5b. The specimen is placed in the pfg so that the axes EA and R_1 coincide with Y_0 and Z_0 . It is mounted so that the center of rotation of the sphere coincides with the center of rotation of the pfg. The assembly is then mounted on an XRD diffraction unit, in the manner illustrated in Figures 3.6 and 3.7. The diffractometer is set at $2\theta = 0$ and the goniometer translated along y_0 so that the end of the sphere is tangent to an alignment bar, which determines the midplane of the X-ray beam. This is shown in Figure 3.7. When the alignment is completed, the bar is replaced by a solar slit and the diffractometer is rotated to a selected value of 2θ . At any value of 2θ , the point P remains at the midpoint of the incident X-ray beam, at the center of rotation of the diffractometer, as shown in Figure 3.7. Since the sphere is mounted with its center at the center of rotation of the pfg, a constant surface area will always be in the X-ray beam as the sphere is rotated through angles of α or ϕ at any preselected and fixed value of 2θ .

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FIGURE 3.3

Ratio of In-Reactor to Ex-Reactor Creep Rate at Various Flux Levels Computed from Shoeck Equation

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FIGURE 3.4

Illustration of Types of Specimens Used for Pole Figure Studies, and the Relationships to the Rod or Tube from Which They are Cut. Schematic, Not to Scale

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(b)

(a)

FIGURE 3.5

- (a) Pole Figure Goniometer
 (b) Sphere, Showing Center of Rotation C. (Sphere is rotated about the stem by an amount ϕ ; at the same time, the stem is rotated about C by an amount α in the Y_0-Z_0 plane. This results in spiral motion with respect to the point P.)

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FIGURE 3.6

Pole Figure Goniometer and Uranium Sphere Mounted on XRD,
with Alignment Bar

FIGURE 3.7

XRD Goniometer is Rotated to a Diffracting Position, $2\theta = 60.42^\circ$
X-Rays are Diffracted from the 131 Planes Parallel to the Tangent
Plane at a - b, and Are Received by the Geiger Counter

To determine a pole figure for a given family of planes, hkl , the XRD goniometer table must first be rotated to the proper value of 2θ . This is the diffraction angle which satisfies the Bragg relationship $n\lambda = 2d \sin \theta$ for the hkl family in question. The symbol λ is the wave length of the incident x radiation, d is the spacing between parallel hkl planes and θ is the angle which the incident radiation must make with the hkl planes in order for diffraction to occur. It is also the angle between the diffracted beam and the hkl planes from which diffraction occurs. The X-ray diffractometer is designed to take advantage of this angular relationship and, the geometry is such that at a given setting of θ the diffracted intensity received by the Geiger counter comes from those hkl planes oriented parallel to the plane tangent to the sphere at point P and whose diffraction angle is θ . In other words, the diffracted intensity received by the counter is for hkl planes satisfying the selected θ angle whose poles lie parallel to the Y_0 axis. The diffracted intensity is proportional to the volume of material in the X-ray beam which has planes in the diffracting position, and gives a measure of the pole density in the orientation parallel to Y_0 . By rotating the sphere in the pfg, various pole orientations can be brought into the diffracting position, and the corresponding pole density can be measured. A pole figure is just a plot on a stereographic projection of pole density as a function of pole orientation for a given set of planes, hkl . With the pole figure goniometer the necessary data for plotting a pole figure is obtained from a spiral scan of the surface by simultaneously rotating the sphere through the angles α and ϕ , at rates of $\frac{d\alpha}{dt} = 1^\circ$ per minute and $\frac{d\phi}{dt} = 36^\circ$ per minute, and recording the diffracted intensity on a strip chart as a function of α , ϕ . That these motions cause the sphere to move in a spiral can be seen by examining Figures 3.8 and 3.9, which show that as the sphere is rotated about the assembly and angle ϕ , the stem is being rotated through an angle α in the z_0y_0 plane about the center, C. The point P, at the intersection of the sphere with the y_0 axis, traces a spiral path on the sphere as it rotates from $\alpha = 0$ to $\alpha = 90$. Crystallographic hkl planes tangent to this spiral path are brought continuously into the diffracting position. This path, then, is that traced by the hkl poles. Their diffracted intensity which is directly proportional to the pole density is recorded on the strip chart and then manually transferred to a stereographic projection by use of the stereographic net shown in Figure 3.10. An example of the strip chart recording obtained from a spiral scan of the 112 planes of α -extruded uranium is shown in Figure 3.11. The pole figure obtained from this chart is shown in Figure 3.12. If more data is required, a second spiral scan may be obtained by setting the sphere at a starting position at $\alpha = 0$ and $\phi = 180$, and rotating at the same speeds as before. This scan gives the pole densities along an interlocking spiral path displaced by $\phi = 180$.

FIGURE 3.8

Motion of the Sphere in the Pole Figure Goniometer. (a) Shows the starting position, (b) shows how the axis of the sphere EA, R_1 , R_2 move about the original coordinate system X_0 , Y_0 , Z_0 of the pole figure goniometer, (c) shows the final position of the sphere. The spiral path traced by the point P is shown.

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FIGURE 3.9

Imaginary Spiral Path Traced Out on Sphere by the Intersection of the Surface with the Fixed Goniometer Axis Y_0 as the Sphere is Simultaneously Rotated Through α and ϕ

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FIGURE 3.10

Spiral Net Showing Loci of hkl Poles on Stereographic
Projection as Traced by α , ϕ Rotation, Pole Figure
Goniometer Starting at $\alpha = 0$, $\phi = 0$

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FIGURE 3.11

Drawing of Strip Chart Record of Diffracted Intensity from 112 Planes of α -Extruded Uranium as a Function of α and ϕ

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FIGURE 3.12
112 Pole Figure of α -Extruded Uranium

Evaluation of High Strength Zirconium Base Alloys - J. J. Holmes

The mechanical, metallurgical and corrosion properties of the heat treatable alloy Zr-2a/o Nb-2a/o Sn (HSZA) are being investigated. Determination of these properties is required for an evaluation of the usefulness of this alloy in fuel element applications.

Tensile properties of HSZA annealed at 950 C have been determined at room temperature and 300 C. Testing of specimens water quenched from 950 C was extended to 450 C. The tensile strength of the alloy in both conditions of heat treatment is plotted as a function of temperature in Figure 3.13. The water quenched material failed in a brittle manner outside the gage length, making measurement of elongation to fracture impossible. The annealed material fractured in a ductile fashion giving an elongation to fracture of 27 per cent to 30 per cent.

Erichsen cup tests on annealed and water quenched Zr-2a/o Nb-2a/o Sn demonstrate the excellent ductility of the annealed material, and, in contrast to the brittle fracture encountered in tensile tests, show good ductility in the water quenched material.

Since welding plays an important role in the fabrication of fuel elements the weldability of HSZA has been evaluated. Free bend welding test specimens 2 x 0.080 x 1/2-inches were welded by the TIG technique. Free bend tests indicated sound welds were obtained by the TIG technique. The ductility of the weld is less than that of the annealed material but greater than that of the water quenched alloy.

The effect of quenching temperature on the hardness and microstructure of HSZA is being studied in order to develop a heat treatment, compatible with that given metallic uranium fuel elements, which will give high strength and ductility in HSZA cladding. Hardness values are plotted as a function of quenching temperature in Figure 3.14. HSZA is seen to respond well to heat treatment in the 650 C to 800 C temperature range which includes the temperatures used in the beta treating of uranium fuel elements. The metallography of specimens quenched from various temperatures is nearly complete. Material quenched from 950 C consists of needles of α' in a matrix of transformed beta. Alpha prime is a solute rich phase formed by a shear process from the beta phase upon rapid quenching. The alpha prime needles formed upon quenching HSZA from the beta phase display the definite crystallographic relationship with the matrix characteristic of martensitic material (Figure 3.15). Zr-2a/o Nb-2a/o Sn quenched from 900 C consists of alpha zirconium solid solution, alpha prime and transformed beta (Figure 3.16). The quantity of alpha prime and transformed beta present in the microstructure decreases with quenching temperature to 800 C. Below 800 C the microstructure consists of alpha and the equilibrium intermetallic compound, probably Zr_4Sn (Figure 3.17).

The strengthening of this alloy can not be attributed to the formation of alpha prime on quenching. The hardness of the material tested at Hanford is little affected by the presence of alpha prime in the microstructure. The

FIGURE 3.13

FIGURE 3.14

Effect of Quenching Temperature on Hardness of Zr-2 a/o Nb-2 a/o Sn

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FIGURE 3.15

Zr-2 a/o Nb-2 a/o Sn Water Quenched from 950 C After 1/2-Hour
at Temperature, 250 X, Bright Field

FIGURE 3.16

Zr-2 a/o Nb-2 a/o Sn Water Quenched from 900 C After 1/2-Hour
at Temperature, 250 X, Bright Field

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FIGURE 3.17

Zr-2 a/o Nb-2 a/o Sn Oil Quenched from 780 C After 24 Hours
at Temperature, 500 X, Bright Field

greatest hardening occurs in quenching from temperatures too low for the formation of alpha prime. Further metallographic and X-ray studies will be carried out in order to define the strengthening mechanism in Zr-2a/o Nb - 2a/o Sn alloy.

FUNDAMENTAL STUDIES

Effect of Irradiation on Thin Films - T. K. Bierlein and B. Mastel

Electron microscopy with its high resolving power permits direct examination of defects such as dislocation loops and fission fragment damage in thin films and foils. The extent of radiation damage in various materials can be determined by comparing pre- and post-irradiation states. This information in turn permits a better understanding of the basic mechanisms of radiation damage.

Fission fragment tracks are observed in fissionable and nonfissionable films by transmission electron microscopy. As indicated in previous reports, these tracks are regions in the film from which material has been removed. In order to establish if vaporization, chemical reaction, and condensation of material are associated with the registration of damage in fissionable uranium dioxide films, the following experiments were performed. A 200 Å thick carbon substrate, the collector, was positioned at a distance of 1 mm from a 100 Å thick uranium dioxide film on aluminum during an irradiation period which resulted in an exposure of approximately 4×10^{18} nvt (thermal). Examination of the carbon collector taken from an air filled capsule revealed the presence of rosettes with diameters as large as 5 microns, Figure 3.18a. Electron diffraction patterns of these deposited rosettes yielded interplanar spacings of 2.012 and 1.723 Å, which compare quite favorably with U_3O_8 (1.99 and 1.71 Å). The uranium dioxide may have reacted with oxygen to form U_3O_8 before, during, or after sublimation. A similar exposure in an evacuated capsule, however, does not yield deposited rosettes, but many very dense, small particles having a maximum diameter of approximately 400 Å, Figure 3.18b. Electron diffraction analysis of these particles identifies them as uranium dioxide. In a vacuum environment the uranium dioxide apparently sublimates as uranium dioxide and its growth differs from that of U_3O_8 . Since such condensed particles are continuously being perturbed by fission, a study of the type of recondensed particulates as a function of burnup should be of interest. Such irradiations are in progress.

Attempts at preparing thin metallic films of chemically reactive metals for subsequent irradiations has continued. An evaporation chamber containing a movable, water cooled anode and a self-biased electron gun has been utilized. Uranium at the end of a 1/8-inch diameter rod is superheated by the impinging electrons to a sufficiently high temperature to permit rapid evaporation. Films formed by this process are far superior to conventional tungsten basket evaporations of uranium for the following reasons: (1) a small point source of evaporating material is achieved; (2) alloying with tungsten is avoided; (3) evaporation rates are easily controlled over longer periods of time (larger available stock of evaporating material); (4) gettering of residual impurities and purification of evaporating stock can be accomplished more easily; (5) much higher temperature of evaporation can be achieved.

(a) 19,000 X

(b) 103,000 X

FIGURE 3. 18

Electron Micrographs Illustrating the Type of Sublimate Which Forms
During Fission in Thin Films of UO_2

Left (a): Sublimate formed on carbon collector after 4×10^{13} nvt exposure in an air environment
Right (b): Same, but in a vacuum environment.

Sandwich films obtained by evaporating uranium onto carbon substrates and coating the evaporated uranium without breaking vacuum with, for example, a layer of aluminum may yield a uranium film which will not oxidize on exposure to air. Such sandwich films containing evaporated uranium or evaporated alloys of uranium are currently being prepared and studied. Figure 3.19 shows the appearance of sandwich films prepared by the electron beam evaporation technique. The sandwich film consists of carbon, evaporated uranium (after air exposure the film is present as uranium dioxide), and an evaporated coating of WO_3 . Since the manner and extent of fission fragment damage may depend quite strongly on the composition, thickness and types of interfaces present in films, it is necessary to compare the behavior of various films during irradiation. As illustrated in Figure 3.19 the sandwich film containing no free surface of fissionable material reveals very sharp, contrasting fission fragment tracks after irradiation to an exposure to 2×10^{16} nvt. Explanations for the relatively large number of deviated tracks present in this film and the changes in track widths, as compared to multilayered films containing a UO_2 free surface, requires additional experimental work.

In order to study the effect of neutron irradiation on foils of nonfissionable materials such as aluminum, Zircaloy-2, etc, a special irradiation facility will be utilized. The irradiations, covered by Production Test IP-346-D, HAPO-243, will be conducted in the 4B Snout at 105 KW at Hanford. Figure 3.20 is a pictorial representation of the capsule holder and its capsules which have been used for establishing thermal flux and temperature characteristics in this facility. The capsules constructed of 99.99 per cent aluminum, are loaded with foils of the materials of interest. The maximum temperature in the interior of the capsule during electron beam welding has been established to be less than 50 C. Eighteen capsules containing a variety of specimens, prethinned and bulk foils of annealed and cold-worked high purity aluminum, prethinned and bulk annealed electron microscope tensile specimens of high purity aluminum, and bulk foils of cold-worked Zircaloy-2, are being assembled for immediate irradiation. Since the damage observable in prethinned specimens may be considerably different than in bulk specimens in which a relatively smaller surface to volume ratio prevails, both prethinned and bulk specimens are being utilized.

Basic Swelling Studies - T. K. Bierlein, B. S. Kosut, R. D. Leggett, B. Mastel, and J. W. Weber

When uranium is irradiated at elevated temperature, drastic increase in fuel volume are frequently observed. This swelling is thought to be due to the accumulation of inert fission gases, krypton and xenon, in pores in the metal matrix. The irradiation temperature, fuel restraint, burnup, burnup rate, and metal parameters such as purity, residual work, structure and second phase distribution are thought to influence the volume increases, but the exact nature or magnitude of the dependence is not known. This investigation is being made to establish the precise influence of these variables on swelling.

FIGURE 3.19

Transmission Electron Micrographs of C-UO₂-WO₃ Sandwich
Films Prepared by Electron Beam Evaporation, Before and After
Irradiation to 2×10^{16} nvt (Thermal) 66,000 X

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FIGURE 3. 20

Capsule Holder and Capsules for Irradiation of Thin
Foil in a Hanford Snout Facility

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Irradiation Program. Two types of capsules are currently being constructed to irradiate high purity uranium at a known and carefully controlled temperature under conditions of minimal restraint: a metallographic specimen capsule, and a general swelling capsule. Two general swelling capsules containing one centimeter diameter uranium spheres have now been charged.

General swelling capsule No. 4 containing four natural uranium spheres, has been undergoing irradiation at a control temperature of 550 C for the past seven months. All of the capsule components are exhibiting their original high integrity. Some difficulty was experienced with control instrumentation in the early stages of the irradiation of this capsule but this was overcome by replacing the potentiometers with an improved model and using premium, pre-aged vacuum tubes. This equipment is now functioning satisfactorily. General swelling capsule No. 5 containing two natural spheres and one three per cent enriched sphere was irradiated during the quarter. It operated at a control temperature of 525 C. The operation of this capsule revealed that placing specimens of more than one enrichment in a single capsule of this geometry results in very large thermal gradients when the reactor is operating. Figure 3.21 summarizes the thermal history of capsule No. 5 during its irradiation. It can be seen that the thermal gradients were as large as 150 C due to the different specific rates of heat generation along the capsule because of the varying enrichment. In order to minimize effects of this sort, future capsules of this configuration will employ a single enrichment. This will not, of course, eliminate the temperature gradient that exists from the center to the edge of the solid spheres. In order to overcome this, the capsule has been modified to accommodate half-inch diameter, thin-walled, 0.030-inch, split, hollow cylinders in place of the spheres. Table 3.2 indicates the maximum temperature difference that will exist in specimens of this geometry as a function of flux and enrichment assuming an adiabatic inner surface. Since the assumption of an adiabatic inner surface is extremely pessimistic, specimens of this sort will operate at a temperature that is as uniform as present control systems will hold it constant, i. e. ± 5 C. Thus, a "constant" temperature irradiation assumes real significance.

TABLE 3. 2

ΔT IN $^{\circ}\text{C}$ OF A URANIUM HOLLOW CYLINDER IN A NEUTRON FLUX

<u>Per Cent U²³⁵</u>	<u>10¹² nv</u>	<u>10¹³ nv</u>	<u>10¹⁴ nv</u>	<u>10¹⁵ nv</u>
0.72	0.04	0.4	4.4	43.7
1.44	0.08	0.9	8.7	87.4
2.88	0.17	1.7	17.5	174.8

Capsule No. 6 employing the split, hollow cylinder specimen configuration has now been assembled, successfully bench tested and shipped to the reactor for charging. This capsule was X-rayed at operating temperature to determine the NaK level during simulated reactor conditions. The bellows-syphon was observed to be functioning as designed and maintaining NaK around

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FIGURE 3.21

In-Reactor Thermal History of General Swelling Capsule No. 5

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the specimens regardless of temperature or capsule orientation. Interestingly enough, the level of NaK in the expansion chamber was observed to decrease as the capsule was heated the first time in spite of the fact that NaK has a much higher coefficient of expansion than does the stainless. Concurrent with this the shape of the NaK meniscus was observed to change from convex to concave indicating a nonwetting and subsequent wetting action. As there are numerous crevices in this capsule, it is postulated that the lowering of the NaK level is brought about by the wetting of the internal surfaces of the capsule and the attendant flow of NaK into cracks and crevices. The capsule contains three specimens, two of which are as-extruded and a third that was oil quenched in vacuum from 730 C after a holding time of fifteen minutes.

Components are now on hand for eight metallographic specimen capsules but actual construction awaits delivery of a redesigned heater. Experience with the bench testing of these capsules indicated that the heater connection formerly employed could not be relied upon to perform adequately. The old configuration is being replaced with a sheathed heater with an integral lead wire that will extend back to the face of the reactor.

Pore Size and Distribution. Optical and electron microscopy afford a direct means for detecting accumulated fission gases in irradiated uranium. Diameter measurements of pores exposed on the surfaces of specimen subjected to various annealing treatments permits calculation of pore void fraction and pore void density values.

Multiple specimens cut from clad rods of uranium irradiated to burnups of 0.29 a/o and 0.41 a/o have been annealed to temperatures of 600, 650, and 700 C, respectively. Heating and cooling rates were 50 C/hr with the exception of the anneal to 700 C. In this case, the heating and cooling rates in the range 620 to 700 C were reduced to 10 C/hr. Time of holding at maximum temperature was one hour for each specimen. Replicas obtained from the etched specimens have been studied.

Figures 3.22 and 3.23 show representative optical micrographs of replicas of the specimens and illustrate the influence of the annealing treatments on swelling.

It is quite apparent that burnup, 0.29 a/o versus 0.41 a/o, has a marked influence on the amount of swelling that occurs during post-irradiation annealing of unrestrained specimens. Figure 3.23 shows the heterogeneity of porosity and microstructure in the immediate vicinity of impurity inclusions which has developed during the short time alpha phase annealing treatment. This heterogeneity is similar to that observed during longer time anneals (100 hours) at the same temperatures. Figure 3.24, an electron micrograph of the replica for the specimen heated to 600 C, shows the marked influence of impurity inclusions on the size of the gas pores and the increase in the number of pores per unit volume in the vicinity of impurity inclusions. Pore size diameters, obtained from pore diameter measurements on electron micrographs, have yielded estimates of the total pore void fraction and pore void

FIGURE 3.22

Optical Micrographs of Replicas of Uranium Irradiated to a Burnup of 0.29 a/o and then Annealed. 250 X (a) As Irradiated (b) After 1 hr. Anneal at 600 C (c) After 1 hr Anneal at 650 C (d) After 1 hr Anneal at 700 C.

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FIGURE 3. 23

Optical Micrographs of Replicas of Uranium Irradiated to a Burnup of 0. 41 a/o and then Annealed. 250 X (a)As Irradiated (b)After 1 hr Anneal at 600 C (c)After 1 hr Anneal at 650 C (d)After 1 hr Anneal at 700 C.

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FIGURE 3.24

Electron Micrograph of Negative Replica of Uranium Irradiated to a Burnup of 0.41 a/o and then Annealed 1 hr at 600 C. Note the Fine Porosity in the Regions Around Impurity Inclusions. 5, 600 X

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densities in the annealed specimens. Such estimates have been made for regions in the specimens which were irradiated at a high (approximately 500 C) and a low (approximately 300 C) temperature. Table 3.3 lists these values as well as the experimentally determined changes in uranium density associated with the annealing treatment. Tetrabromoethane was used as the immersion medium for determining the density of the uranium.

Inspection of Table 3.3 reveals the good agreement between the experimentally determined $\Delta\rho/\rho$ ratios and the calculated pore void fraction values determined statistically from electron micrographs. If the void fraction and void density values are accurate, there appears to be a distinct difference in the growth and agglomeration of the gas into observable pores in the edge and center regions of the specimen, for which the temperature during irradiation was approximately 300 and 500 C, respectively. That gas agglomeration does occur is indicated by the decrease in total number of pores per unit volume which occurs at temperatures of annealing above 650 C. Pore elimination may occur by either of two processes: (1) solution of gas atoms from small pores and precipitation on larger, growing pores; and (2) growth of large pores which consume the essentially stationary smaller pores. Which of these two processes is applicable should be determinable from comparison of the pore size frequency curves as a function of time and temperature of annealing. Such frequency curves and comparisons are being made for the specimens which have a burnup of 0.29 a/o. Specimens having the higher burnup, 0.41 a/o, will be treated in a similar fashion. As the optical micrographs of Figure 3.23 indicate, the large pores present in optical micrographs constitute a large proportion of the void fraction of pores present in the specimens with a burnup of 0.4 a/o. The void fractions determined from electron micrography, which include pores having diameters in the range 800 Å to 3 μ , must, therefore, be combined with the void fraction of pores revealed by the optical micrographs.

Metallography and density measurements on an unrestrained specimen of uranium irradiated to a nominal burnup of 0.07 a/o at a temperature less than 250 C have been completed. The surface of this specimen has undergone extensive roughening, Figure 3.25. It is extremely difficult to recognize and focus on identical pre- and post-irradiation areas in the sample. All areas of the specimen show considerable plastic deformation and lineage structure.

Geometry measurements on this specimen indicated an approximate volume increase of 3 per cent. Density measurements, however, indicate little change in volume. Since the final density is 18.95 g/cc ($\Delta\rho = -0.3$ per cent). The geometry measurements apparently yield very inaccurate estimates of the final volume. Electron microscope study of this specimen revealed no gas porosity in the as-irradiated state. Both this specimen and the lower burnup specimen, GEH-14-33 and 35, were reground, polished, etched, and replicated. No internal porosity other than the fine 200 Å diameter "decorated" porosity was detected. Microhardness measurements on these two specimens will be attempted.

TABLE 3.3
DATA OF SWELLING IN URANIUM DUE TO POST-IRRADIATION ANNEALING*

Specimen History	Measured Density, g/cc ρ	Fractional Density Change $\Delta \rho / \rho$	Pore Void Fraction ¹⁾			Pore Void Density ²⁾			Avg. Vol. Per Pore, μ^3
			Edge** ~300 C	Center** ~500 C	Average	Edge** ~300 C	Center** ~500 C	Average	
Pre-Irradiation	18.92								
Post-Irradiation 0.29 a/o Burnup	18.62	0.016	0.030	0.026	0.028	2.77	3.34	3.05	0.009
600 C Anneal	17.68	0.066	0.077	0.060	0.068	7.65	7.78	7.72	0.009
650 C Anneal	17.20	0.091	0.106	0.086	0.096	4.47	7.86	6.16	0.016
700 C Anneal	17.18	0.092	0.105	0.091	0.098	2.77	3.15	2.96	0.033

1) Pore void fraction in μ^3 of pores per μ^3 of uranium.

2) Pore void density in number of pores per μ^3 of uranium.

* Specimens were held at annealing temperatures for one hour; heating and cooling rates were 50 C/hr and these were reduced to 10 C/hr in the range 620 to 700 C during the 700 C anneal.

** The estimated irradiation temperatures in the edge and central regions of the specimens 300 C and 500 C, respectively.

FIGURE 3.25

Optical Micrographs of Replicas of Uranium Irradiated to a Burnup of ~ 0.07 a/o. (a) and (b) Portray the Same Area on the Specimen Before and After Irradiation. (c) and (d) Illustrate the Lineage Structure Which Developed During Irradiation.

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Specimens have been fabricated for testing the applicability of bonding uranium to uranium at relatively low temperatures by ultrasonic joining methods. If a sharp interface can be achieved (determined by differences in grain structure) similar bonding of irradiated uranium with nonirradiated uranium should permit valuable fission gas mobility and precipitation studies.

Fission Product Mobility. A knowledge of the mobilities of rare gas fission products through uranium is important in understanding the mechanism of pore formation and growth. Rare gases have been introduced into uranium by electrical glow discharge (sputtering) and by ion bombardment and removed again by heating. To facilitate this study a portable, CEC type 21-620 mass spectrometer has been purchased and incorporated into a glow discharge system. The system is still in a state of evolution but the mass spectrometer has been operated extensively under a variety of conditions and appears to be versatile, sensitive and easy to operate. As the output of the mass spectrometer is in arbitrary units it is necessary to calibrate the instrument to allow a determination of the quantitative amount of gas being pumped through the analyzer. To accomplish this, the response of the instrument was observed as a function of the differential pressure across a standard capillary leak. The calibration was first done with helium and then krypton. Figure 3.26 illustrates the response versus differential pressure for the particular standard leak that was employed. The absolute leak rate for helium at any differential pressure was evaluated by first determining the characteristics of the standard leak using a calibrated helium leak detector. The leak rate versus differential pressure curve obtained with the leak detector was then compared with the response versus differential pressure for the mass spectrometer and a curve of response versus leak rate obtained. This final curve for helium is shown in Figure 3.27. The leak rate versus differential pressure curve for krypton was determined by dividing the leak rate values obtained for helium by 6.44 which is the ratio of helium conductance to krypton conductance for capillary tubes as reported by Dushman.⁽¹⁾ The final relationship between response of the mass spectrometer and krypton flow is included in Figure 3.27. It can be seen that the mass spectrometer is much more sensitive to krypton than to helium. Using a 20 μ amp ionizing current, flow rates of krypton can be analyzed in the range 10^{12} - 10^{15} atoms/sec while flow rates of helium must be in the range 10^{13} - 10^{16} atoms/sec to be analyzed. The range can be adjusted downward by about a factor of five by increasing the ionizing current and can be increased another factor of twenty by changing components in the pre-amplifier. Thus, the range 10^{10} - 10^{13} atoms/sec could be employed if this were found to be desirable. At the moment, the higher range appears more practical from the standpoint of the flow rates that are anticipated.

Restrained Irradiations. Reactor swelling experiments of Zircaloy-2 clad uranium fuel rods with various cladding thicknesses, are being irradiated in NaK filled, temperature monitored capsules to selected exposures at predetermined uranium temperatures. Five swelling capsules GEH-14-98, 14-99, 14-101, 14-104, and 14-105 are presently being irradiated in the MTR with goal exposures in the range 0.25-0.31 a/o BU. Exposures and average

FIGURE 3.26

The Mass Spectrometer Response for Krypton and Helium vs
Differential Pressure Across a Standard Capillary Leak

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FIGURE 3.27

The Mass Spectrometer Response vs Flow Rate for He and Kr

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center uranium temperatures for the fuel rods in these capsules through MTR Cycle 143 are, respectively, 0.25, 0.09, 0.29, 0.29, and 0.36 a/o BU and 500, 290, 450, 600, 335 C. A sixth capsule of similar design, GEH-14-102, was charged in the ETR at the beginning of Cycle 30. Center uranium temperature is 425 C. Two swelling capsules GEH 14-97 (0.42 a/o BU and 650 C) and 14-103 (0.21 a/o BU, and 300 C) are now awaiting examination at the Radiometallurgy Operation.

Radiometallurgy examination on nine 1.6 per cent enriched uranium fuel rods irradiated in NaK swelling capsules in a Hanford reactor is complete except for three density determinations. Density decreases of up to 3.1 per cent were measured on three of the declad fuel rods irradiated to 0.25 a/o BU at center temperatures ranging from 480-525 C. Three other fuel rods irradiated under identical conditions but which suffered cladding failures at some time during the irradiation have uranium density decreases in the range 3.7 to 7.3 per cent. Table 3.4 lists the fuel rods with the irradiation conditions and the density results. The difference in density changes between the failed and unfailed rods demonstrates the effect of external restraint on the uranium. Three fuel rods from this same series of capsule irradiations but exposed to a lower burnup (0.14 a/o) are to be examined soon for cladding condition and volume increases in the uranium.

Effects of Irradiation on Molybdenum and Zirconium - D. L. Gray

The effects of irradiation on molybdenum and zirconium are being investigated with X-ray diffraction techniques. The studies of irradiated molybdenum have been extended to include effects of annealing highly irradiated ($> 2.3 \times 10^{19}$ nvt fast) material. Isochronal anneals for molybdenum irradiated to 5×10^{19} fast nvt have been carried out to 1100 C and changes in X-ray line shape and lattice parameter have been observed. Anneals of 30 per cent cold rolled molybdenum (rolling temperature 400 C) have been run simultaneously, for comparison purposes, and changes in line shape and position have been observed with increasing temperature above 400 C.

A comparison of the isochronal lattice parameter annealing curves for molybdenum exposed to 5×10^{19} nvt with those for 6.5×10^{17} and 6.5×10^{18} nvt exposures reveals significant differences in the mode of lattice parameter recovery. As can be seen in Figure 3.28, only about 6 per cent of the total lattice parameter increase is recoverable in the temperature range between 25 and 200 C for molybdenum irradiated to 5×10^{19} nvt. Approximately 20 to 25 per cent recovery occurs in this same temperature range for the molybdenum irradiated to lower exposures. The recovery in the temperature range between 200 and 400 C accounts for a larger fraction of the total decrease as the irradiation exposure is increased. Recovery of line width more or less parallels the lattice parameter recovery above 400 C in both the irradiated and cold worked molybdenum. Curves showing the decrease in these properties for 30 per cent cold worked molybdenum appear in Figure 3.29.

TABLE 3.4

FUEL RODS WITH IRRADIATION CONDITIONS AND THE DENSITY RESULTS

Sample Number	Fuel Rod OD Inches	Nominal Cladding Thickness Inch	Exposure		Uranium Center Temp. Range °C	Cladding Outside Temp. Range °C	Uranium Volume Avg. Temp. Range °C	Uranium Per Cent $\Delta\rho$ **
			MWD/T	f/cm ³				
4A7	0.610	0.020	1830	1.1×10^{20}	480-520	215-235	380-407	3.7*
5A11	0.610	0.020	1860	1.11	485-525	220-235	382-415	3.1
5A2	0.630	0.030	1815	1.08	480-520	195-210	382-415	2.5
5A15	0.630	0.030	1870	1.12	490-530	200-215	390-425	1.85
3B10	0.610	0.020	2080	1.25	480-520	250-270	390-425	7.3*
5A7	0.630	0.030	2060	1.23	485-525	250-270	400-432	6.0*

* Cladding Split

** Density measurements were made after the Zircaloy-2 clad was removed.

FIGURE 3.28

Lattice Parameter Recovery of Irradiated Molybdenum, 1 hr Anneals

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FIGURE 3.29
Recovery of Molybdenum Cold-Rolled 30 Per Cent at 400 C

The annealing stages can be interpreted in terms of the model proposed earlier, ⁽²⁾ namely that trapped interstitials and interstitial clusters are present in molybdenum after irradiation. Vacancies are present also, probably as singles, di-vacancies and higher order groups. The number of interstitial clusters and higher order vacancy groups are postulated to increase with increasing irradiation exposure. It was suggested that interstitial migration occurs in the low temperature annealing stage. If the idea of interstitial migration is given credence as a primary mechanism in the low temperature stage (25 to 200 C) and if it is accepted that lattice parameter expansion is directly proportional to the amount of defect recovery; then, for the highest exposure specimen (where only approximately 6 per cent recovery occurs) the implication is that most of the interstitials are present as clusters, or are in fairly deep traps as previously inferred from the neutron exposure data. The 200 to 400 C recovery could possibly be due to the release of interstitials from deep traps or to the migration of di-vacancies. The number of di-vacancies would doubtless increase as irradiation progresses and the increased percentage recovery at 200 to 400 C would then reflect this increase in concentration. The recovery from 400 to 800 C which is seen in both the cold worked and irradiated material must be due to a number of reactions, which depend on vacancy and dislocation movements as a primary mechanism. The annealing kinetics above 400 C will be investigated during the coming quarter.

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2. Gray, D. L. and W. V. Cummings. "An X-ray Diffraction Study of Irradiated Molybdenum," Acta Met., 8: 446-452. 1960.

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METALLIC FUEL DEVELOPMENT

NEW FUELS DEVELOPMENT

The Effects of Cladding Thickness Variations in Zircaloy-2 Clad Coextruded Rods - J. W. Weber

Zircaloy-2 cladding failures of coextruded uranium rods have occurred in NaK capsule and high temperature recirculating water loop irradiations. From the appearance of the fuel rods the same failure mechanism is apparently operating in both cases. In the previous quarterly of this series a description and comparison of the loop test and one of the capsule tests, 4A7, is given.

Of the five cladding failures recently found in the NaK capsule irradiations (Figure 4.1) four occurred in rods with 0.020-inch nominal cladding thickness and one in a rod with 0.030-inch thick cladding. As reported in the previous quarterly the minimum cladding thickness of the 0.020 and 0.030-inch clad rods was estimated to be 0.010-0.014 inch and 0.020-0.024 inch respectively. There were three rods with 0.030-inch thick cladding and one rod with 0.020-inch thick cladding in this test of nine capsules which showed no external evidence of cladding failure. To see if there may have been large differences in cladding thickness between the failed and unfailed rods a metallographic examination was performed on a cross-section from a failed and an unfailed rod in both the 0.020-inch and 0.030-inch clad rods.

Rod 4A7 - Failed, 0.020-Inch Cladding. This rod served for the examination of the failed 0.020-inch thick cladding; minimum thickness was probably about 0.014 inch.

Rod 5A11 - Unfailed, 0.020-Inch Cladding. This fuel rod had no outwardly visible evidence of failure, however, on sectioning, a necking of the cladding was found (Figure 4.2). Minimum thickness at the necked region is 0.011 inch. It is estimated that the original cladding thickness, before necking occurred, was 0.014-0.015 inch. Figure 4.3 is a higher magnification view of the necked area showing the localized grain deformation accompanying the necking.

Rod 5A7 - Failed, 0.030-Inch Cladding. The cladding on this rod has failed with little or no reduction in thickness indicating a brittle type failure (Figure 4.4 a). The clad split has adjacent to it a section of cladding thinner than where the split occurred. However, it must be remembered that cross-section shown is not necessarily at the initial point of failure. Note in Figure 4.4 b how the uranium and cladding have sheared past one another at the interface. The crack in the uranium terminates in a ductile manner.

Rod 5A2 - Unfailed, 0.030-Inch Cladding. This rod shows no evidence of necking or clad failures. At two locations on the cross-section minimum cladding thickness of 0.023 inch were measured.

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4A7
Cladding
0.020
Inch

2B16
Cladding
0.020
Inch

2B25
Cladding
0.020
Inch

3B10
Cladding
0.020
Inch

5A7
Cladding
0.030
Inch

FIGURE 4.1

Cladding Splits in Coextruded Fuel Rods Irradiated in NaK Capsules 1 X

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REC-62 RICHARD, JAMES

FIGURE 4.2

Transverse Cross-Section of Fuel Rod 5A11
Showing the Striation in the Cladding 5X

FIGURE 4.3

Photomicrograph of Cladding from Cross-Section of Figure 4.2 at the Striation (Note the grain deformation in the striated region in the center of the picture.) 250 X

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(b) Photomicrograph of cladding at the split on the right of Figure (a)

Zircaloy-2 Cladding Splits in Fuel Rod 5A7

FIGURE 4.4

(a) Transverse cross-section showing the splits in the cladding with little or no associated reduction in thickness. 5X

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There does not appear to be a correlation of clad thickness with failure for either the 0.020 or 0.030-inch clad rods. However, as pointed out before, the initial point of failure is not necessarily the section examined. An experiment has been designed in an effort to measure the effect of cladding thickness variation on the susceptibility to failure. This experiment will use fuel rods with cladding thicknesses of 0.030 and 0.040-inch with striations machined in the outer surface of the cladding. The fuel rods will be encapsulated in NaK capsules to provide cladding surface temperatures of 240, 290, and 320 C. The cladding surfaces and thicknesses will be carefully precharacterized before irradiation.

Thermocoupled Fuel Element Tests - R. L. Call

A series of thermocoupled fuel element tests is being performed in a high temperature, high pressure, recirculating loop in one of the Hanford reactors. These tests are being performed to measure the temperature effects of "crud-film" deposition on fuel elements. A description of the fuel elements and the results of the first test of the series are given in a previous quarterly report.

The thermocoupled fuel element from the first test of the series has been examined in a radiometallurgy cell. Figure 4.5 shows the fuel element "as received" in the cell. The stainless steel to Zircaloy-2 braze joints which provide the transition from the Zircaloy-2 end cap to the stainless steel thermocouple sheath can be seen at the left. For the first portion of the irradiation, the local bulk water temperature was 260 C. For the remainder it was maintained at 60 C because of a leak in the loop piping. The thermocoupled fuel element and its downstream heater elements were discharged because of rupture indications. No evidence of failure was found during the radiometallurgical examination of the thermocoupled fuel element. The three stainless steel to Zircaloy-2 braze joints were examined carefully and were found to be sound. The downstream heater elements are now believed to be responsible for the rupture signal. They have not been examined in a radiometallurgy cell, however.

The second thermocoupled fuel element was charged into the same recirculating loop used for the first test of the series. This fuel element was irradiated in 260 C water and then the reactor was shut down. About 24 hours after the reactor was started up rupture signals were again received by the loop rupture monitors. The thermocoupled fuel element and its downstream heater elements were discharged from the loop. Figure 4.6 shows the thermocoupled fuel element "as received" in the radiometallurgy cell. As can be seen in Figure 4.6, the cladding was split on each side of the rod adjacent to the peripheral thermocouples. The uranium core was split in the plane of the thermocouple holes. The stainless steel to Zircaloy-2 braze joints were sectioned and found to be sound. It appears likely that the initial water entry was through the cap-to-cladding weld. The formation of UO_2 in the thermocouple holes then caused the uranium core and the cladding to split.

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FIGURE 4.6

Views of Opposite Sides of the Second Thermocoupled Fuel
Element "As Received" in the Radiometallurgy Cell After
Irradiation to 110 MWD/T in 260 C Water ~0.5 X

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Additional thermocoupled fuel elements and heater elements are being prepared. These will be charged and the program to investigate the effects of "crud film" deposition on fuel elements will be continued.

Fuel Element Supports - Bearing Surfaces - J. W. Weber

Tests made ex-reactor show that severe scratching of horizontal Zircaloy-2 process tubes will occur when full-sized NPR fuel elements with Zircaloy-2 supports are charged in them. In addition to the loss of metal and eventual thinning of the process tube after a number of charge-discharge operations, these scratches may lead to localized corrosion or hydriding or act as regions of stress concentration. A criteria for the successful performance of a support bearing material is that it (1) not break through the autoclave film of the process tube, (2) that it be compatible with Zircaloy-2, and (3) be corrosion resistant in high temperature water.

In the previous quarterly of this series⁽¹⁾ and in HW-65962 it was reported that plain low carbon steels meet these criteria. Because the fuel element supports will be in sliding contact with NPR fuel charging magazines, a test was made using 1008 steel supports against a 321 stainless steel surface. Results of these tests show severe deep scratching of both the stainless surface and the bearing surface of the support. When the scratched support was subsequently tested against an autoclaved Zircaloy-2 surface, severe scratching and galling of the Zircaloy-2 occurred. The addition of one per cent by volume of a water soluble charging oil to the water lubricant eliminates galling of the stainless steel by ingot iron supports. However, with low carbon steel up to 25 per cent additions of charging oil will not prevent galling.

To improve the flow and pressure drops attributed to the supports, it may be desirable to change the cross-sectional contours. Ingot iron supports with a cross-sectional radius reduced to 3/8 inch did not scratch the autoclaved Zircaloy-2 surface within a wear distance of 180 feet. A final report is being written on this series of wear tests against autoclaved Zircaloy-2.

Self Supports for NPR Fuel Elements - R. G. Wheeler

An all steel self support and method of attachment was developed to satisfy the nongalling, corrosion and mechanical requirements for NPR outer tube fuel elements. Report, HW-65807, Steel Supports for NPR Fuel Elements, describes the supports and attachment method. A charge of NPR inner tube elements fitted with these supports will be tested in a KER high pressure loop.

A clip designed to attach the inner tube of an NPR element to the outer tube will be tested in the same loop charge mentioned above. The function of the clip is to prevent telescoping of the outer and inner fuel element tubes during charge or discharge operation. The clip is made of Zircaloy-2

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and projection welded to the inside surface of the outer tube element, as illustrated in Figure 4.7. Rotation of the outer fuel element tube, with respect to the inner, after they are assembled locks the clip into a self support on the inner tube as illustrated in Figure 4.7.

Plastic Deformation and Failure of Coextruded Jackets - R. G. Wheeler

Early in the year, Zircaloy-2 tubes were burst in reactor with no loss in average tangential strain at fracture after about 10^{19} nvt fast neutron exposure. In contrast, some failures in Zircaloy-2 clad experimental fuel elements show highly localized necking at the fracture with little or no plastic strain in the remainder of the jacket. The nearly complete loss of uniform elongation in Zircaloy-2 irradiated to 10^{21} nvt (2) or more, is considered to be the major cause of jacket splitting without significant diameter change. Recent tests on unirradiated coextruded Zircaloy-2 jackets from which the uranium core was removed, however, showed the state of stress significantly affected the fracture mode. The tests were made in the temperature range of a fuel element jacket operating in pressurized water (275-300 C). Jackets burst with internal pressure alone had an average circumferential strain of 16 per cent at fracture. Jackets burst by compressing silicone rubber inside the tubes had an average circumferential strain of 5 per cent. In the latter case, most of the strain was in localized necked areas; whereas, internal pressure produced more uniform elongation. The ratio of tangential to longitudinal stress is 2:1 for internal pressure. With the rubber filled tubes, the tangential to longitudinal stress ratio was an undetermined large negative number. A study of the strain and fracture of coextruded Zircaloy-2 jackets over a wide range of controlled biaxial stress ratios will be used to help differentiate effects of stress state from radiation damage as a cause of jacket failure at low average strain.

Fuel Element Model to Predict Cladding Deformation - K. R. Merckx

Localized straining in Zircaloy cladding has caused failures of irradiated uranium fuel elements. The localized straining occurred with very little uniform elongation in the majority of the cladding material. If the deformations due to the thermal expansion and swelling volume changes could be shifted in tubular fuel elements from the outer to inner cladding, then the possibility of localized straining failures should be minimized. In order to determine what geometries are necessary, a mathematical model of a plastically deformed fuel material, with strain hardening material for the cladding, has been developed. The entire fuel material region is assumed to be plastic with low yield strengths; 500-1000 psi being assumed for the uranium fuel. Parameters which can be evaluated with this model are yield strengths for the uranium, volume expansions or swelling, tube geometries, inner clad thicknesses, and exterior cladding strains. The analysis yields outer cladding thickness, cladding strains and stresses, and the stress and strains within the fuel material. The necessary mathematical relations, method of analysis, and computer program have been obtained.

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FIGURE 4.7

Locking Clip Shown in Engaged Position

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Increased Strain Rates Due to Large Temperature Cycles - K. R. Merckx

The microstress distributions in polycrystalline uranium due to anisotropic thermal expansions of the uranium crystals, change macroscopic strain rates. For constant load tests, comparable strain rates have been obtained for uranium specimens undergoing cyclic temperature variations, (150 C-450 C) at stresses an order of magnitude lower than uranium specimens at constant temperature, (450 C). Previous test results have been reported in this series. An additional test at 750 psi cycled once a day between 155 C and 435 C has been completed while a similar test at 1000 psi is being run. Figure 4. 8 is a graph of the strain data for the 750 psi test.

High Energy Rate Forming - R. G. Wheeler

The Dynapak, high energy rate forming machine, was used mostly for compaction of UO₂ as reported by Brite.⁽³⁾ Another application of the Dynapak during the quarter was forming of steel jackets for plate type fuel elements. The material formed was 0. 010 and 0. 020-inch thick sheets of stiff steel alloys such as cold-worked AISI 406 and an alloy of 7. 6 per cent aluminum, 5. 1 per cent chromium, balance iron. These materials were formed into long shallow (0. 15-inch deep) cups that will be seam-welded to form unbonded jackets for plate elements. A steel male die formed the cups against a rubber backup pad. A comparison to determine if high energy rate forming of sheet material has any advantages over forming with a relatively slow press is underway. There are indications of less springback and wrinkling in high energy rate formed pieces.

FABRICATION DEVELOPMENT

"Extrusion Closure" Process - E. A. Smith

This method of making closures on coextruded Zircaloy-clad uranium tubular fuel elements has been developed to a stage where it shows considerable promise. The procedure as first tried involved:

- a) Mechanically and chemically removing the uranium from between the Zircaloy inner and outer jacket walls to the desired depth at each end of the clad tube.
- b) Inserting an annular cap of Zircaloy in the groove so formed.
- c) Vacuum-electron welding the inner and outer jackets to the annular cap.
- d) Heating the assembly into the uranium β -phase (715-730 C).
- e) Extruding the welded closure, cap, and jacket walls in a closed die to form an extended hollow cylinder of composite material with a thin cross-section.
- f) Facing off the extruded portion and if desired, rewelding.

Metallographic study shows that the composite metal in the extrusion flash and for an appreciable distance below it has become blended and homogenized, thus insuring a leakproof closure.

This simple process, however, does not provide for bonding the uranium core material to the cap, as is currently believed necessary for adequate quality. Hence, efforts have been turned to placing a very thin layer of some suitable material at the interface to bring about bonding under the conditions of the extrusion forming.

After trying platings of nickel, iron, tin and copper, and solid shims of the same metals, it became apparent that under the extrusion conditions wrought nickel bonds well to zirconium, but not to uranium; wrought iron bonds well to uranium, but not to zirconium; wrought copper bonds well to both; while plated metals, except electroplated copper, appear to form generally poor bonds, possible due to nonoptimum plating conditions. Thus, it

TEST CONDITIONS: Stress-750 psi Gage Length-1.5 inch Area-0.077 inch²
 Temperature Cycle-155-435 C (4 hr. Temp. Cycle, 20 hr. at 435 C)

FIGURE 4.8
 Constant Load Cyclic Temperature Test

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appears feasible to use either a thin shim of copper (~0.003 inch) or of nickel-clad iron at the interface between the uranium and the Zircaloy cap. It should extend upward for a short distance along the interface with the Zircaloy jacket, to form a Zircaloy-to-Zircaloy seal at the base of the cap and insure that a leak in the primary closure will not connect with the uranium core.

Advantages of this method over the brazing method include the following:

1. The relatively low temperature of the operation (~725 C) leads to better uranium and Zircaloy structures.
2. The 725 C temperature does not damage the already-established uranium-Zircaloy wall bond.
3. Solid state diffusion, under pressure, insures freedom from bubbles and voids at interfaces.
4. Pre-welding, and coextrusion insures sound bond.
5. Elimination of beryllium-bearing braze material reduces the health hazard, and the precautions required.
6. Pressing in a die avoids offsetting the cap with respect to the tube.

A test lot of fuel elements is being prepared by this process for evaluation in the leak-vulnerability test.

Projection Welded Brazed Closure - P. A. Ard and W. I. Steinkamp

The projection welded brazed closure is being evaluated for use in closing coextruded Zircaloy-2 clad uranium fuel elements and consists of projection welding a Zircaloy-2 cap directly to the cladding and brazing the cap to the uranium surface in one operation. The welding set-up is shown in Figure 4. 9. Approximately 0.030 inches of uranium is acid milled from the end of the element to produce a uranium free cladding ring which is matched with a projection machined in the cap. A layer 0.010 to 0.015-inch thick of braze material is placed over the recessed uranium. The element is first closed with the projection weld with the pressure being maintained on the cap and the power continued for several cycles in an attempt to resistance braze the cap to the uranium. The cap contacts the braze material after settling from the initial projection weld.

This closure concept is initially being tried on KER sized rods (0.593-inch OD by 0.020-inch clad) as the capacity of the available spot welding machine is not sufficient to test this concept on tubular elements. The power required is expected to be approximately the same as required for the hot headed projection welded closure.

The projection weld between the Zircaloy-2 cap and the Zircaloy-2 clad has to date produced a very promising and reproducible projection weld. Approximately 35 per cent of the uranium surface was bonded to the Zircaloy-2 cap in one attempt in which no braze material was added. A Zircaloy-2, Be, Cu braze material will be tested in the very near future along with a 0.014-inch thick bimetallic strip of nickel-iron.

An offsite contract is being considered with a firm which has a welding machine of adequate capacity to test the projection welded brazed closure concept on tubular elements. Initial tests would be on KER inner tubes (1.050 inch OD by 0.500 inch ID).

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FIGURE 4.9

Welding Set up for Projection Welded Braze Closure
on Coextruded Zircaloy-2 Clad Rods

This closure concept is initially being tried on KER sized rods (0.593-inch OD by 0.020-inch clad) as the capacity of the available spot welding machine is not sufficient to test this concept on tubular elements. The power required is expected to be approximately the same as required for the hot headed projection welded closure.

The projection weld between the Zircaloy-2 cap and the Zircaloy-2 clad has to date produced a very promising and reproducible projection weld. Approximately 35 per cent of the uranium surface was bonded to the Zircaloy-2 cap in one attempt in which no braze material was added. A Zircaloy-2, Be, Cu braze material will be tested in the very near future along with a 0.014-inch thick bimetallic strip of nickel-iron.

An offsite contract is being considered with a firm which has a welding machine of adequate capacity to test the projection welded braze closure concept on tubular elements. Initial tests would be on KER inner tubes (1.050-inch OD by 0.500-inch ID).

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Closure and Joining - W. I. Steinkamp

The attachment of steel supports to Zircaloy clad fuel elements has been achieved by means of the riveted Zircaloy stud. This stud is resistance welded to the fuel element through the support and the head of the stud riveted to secure the support. This can be accomplished in one operation on the resistance welding machine or the support can be attached later. Average shear strengths of 1100 pounds have been obtained for 1/8-inch diameter studs.

Another method of obtaining steel wearing surfaces is the direct attachment of supports formed from steel clad Zircaloy. A small amount of this material was available and has been successfully welded to Zircaloy clad fuel elements.

Component Fabrication - R. S. Kemper and R. N. Johnson

Sections of KER single tube (1.752 OD x 1.052 ID) with 0.027-inch inner and outer cladding were heat treated with several quenching cycles and examined to determine the best treatment for a production test lot of this material. Heating and cooling curves were recorded and midwall quench rates determined. Sections of this geometry are heated to the beta phase in two minutes and to 730 C in 3.5-4 minutes in a NUSAL bath maintained at 730 C. This geometry is cooled from 730 C to the transformation in 36 seconds in air. The quenching rates were determined as; 7700 for 20 C water quench, 4650 C/min for 10 second air delay and oil quench; and 3700 C/min for 20 second air delay and oil quench. The best macro and microstructure was observed for the 3700 C/min quenching rate. Some columnar structure was still present; however, and the twenty-two production test elements were treated by holding 5 minutes at 730 C in NUSAL and air cooling for 30 seconds before oil quenching. Two 6-inch elements that were brazed and welded at both ends were heat treated and examined for effect on the bond and refinement of the uranium in the heat affected zone. Oil quenching did not adversely affect the clad-uranium or cap-uranium bonds. Some porosity was observed in the uranium near the cap as shown in Figure 4.10. Typical cap-uranium braze zone and clad-uranium bond zone near the cap are shown in Figure 4.11. This treatment sufficiently refined the core structure in the braze heat affected zone for reactor operation.

Component Fabrication - R. S. Kemper

Eight additional 18-inch lengths of KER outer tube of enriched (1.6 per cent) uranium-2 w/o Zr alloy were inspected and heat treated prior to braze closure for irradiation test elements. An additional extrusion (NMI #21074) was used for this stock. Two of the elements had surface defects that will reject the elements. The pieces were heat treated by salt bath heating in NUSAL 10 minutes at 730 C, 10 second air delay and oil quenching. Warp and dimensional changes were recorded. Double throw warp varied

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FIGURE 4.10

Longitudinal Section of Braze Closed Fuel Element
Showing Porosity Generated in Uranium During Brazing

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FIGURE 4.11

Typical Bond Zones Between Cap and Uranium,
and Clad-Uranium in Braze Heat-Affected Zone

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from 0.001 to 0.038 inches before heat treatment and varied from 0.009 to 0.040 inches after treatment with no consistent changes. Sections were taken from each element after treatment for evaluation of core structure and core to clad bond zone. Considerable variation in structure was observed that appears to be due to variation in alloy content. Typical areas are shown in Figure 4.12. This was observed only on extrusion Number 21074 and not on two elements of a different extrusion. Chemical analysis of these areas will be made.

Arc Melting and Zirconium Base Alloys - R. G. Nelson

An arc melting facility has been purchased and installed in the 306 Building. This furnace is capable of making consumable or nonconsumable ingots up to 6 inches in diameter by 18 inches long. In addition it can make small castings of special shapes up to about 20 and 30 pounds through the use of a skull melting attachment.

A series of zirconium base alloys are currently being prepared and melted in the arc furnace. The alloys are development alloys for the Corrosion and Coatings group and for the Physical Metallurgy group.

Listed in Table 4.1 are the intended compositions of these alloys.

TABLE 4.1

ALLOY COMPOSITIONS

(Weight per cent)

<u>No.</u>	<u>Cr</u>	<u>Fe</u>	<u>Cu</u>	<u>Nb</u>	<u>Mo</u>	<u>Sn</u>	<u>Ni</u>	<u>V</u>
1	0.1	0.15	--	--	--	1.5	0.1	--
2	0.1	0.15	--	--	--	1.5	--	--
3	--	--	--	--	--	2.0	--	2.0
4	0.5	--	--	1.0	--	2.0	--	1.0
5	--	--	0.5	--	0.5	--	--	--
6	--	--	--	1.0	--	--	--	--
7	--	--	--	--	--	--	--	1.0
8	--	--	--	0.5	--	--	--	0.5
9	0.1	0.3	--	--	--	1.5	--	--
10	1.0	--	--	0.5	--	--	--	1.0
11	--	0.1	--	3.0	--	2.0	--	0.05
12	--	--	--	--	2.0	4.0	--	--
13	--	--	--	1.0	--	2.0	--	1.0

Raw material is Japanese sponge, reactor grade, with a 120 BHN. Alloying elements are purchased in a high purity powder form.

Electrode Preparation. Electrodes for each primary melt consist of four 1-1/2 inch octagonal by 12 inch long compacted bars. These are welded together end to end by electron beam and a threaded Zircaloy-2 holder piece

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FIGURE 4.12

KER Outer Tube Sections, 1.6 Per Cent Enriched, 2 w/o Zr-Uranium Alloy Core. (As polished and oxidized after heat treatment showing nonuniform structure areas. Top of picture is top of extrusion die. 2X Top N-18-40, Bottom N-18-96)

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welded to the top end of the assembly. Alloying elements for each bar are weighed and then blended in a glass vial. The alloy blend is then pressed into pills about 1/8-inch thick and 1/2 inch in diameter.

Compacting of the bar is performed on a 700-ton press with the load applied perpendicular to the longitudinal axis of the bar. Approximately 30-tons per square inch of pressure is required to produce a bar of sufficient strength to transport, weld, and melt. One-half of the zirconium charge is placed in the die, followed by the alloy pills. These are distributed evenly along the bar length, followed by the balance of the zirconium charge.

Melting. Melting takes place in a 2.9-inch diameter water cooled copper crucible. A small amount (1/4 lb) of pure zirconium sponge is placed in the bottom of the crucible to start the arc and protect the crucible bottom from the direct impingement of the arc.

The furnace is evacuated to less than 3 microns and pumping continued until the rate of rise of the static system is less than one micron per minute. The electrode is slowly lowered until an arc has been established. The arc current is adjusted to 1400 amperes and the arc gap between electrode and ingot top is maintained at a constant distance as indicated by the voltage between the electrode and crucible. As the electrode is melted off it is lowered to maintain the constant voltage. Experience to date has shown that a starting voltage of about 35 and finishing at 30 gives best results. The five volt difference can be attributed to the voltage drop in the electrode itself. Using 1400 amperes and 30-35 volts the 14-pound electrode is consumed into an ingot in 4 to 6 minutes, thus giving a melting rate of 2.8 pounds per minute and a power consumption of about 0.25 kilowatt hours per pound.

Eight of the above alloys have been melted. Current plans are to forge these into electrodes and remelt into the same size crucible. Double melting of each alloy is used to insure alloy homogeneity and reduce the side wall porosity.

Spider End Plates - F. B. Quinlan

A means of maintaining concentricity and axial alignment of "tube in tube" type fuel elements is being developed. Such a device is required to maintain concentricity of the tubes with a minimum of resistance to the water flow, to be able to withstand charging and discharging impacts, to be free of interference with nondestructive tests and assembly procedures and to be free of any mechanical or metallurgical techniques which may tend to promote corrosion.

Some of the spiders produced to meet the above mentioned conditions are shown in the following figures, 4.13 and 4.14.

In Figure 4.13 the spiders may be made of any desirable material, most likely Zircaloy-2 or mild steel, since the fastening of the spider is purely mechanical. The 4-40 machine screws shown may be replaced by studs that rivet to the ring. It should be noted, in these two cases, that all of the components

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FIGURE 4.13
Zircaloy-2 or 1020 Steel Spiders

FIGURE 4.14
Zircaloy-2 Only

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may be given an autoclave film separately and then assembled without disturbing the film. However, the welded spider shown in Figure 4.14 may require removal of the oxide film prior to spot or projection welding. As the Zircaloy-2 end ring is a comparatively massive piece of metal and as the spider legs would not be too greatly weakened by white oxide corrosion, it may be possible to make such welds without removal of the autoclave film.

Figures 4.15a and 4.15b shows a mild steel stamping which can be readily bent into a combination spider and support. Admittedly this carries the simplicity of the "spider end plate" to the extreme and it could only be considered in terms of 12-inch fuel elements but it does present a concept which may stimulate some constructive thought.

Metal Fabrication - F. B. Quinlan

In support of the work being done to explore the feasibility of the 406 type alloys as a high temperature cladding material, some high temperature rolling experiments have been made.

When two sheets of the 406 alloy were TIG welded together, the weld produced was very brittle. Hot rolling of this welded sheet was considered as a means of changing the cast structure of the weld metal to a wrought structure. As the sheet was thin, approximately 30 mils, it could not be expected to maintain the hot working temperature of 1050-1100 C during the rolling operation.

To provide an ample heat reservoir for this operation, the 406 sheet was placed between two 3/16-inch thick bars of 304 SS. The bars were tack welded together to prevent loss of the sheet. This SS compact containing the welded 406 alloy sheet was then heated in air to 1100 C and hot rolled, in three steps, from a thickness of 0.405 inch to 0.135 inch.

Upon opening the compact it was observed that the 406 sheet parted readily from one SS surface but adhered with a tenacious mechanical bond to the other SS surface. Severe flexing of the adhering piece broke the bond and exhibited a clean metal interface. The opposite face, the one that separated readily had a slight bluish oxide finish. Three subsequent experiments produced the same "one side" tendency to bond which is difficult to explain especially as no fixed orientation of the compact was maintained during rolling.

When the welded 406 alloy was removed from the compact, it was found to be very ductile and numerous severe bends did not cause cracking of the weld metal.

Rod Forming and Surface Expanding Methods - F. B. Quinlan

Fuel rods, either Zircaloy-2 clad coextruded uranium or Zircaloy-2 clad UO_2 may be reformed to have something other than a circular cross section. Specially formed cross sections may be desired on certain fuel elements to increase the surface area for better heat dissipation, for self supporting features or for changing the nature of the thermal stresses produced within the element.

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FIGURE 4.15a

FIGURE 4.15b

Spider Supports

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Such changes in cross section may be made by means of a specially designed "turk's head" as shown in Figure 4.16 and Figure 4.17. By changing the peripheral contour of the three rolls in Figure 4.16, cross sections such as Figure 4.18 (1), (2), (3), and (4) may be made from a bar or powder filled tube of circular cross section. The "turk's head" shown in Figure 4.17 is a commercial model that may be used to produce a variety of rectangular cross sections, Figure 4.18, (5) and (6).

FUEL EVALUATION

Tubular Fuel Elements - G. T. Geering

Four KER tubular fuel elements were irradiated to 3200 MWD/T before failure. The test was designed to compare uranium fuel performance with uranium-2 per cent zirconium fuel performance. A description of the test elements is given in Table 4.2.

TABLE 4.2

DESCRIPTION OF KER TUBULAR FUEL ELEMENTS FOR LOOP 2 TEST

Core Material	Uranium (2 Elements), U-2 per cent Zr (2 Elements)
Bond Type	Coextrusion
Clad	Zircaloy-2 0.020-Inch Thick
Outer Tubes	1.820 x 1.450-Inch Diam x 18-Inch Long
Inner Tubes	1.050 x 0.500-Inch Diam x 18-Inch Long

The four elements operated with high temperature pressurized water coolant in KER Loop 2. The operating conditions for the failed element are given in Table 4.3.

TABLE 4.3

OPERATING CONDITIONS FOR KER TUBULAR FUEL ELEMENTS IN KER LOOP 2

Temperatures at Point of Failure

Bulk Coolant	250 C
Maximum Fuel	410 C

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FIGURE 4.16
Three-Roll Turks Head

FIGURE 4. 17
Four-Roll Turks Head

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FIGURE 4.18

Fuel Element Cross Sections

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The failure was due to a longitudinal clad split. Grooves extended away from the ends of the split in both directions (Figure 4.19a). Several deep grooves running diagonally across the tube appeared on the opposite side from the split (Figure 4.19b). A deep longitudinal groove also appeared near the midlength of the element. A composite photomicrograph transverse to this groove is shown in Figure 4.20. Marked elongation of the grains is seen at the bottom of the groove. Evidently the clad failed due to local plastic deformation imposed by the expanding uranium fuel core. The irradiation damage to the clad may have contributed to the localized nature of the deformation. The diagonal grooves, however, have a different origin. They are associated with internal cracking of the uranium, whereas, the longitudinal grooves were independent of cracks in the uranium. A transverse section through one of the diagonal grooves is shown in Figure 4.21a. A large crack lies directly under the groove, which itself cracked open during handling in the hot cell. The crack and a network of cracks extending away from it are filled with uranium oxide. Water entering through the clad split must have penetrated a crack network in the uranium. The prying action of the oxide formed in the crack network then caused the diagonal groove in the fuel clad.

The oxide filled internal crack network is shown in Figure 4.21b. Measurements of the failed tube are not complete. However, the inside diameters at midlength show that the central coolant channel has developed an out-of-round shape. This is shown in Figure 4.21b. The outside of the tube at midlength remained round, with the average diameter increasing from 1.046 to 1.065 inch.

Brazed Closures - J. C. Tverberg

The first brazed fuel elements were charged in-reactor together with several other types of experimental material. Less than four days later the loop was discharged due to a rupture. The brazed elements were given a detailed examination in Radiometallurgy and no evidence of failure was observed. The activity probably originated in other elements in the loop. The fuel element closure was made by brazing an end cap into each end of the NPR-inner size coextruded fuel tube with the 5 w/o beryllium + 95 w/o Zircaloy-2 brazing alloy. In brazing the elements were heated to 1035 C in 30 seconds, then held for one minute. External pressure of 25 psi was applied to the cap for one minute, the power was shut-off, and the elements cooled in vacuum. Swelling occurred in the end cap region because of the differential thermal expansion of gamma uranium and Zircaloy-2 at the high brazing temperature. Gas bursts during brazing gave considerable trouble, causing voids in the braze zone. This was controlled by outgassing both the brazing alloy and uranium at 880 C for three minutes prior to use. A slower heating rate was used to allow the gas to escape before the braze became molten. Uranium which was mechanically counterbored had smaller gas bursts than that which was acid milled.

Partial results were reported in the last quarterly report on flow tests with brazed material in the Elmo-7 test loop. This test involved six brazed

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Clad Split in Tubular Fuel Element Exposed to 3200 MWD/T

FIGURE 4.19b

Diagonal Grooves on Side Opposite Split 2X

Clad Split and Longitudinal Grooves in Inner Tube 2X

FIGURE 4.19a

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Composite Photo Micrograph

FIGURE 4. 20

Transverse Section Through Groove in Clad of Tubular Element
Exposed to 3200 MWD/T (~150 X, Polarized Light)

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Ion Etch 2X
(A) Transverse Section of Internal Crack and Diagonal Groove

Ion Etch ~15X
(B) Propagation of Oxide Filled Crack into Fuel Clad

FIGURE 4.21

Internal Cracking in Tubular Element Exposed to 3200 MWD/T

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fuel specimens run in 300 C (572 F) water at pH 10.0 adjusted with LiOH and 1600 psi pressure. Samples included two unwelded unheat-treated specimens, two unwelded heat treated specimens and two welded and heat-treated specimens. Flow data are summarized in Table 4.4.

TABLE 4.4

FLOW TEST DATA

<u>Exposure Hours</u>	<u>Cumulative Exposure Hours</u>	<u>Remarks</u>
152	152	One unwelded heat treated element ruptured. Remaining elements looked good.
333	485	Discharged three unwelded elements due to pinhole formation in braze. Welded elements looked good.
801	1286	Two welded specimens discharged due to breakaway corrosion.

Figure 4.22 shows the first ruptured element. Rupture probably occurred through a pinhole in the braze zone. The rupture occurred long before it was discovered as shown by the complete disintegration of the uranium. At the end of 485 hours the three remaining unwelded elements were discharged due to the formation of pits and pinholes in the braze zone, as seen in Figure 4.23. These pits were probably caused by erosion of braze metal from over gas voids in the braze. After 1286 hours the welded specimens were removed due to the formation of a heavy, white, loosely adhering corrosion product. Break-away corrosion as shown in Figure 4.24 had occurred between 485 and 1286 hours. This corrosion may have been accelerated by exposure to rupture products during the first 152 hours of flow exposure. This test shows the value of a welding in removing gas bubbles formed during brazing. Several autoclave ruptures occurred which indicate that certain gaseous impurities drastically reduce the corrosion resistance of the reference brazing alloy. Figure 4.25 shows a ruptured brazed fuel specimen which had been autoclaved for 20 hours in 1500 psi 400 C steam. Prior to autoclaving the braze was smooth and unbroken. An analysis of the braze alloys showed an abnormally high oxygen content. Braze alloy from the same batch was vacuum outgassed and specimens behaved satisfactorily during autoclaving. As a final test a fuel element was brazed with the high oxygen containing braze on one end and the outgassed braze on the other and both ends were welded. This element was then autoclaved for 20 hours in 400 C 1500 psi steam. The element ruptured on the end brazed with the high oxygen content alloy, as shown in Figure 4.26. Gas content of the brazing alloys are given in Table 4.5.

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1/2 X

FIGURE 4.22

Brazed Fuel Section Which Ruptured in Less Than 152 Hours of Reactor Flow Conditions. The Braze Heat Affected Zone is Heavily Deformed

1 X

FIGURE 4.23

Pinholes Which Developed After 485 Hours of Exposure to Reactor Water Conditions. These Pinholes were Probably Due to Erosion of Braze Metal From Over Gas Voids in the Braze Metal.

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FIGURE 4.24

Brazed and Welded Fuel Specimens Which Showed Break-Away Corrosion in the Weld-Braze Region After 1286 Hours Exposure to Reactor Water Conditions. 1 X

FIGURE 4.25

Brazed Fuel Section Which Failed in a Steam Autoclave in Less than 20 Hours. The Braze Alloy was Abnormally High in Oxygen Content (1800 ppm) 1 X

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Brazed with Alloy Containing
1800 ppm Oxygen

FIGURE 4.26

Fuel Element Brazed with 1800 ppm Oxygen BeZirc Alloy on One End (Top) and 900 ppm Oxygen BeZirc Alloy on the Other (Bottom). During Autoclaving the End with the High Oxygen Containing Braze Alloy Ruptured, Thus Indicating the Adverse Effect of Oxygen on the Braze Alloy. The Element was Welded Prior to Autoclaving. 1 X

Brazed with Alloy Containing
900 ppm Oxygen

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Centimeter

Inches

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TABLE 4.5

GAS CONTENT OF BRAZING ALLOYS
(Parts per Million)

<u>Specimen</u>	<u>Oxygen</u>	<u>Hydrogen</u>	<u>Nitrogen</u>
Base Metal	1015	25	64
High Oxygen Braze	1800	60	81
Outgassed Braze	940	85	98

Work has continued on alternate brazing alloys which either melt at a lower temperature than the reference alloy or that do not contain beryllium. The first of the alloys which looks promising is one composed of 12 Fe + 4 Be + 84 Zircaloy-2. This alloy melts around 900 C, brazes at 950 C and readily wets both uranium and Zircaloy. Also, this alloy shows exceptionally good short-term corrosion behavior. The resulting autoclave film is extremely black and tenacious, similar to that of Zircaloy-2. The only disadvantage of this alloy is its brittleness. A production test consisting of KER size tube elements with a 1.6 per cent enriched 2 w/o zirconium core is being brazed, with the 12 Fe + 4 Be + 84 Zircaloy-2 alloy. Brazing is performed by heating to 950 C in vacuum, holding for 60 seconds, then vacuum cooling. The closures are surprisingly free of voids and extremely well bonded. The braze alloy is placed under the cap and brazed with a downward pressure of 15 psi on the cap.

Alternate brazing alloys are being prepared. These include:

Zircaloy-2 + 8 Cr + 8 Ni

Zircaloy-2 + 8 Cr + 8 Fe

Zircaloy-2 + 6 Cr + 6 Fe + 6 Ni

These alloys are the same as those proposed by McCuaig of ANL for brazing Zircaloy.

Fuel Rupture Tests in ETR - R. L. Call, D. C. Kaulitz

The radiometallurgy examination of the defected fuel element from the fourth ETR rupture test has been completed. This test is one of a series of "push-button" in-reactor rupture tests which is being performed at the ETR. These tests are being performed to evaluate the rupture characteristics of the fuel proposed for the Hanford New Production Reactor (NPR). The tests will determine the uranium corrosion rate and the failure delay time or "incubation period" of irradiated and unirradiated fuel.

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The GEH-3X3P7 loop in which the rupture tests are being performed, the rupture mechanism, the rupture detecting instruments and the results of the first three tests of the series have been described in previous quarterly reports. The fuel elements used in the fourth rupture test consisted of two sets of concentric tubes, each 14-3/4 inches long. The outer tube of each set had an OD of 1.82 inches and ID of 1.45 inches. The inner tube of each set had an OD of 1.05 inches and an ID of 0.52 inches. The tubes were formed of natural uranium clad in 20 mils of Zircaloy-2 by coextrusion. The defect (which was opened in the reactor by remote control) was on the outer surface of one of the outer tubes.

The fuel elements for this test had not been irradiated prior to their being charged into the ETR. After they were charged, the reactor was operated at full power for approximately 10 hours. The defect cap was then sheared off and the reactor operated for 30 minutes more. Temperature conditions at the defect during the test were:

Local bulk water temperature	-	262 C
Cladding surface temperature	-	301 C
Uranium surface temperature	-	322 C
Uranium core temperature	-	343 C

The incubation period for this test was 20 minutes; that is, for the first 20 minutes after the defect cap was sheared off, there was no increase in the coolant water radioactivity in the loop. Over the next ten minutes there was a uniform rise in radioactivity to a level which required the loop to be shut down.

Figure 4.27 shows the defected piece and the rupture mechanism "as received" in the radiometallurgy cell. The figure shows that considerable corrosion of the uranium and deformation and cracking of the outer cladding took place after the defect cap was sheared off. The damage to the chisel of the rupture mechanism (visible at the left in Figure 4.27) occurred after the test was discharged from the reactor. The weight of the uranium converted to oxide was calculated to be 43 grams.

Figure 4.28 is a cross section through the defect. The extent of the uranium corrosion can be seen here. Note that the corrosion extended out along the outer cladding much more rapidly than it penetrated through the uranium toward the inner cladding.

Figure 4.29 shows the upper left corner of the corrosion pocket shown in Figure 4.28. The cladding here is cracked from the inner side. Figure 4.30 is a higher magnification composite, photomicrograph of the upper left corner of Figure 4.29. Note that the corrosion is extending along the cladding-to-core bond line.

An ex-reactor test has been run with identical material and under identical conditions except that the water and uranium temperature were 300 C. A comparison of the incubation periods and corrosion rates shows that the incubation period is a little shorter and the corrosion rate about four times faster in-reactor and ex-reactor.

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4.38

FIGURE 4.27

A Defected Tubular Fuel Element Exposed In-Reactor to 260 C Water
for 30 Minutes After the Defect Cap was Sheared Off ~0.3 X

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4.39

HW-69085

FIGURE 4.28

A Cross Section Through the Defect of a Tubular Fuel Element
Exposed In-Reactor to 260 C Water for 30 Minutes After the Defect
Cap was Sheared Off. As Polished ~2 X

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4. 41

250 X

FIGURE 4. 30

A Higher Magnification Composite Photomicrograph of the Upper Left Corner of Figure 4. 29. Etched by Ion Bombardment

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DECLASSIFIED 4.12

HW-69085

FACILITIES AND EQUIPMENT

Fuel Element Measurement - K. R. Merckx

In an effort to find accurate geometrical descriptions of test fuel elements, development work was done on fuel element measuring devices. A method of characterizing the external surface of cylindrical fuel elements has been developed. Displacement readings from linear differential transformers mounted on the carriage of a lathe are continuously recorded while the fuel element is rotated in the lathe. Readings are made at different axial locations or stations. Both radial displacements and diameter readings are recorded for each axial station. The graphs of these readings for each axial station are used to determine eccentricity from an axis of rotation and the angular location of this eccentricity as well as mean diameter, ovality and angular location of the axis of ovality. Measurements and analyses of sample readings have been successfully completed. This method of measurement is being used on fuel elements being prepared for reactor testing. No method of making internal measurements on tubular fuel elements has been perfected. Operational requirements on warp, diameter, and thickness readings have been established and commercial equipment capable of making suitable measurements is being sought.

REFERENCES

1. Quarterly Progress Report, Fuel Development Operation, April, May, June, 1960, HW-67904. July 15, 1960. (SECRET)
2. Salvaggio, G. J., R. E. Johnson and S. W. Parembka. Mechanical Properties of Irradiated SIW Core 3 Zircaloy-2, WAPD-TM-184. November, 1959.
3. Brite, O. W. and R. J. Anicetti. High Energy Impact Forming of UO₂. See Section 5 of this Quarterly Progress Report.

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CERAMIC FUELS DEVELOPMENTFABRICATION DEVELOPMENTSpecial Fuel Elements and Components for PRTR Start-up Tests -
M. K. Millhollen

Four, full-size, 19-rod cluster elements were fabricated for PRTR start-up tests. Three elements for preliminary loading tests contained lead powder swaged in Zircaloy-2 cladding. One element for critical tests contained lead-cadmium powder swaged in Zircaloy-2 cladding.

A fifth special fuel element of UO_2 swaged in Zircaloy-2 cladding was assembled for physics tests. In this element, the center rod and one rod in each of the inner and outer rings of rods were fabricated with two special closures so that the three rods can be disassembled from the element. Each of the special threaded closure assemblies also serves as a measurement foil container. This fuel element has a detachable bottom end hanger. The outer wire wrap of the bundle is not welded to the top and bottom of the fuel element and is readily removable.

Ten neutron source capsule-holders were fabricated and welded to ten Class II, swaged UO_2 19-rod cluster elements, Figures 5.1 and 5.2. These containers can be disassembled and reassembled remotely and will contain plutonium-beryllium and radium-beryllium sources. The center rods of the capsule-holder assemblies were welded to the center posts of the fuel element bottom hanger fittings.

Hot-Swaging of UO_2 - P. L. Farnsworth

Hot-swaging continued to show promise for compacting to high densities UO_2 having poor packing characteristics. For example, minus 65 mesh fused UO_2 which can be compacted on a Syntron vibrating table to only ~60 per cent TD and can be cold-swaged to ~82 per cent TD was hot-swaged at ~850 C to ~93 per cent TD. This UO_2 was contained in Zircaloy-4 tubes which were resistance heated to 850 C just prior to swaging. Zircaloy-4 clad fuel rods containing electrodeposited UO_2 (-48 mesh) were heated by the same method to 850 C and swaged to 91 per cent TD.

Modifications made on the resistance heating apparatus significantly improved fuel rod heating characteristics and temperature control. However, arcing occurred between the resistance heating contacts and the fuel rods, and produced pits which could not be removed by surface grinding without exceeding the wall thickness variation tolerance.

Analyses of hot-swaged Zircaloy-4 cladding revealed high concentrations of hydrogen and oxygen on both the outer and inner surfaces and high concentrations of hydrogen throughout the cladding wall. These results indicated that hydrogen and oxygen were picked up both from the UO_2 and from the atmosphere and that the hydrogen diffused readily through the hot cladding.

FIGURE 5. 1
Source Holders

FIGURE 5.2

Source Holder Attached to PRTR Fuel Element

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Equipment Development. The resistance heating apparatus which, as an alternate to induction heating, is used to heat fuel rods prior to swaging was modified to include three contact brush holders, rather than two. This arrangement is shown in Figure 5.3. A pre-heat area between the first and second brush holders in which the fuel rod temperature is raised to the swaging temperature, and an area between the second and third brush holders in which the temperature is maintained within ± 50 C of the desired swaging temperature are provided. Using only two brush holders, the "at temperature" zone was about four inches long. With three brush holders, the "at temperature" zone is about 14 inches. The fuel rod is, therefore, at temperature for a longer time, allowing the temperature of the UO_2 to approach that of the cladding. Higher and more uniform UO_2 bulk densities were obtained.

Satisfactory fuel rod temperature control was achieved by maintaining a constant feed rate of the fuel rod through the heating contacts and a constant power output from the motor generators. The temperature detectors and power output controls were used only to prevent temperature overshoots should the motion of the fuel rod through the heating contacts decrease or cease. This modification eliminated the cycling associated with control of fuel rod temperatures by controlling motor generator power output.

Copper bonded tungsten carbide resistance heating contact brushes were substituted for the graphite brushes which often overheated during operation. Copper bonded tungsten carbide brushes have higher thermal and electrical conductivities than the graphite brushes. No excessive heating of the copper-tungsten carbide brushes was observed.

Asbestos rope which was used as end plugs to contain UO_2 in the fuel rods was replaced by Fiberfrax (or Refrasil). During heating, the asbestos released water vapor which often ruptured the cladding.

Process Evaluation. Zircaloy-4 clad fuel rods containing -100 mesh fused UO_2 , -65 mesh fused UO_2 , and electrodeposited (-48 mesh) UO_2 were resistance heated and swaged at ~ 850 C. The bulk UO_2 densities after loading, after cold-swaging, and after swaging at 850 C are compared in Table 5.1.

TABLE 5.1
EFFECT OF HOT-SWAGING ON BULK DENSITIES OF
VARIOUS TYPES OF UO_2 *

<u>UO_2 Type</u>	<u>Loaded Density (Per Cent TD)</u>	<u>Cold-Swaged Density** (Per Cent TD)</u>	<u>Hot-Swaged Density** (Per Cent TD)</u>
-100 mesh fused	55	81	91
-65 mesh fused	60	82	93
electrodeposited (-48 mesh)	63	88	91

* Clad in Zircaloy-4

**Reduced from 0.750-inch OD to 0.620-inch OD cold, followed by reduction from 0.620-inch OD to 0.565-inch OD at 850 C.

FIGURE 5. 3
Resistance Heating Apparatus

The density of the cold-swaged, electrodeposited UO_2 (88 per cent TD) was better than anticipated. Fused UO_2 of similar particle size distribution was cold-swaged to 84 per cent TD. Particle shape may account for the observed difference. Figure 5.4 shows photomacrographs of as-received electrodeposited UO_2 , de-agglomerated electrodeposited UO_2 , and crushed, fused UO_2 . The electrodeposited UO_2 particles are more spherical than the randomly shaped fused UO_2 particles.

The density variation, Figure 5.5, in a hot-swaged Zircaloy-4 clad fuel rod containing -100 mesh fused UO_2 was less than ± 0.5 per cent TD. The wall thickness ranged from 0.033 inch to 0.038 inch.

Hot-swaged (850 C), cold-swaged, and as-received Zircaloy-4 cladding was analyzed for hydrogen content. The results are plotted in Figure 5.6. Conclusions which can be drawn from the results are:

1. The Zircaloy-4 cladding acts as a getter for hydrogen during hot-swaging at 850 C.
2. Hydrogen diffuses through Zircaloy-4 at 850 C. The hydrogen content of the material has increased throughout the cladding wall.
3. Hydrogen is picked up from both the sorbed gases in the UO_2 and the external atmosphere.

Figure 5.7 shows a transverse cross-section of Zircaloy-4 cladding after hot-swaging at 850 C. The four dark areas seen in the photomacrograph are transformed beta phase and indicate the areas which attained the highest temperatures (> 900 C). From the pattern, it appears that these areas are the points where the fuel rod was in contact with the resistance heating brushes. No precipitated hydrides were seen in the specimen. Rapid quenching of the Zircaloy-4 in the swager dies prevented precipitation and growth of the zirconium hydride. The hydrogen is, therefore, either in solution or present as fine precipitate, too small to be determined optically. Contamination from the external atmosphere can probably be decreased by flooding the hot fuel rod with an inert gas during swaging. Reduction of the gas contamination on the inside surface would require vacuum outgassing of the UO_2 followed by tube loading in an inert atmosphere.

Tensile and corrosion specimens of as-received Zircaloy-2, as-received Zircaloy-4, cold-swaged Zircaloy-4, and hot-swaged Zircaloy-4 are being prepared.

Hot-Swaged, PRTR Fuel Rods - Fuel rods for 19-rod cluster PRTR fuel elements were hot-swaged at ~ 850 C. These fuel rods contained -65 mesh fused UO_2 clad in Zircaloy-4. Bulk densities after hot-swaging were 93.0 ± 0.5 per cent TD, and wall thicknesses were 0.035 ± 0.003 inch. Fuel rod diameter was $0.566 \text{ inch} \pm 0.003 \text{ inch}$.

Fluorescent dye penetrant, microscopic and visual inspection of these fuel rods revealed considerable surface pitting. This pitting resulted from arcing between the fuel rods and the contact brushes. The defects could not

As-Received Electrodeposited UO_2

De-agglomerated Electrodeposited UO_2

Crushed Fused UO_2

FIGURE 5.4

Typical Particle Shapes of Electrodeposited UO_2 and Fused UO_2 20 X

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FIGURE 5. 5

Bulk Density and Wall Thickness Variations in Zircaloy-4 Clad Hot Swaged Fuel Rod Containing -65 Mesh Fused UO₂. Swaging Temperature ~850 C

FIGURE 5. 6

Effect of Swaging Temperature on Hydrogen Pickup in Zircaloy-4 Cladding Tubes

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FIGURE 5.7

Heat Pattern and Structure of Resistance Heated Hot-Swaged Zircaloy-4
Cladding

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be removed by surface grinding without exceeding the wall thickness tolerance. The fuel rods will be used by the Chemical Development Operation to determine the dissolution rates of hot-swaged UO_2 in which the particles are partially sintered.

Vibrational Compaction Studies - J. J. Hauth and R. J. Anicetti

To investigate further the effect of particle size composition on the density of vibrationally compacted fuels, three size fractions of fused UO_2 were prepared. The size ranges selected for the coarse, medium and fine fractions were, respectively, -6+12 mesh, -35+65 mesh, and -200 mesh. Particle size compositions investigated ranged from 55-75 w/o coarse, 2.5-25 w/o medium, and 15-35 w/o fine. Each fraction was separately blended before preparation of the various mixtures to insure uniform distribution of particle sizes in each fraction. Screen analyses (Table 5.2) of the separate fractions and of one composite, showed that considerable attrition of the granular material occurred during blending.

TABLE 5.2

PARTICLE SIZE DISTRIBUTION OF FUSED UO_2 AFTER CRUSHING
AND BLENDING

Mesh Size (Tyler Series)	Weight Per Cent			Composite 60 w/o C, 15 w/o M, 25 w/o F
	Coarse (-6+12)	Medium (-35+65)	Fine (-200)	
-6+8	13.2			8.4
-8+10	46.5			23.7
-10+12	16.7			10.7
-12+35	23.6	0.2		16.2
-35+48		14.8		2.5
-48+65		41.8		6.5
-65+200		43.2	2.4	6.6
-200+270			8.9	2.5
-270+325			33.0	15.0
-325			55.7	7.9
Particle Density, g/cc	10.72	10.69	10.62	10.68

Eighteen mixtures were loaded and compacted in 0.505-inch ID Zircaloy-2 tubes on the MB C-50 shaker (5,000 pounds-force rated thrust). The frequency (75 to 4,000 cps) and acceleration (to 100g at the shaker table) were manually controlled. The tubes were vibrated as the UO_2 was loaded, and after loading until the rate of densification appeared to be negligible. Each rod was vibrated for approximately seven minutes, from the beginning of the tube filling through final compaction. The highest UO_2 bulk density (90.5 per cent TD) under these conditions was achieved with the mixture composed of 62.5 w/o coarse, 17.5 w/o medium, 20 w/o fine, for a compaction efficiency* of 93 per cent. Densities of all rods were greater than 88 per cent TD, and most were greater than 88.5 per cent, indicating that wide variations in particle size composition have a relatively small effect on bulk density.

A program was conducted in cooperation with Atomics International for vibrational compaction of uranium carbide in stainless steel tubes.

Fused UC (-4 mesh), supplied by AI, was crushed, screened and blended under an argon atmosphere. Particle size composition selected for compaction was as follows:

50.0 w/o	-6+10 Mesh (Tyler)
12.5 w/o	-10+20
17.5 w/o	-35+65
20.0 w/o	-200

The average particle density was approximately 95 per cent TD.

The UC was loaded and compacted---in an argon atmosphere. Five 9/16-inch OD, 0.010-inch wall stainless steel clad rods shown in Figure 5.8 were fabricated. Four were 2-feet long, and the fifth was 8-feet long. Bulk fuel densities are listed in Table 5.3.

TABLE 5.3

DENSITY OF VIBRATIONALLY COMPACTED UC

Rod No.	Length (ft)	Bulk Density		Compaction Efficiency Per Cent
		g/cc	Per Cent TD**	
1	2	11.65	85.5	90.0
2	2	11.58	85.0	89.5
3	2	11.67	85.6	90.1
4	2	11.63	85.3	89.8
5	8	11.69	85.8	90.3

* Ratio of density of compacted fuel to particle density of the powder employed, times 100.

**Based on a theoretical UC density of 13.63 g/cc.

FIGURE 5.8

Uranium Carbide Fuel Rods Fabricated by Vibrational Compaction
of UC in 0.010-Inch Wall Stainless Steel Tubes

Welded end closures were effected without difficulty, using specially designed end caps.

Facilities. The new vibration system, shown in Figure 5.9, was installed and operated to its rated output of 5,000 pounds force. The system will be used for fabricating multiple rod assemblies and massive fuel elements, such as the tubular assembly shown. Instrumentation shown in the control console permits automatic and continuous variation of displacement and acceleration throughout the frequency range of 5 to 3,000 cps. Maximum values of displacement, velocity and acceleration for continuous operation are, respectively, $\pm 1/2$ inch, 70 in/sec, and 125 g.

Fixtures designed and being evaluated for remote fabrication of partially decontaminated recycled fuel materials include an air chuck to accommodate 0.565-inch OD rods, and a fuel loading device which incorporates a means for determining remotely the level of the fuel material during vibrational compaction.

High Energy Impact Forming of UO_2 - D. W. Brite, R. J. Anicetti

A technique for subjecting heated UO_2 powder to a high energy impact of 500,000 psi or greater, using modified Bridgman anvils and a mild steel reinforcing ring, was described in the preceeding quarterly report.⁽¹⁾ The bulk density of the compacted UO_2 was greater than 99 per cent of the theoretical UO_2 density.

Resistance heating of the stainless steel capsules containing the UO_2 while the capsule was in place in the impact die, was not successful. Although the capsule was insulated from the steel retaining ring, the ring nevertheless became hot enough to flow excessively upon compaction. The UO_2 was extruded into the retaining ring, and the anvil faces almost came together. Densities obtained, using compacting pressures which has previously resulted in UO_2 densities of >99 per cent TD, were 96.4 and 93.5 per cent TD for micronized and PWR type UO_2 , respectively. Apparently the mild steel retaining ring must be cold to retain sufficient strength and to function as a restraining ring.

Electron micrographs of UO_2 compacted by high energy impact at 1200 C reveal the micropores caused by gas trapped in the UO_2 powder. The capsules containing the UO_2 were sealed in vacuum, after outgassing at 600 C. The excess oxygen in the nonstoichiometric UO_2 apparently was liberated at the higher temperatures of impaction (>1000 C). UO_2 capsules which were impacted at 1300 C and at 800 C resulted in UO_2 having a bulk density of 94.2 per cent TD and >99 per cent TD, respectively.

Figure 5.10 is an electron micrograph of the 94.2 per cent dense sample. Many small pores are apparent at the grain boundaries. Where transgranular fracture occurred, no pores are visible.

High energy impacted UO_2 specimens (90-95 per cent TD) were sintered in hydrogen for 12 hours at 1750 C. Sintered densities of the UO_2 were as high as 99.6 TD. Photomicrographs of the samples having densities of 99.6 and 96.2 per cent TD appear in Figure 5.11. The long crack in the sample of 96.2 per cent TD existed in the compacted UO_2 before sintering.

FIGURE 5.9
5000 Pound-Force Vibration System

5000X

Electron Micrograph, Fracture Surface

FIGURE 5.10

Microstructure of UO_2 Heated to 1300 C and Compacted
by High Energy Impact to 94.2 Per Cent TD

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1000X

Compacted to 94.5 Per Cent TD

250X

Compacted to 86.3 Per Cent TD
Sintered to 96.3 Per Cent TD

250X

Compacted to 95.2 Per Cent TD
Sintered to 98.8 Per Cent TD

250X

Compacted to 92.2 Per Cent TD
Sintered to 99.6 Per Cent TD

Polished and Etched

FIGURE 5.11**Microstructure of UO_2 Compacted by High Energy Impact
and Sintered 12 Hours in Hydrogen at 1750 C**UNCLASSIFIED

Welding Fabrication Development - L. R. Vancott, L. E. Mills

Recycle Fuel Rod Closure. The tungsten arc inert gas welding process was employed for remote welding of closures on Zircaloy cladding filled with recycled UO_2 fuel. A glass bottle torch was developed to provide the high purity inert atmosphere required for welding Zircaloy. The torch is 3-inches in diameter by 2-1/2 inches long with the tungsten welding electrode at a 45 degree angle to the center locating plug (Figures 5.12 and 5.13). The centering plug aligns the electrode and bottle with the fuel element and maintains proper arc length as the element rotates within the enclosure. An argon atmosphere is supplied through the top of the closure at four cfh through a porous bronze diffuser.

A copper chill block is fitted to the fuel element to decrease heat conduction to the fuel, thus minimizing out-gassing. The chill block also closes the bottom opening in the bottle and provides a tighter inert gas system.

Total heat input to the rod is also minimized by making the complete closure in 2.4 seconds (44 inches per minute).

Spacer Attachment by Resistance Seam Welding. A three-phase low-frequency converter resistance seam welder was investigated for welding round (0.072-inch diameter) and rectangular (0.075 inch by 0.135-inch high) spacer ribs on Zircaloy tubing (3-1/16 inch OD by 0.060-inch wall).

The welding parameters that produced the most satisfactory weld on 0.072-inch diameter wire were 100-pounds electrode force, continuous electrode rotation, an intermittent weld current pulse of 1/180-second weld time using 95 per cent capacity of the 150 KVA machine and 1/20-second cool time. The short weld pulse followed by the rapid cooling allowed the welds to be made without rigid inert atmosphere protection. Some atmosphere contamination was encountered with this procedure but was easily removed with light wire brushing. Weld strength was evaluated by shearing a 1-inch section of the welded rib from the tube. A shear strength of 425 pounds per inch was average. The pressure and heat deformed the rib during formation of the weld. This deformation resulted in an increased contact width between the rib and the tube. The contact width increased at a faster rate than the growth of the weld nugget and resulted in a contact width at least three times the width of weld. Thus, a crevice equal to the width of the weld appeared on each side of the weld. A typical weld area is shown in Figure 5.14. The etch solutions used prior to autoclaving could not be consistently rinsed from the crevice and resulted in corrosion during autoclave tests.

The rectangular rib material was machined to various shapes to evaluate the effect of rib shape on weld quality. The optimum rib shape was a diamond configuration that provided a rib to tube contact width of 0.020 inch and a 60 degree included angle top section. The tapered upper section mated an electrode and gave excellent pressure and contact characteristics. The shear attained with this rib was 800 pounds per inch of weld.

FIGURE 5.12
Fully Assembled Fixture

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Bottle Type Torch with Chill Block and Fuel Rod

FIGURE 5.13

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50 X

FIGURE 5.14

Photomicrograph of 75-Mil Wire on 3.062-Inch OD Zircaloy-2
Tube. Note the Deformation of Wire and Deep Crevice on Each Side

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The rib deformation that was encountered in the round wire was also encountered with the rectangular wire and resulted in a crevice on each side of the weld.

High Frequency (450 KC) Resistance Welding. Attachment of spacer ribs (0.072-inch wire) to 9/16-inch diameter stainless steel tube by high frequency resistance welding was the objective of the following tests. Wire was imultaneously wrapped on the tube and continuously welded at a 10-inch helical pitch.

Mechanical development was first necessary to provide uniform velocity of the work piece, internal support mandrel for the tube directly under the weld area and mechanical guide supports for the tube and wire.

The argon atmosphere purge box system was replaced by a localized porous bronze diffusion baffle which provides a uniform laminar gas flow over the weld area. A liquid CO₂ quench nozzle was located adjacent to and after the argon atmosphere protection.

Weld quality was based on (1) the crevice formed between the wire and tube, (2) expulsion of metal or beading along the joints, (3) continuity of weld, (4) width of weld and (5) shear load (pounds per inch) of rib to tube for a 1/2-inch section.

Welding parameters, and their effects, are summarized:

1. Contact pressure of wire to tube had very little effect on balancing heat in the wire and tube but it did have an effect on wire deformation.
2. Relative position of the electrodes had a definite effect on the quantity of heat in each member being welded. A marked difference in heating rate was produced by placing the wire electrode 5/8-inch and the tube electrode 1-3/8-inch from their contact points on the wire and the tube, respectively. The optimum setting was not determined; however, electrode positioning will be of primary concern in future tests.
3. The usable welding current range was very narrow. The current carrying capacity of the 0.075-inch diameter wire determined the maximum welding current that could be employed. That maximum current was required to produce a reasonable weld. A weld current five per cent less than maximum produced no weld. Even the maximum welding current produced a weld having a large crevice to weld nugget width ratio because of flattening of the wire and of the relatively small amount of heat generated in the tube wall. The uneven heating resulted in expulsion of small amounts of molten surface metal from the wire to form an intermittent bead along the weld joint. Figure 5.15 shows evidence of insufficient heating of the tube, slight deformation, metal expulsion and severe crevice formation.

FIGURE 5.15

Photomicrograph of 72-Mil Wire on 9/16-Inch OD Stainless Steel Tube. Note Expelled Metal and Lack of Heat in the Tube

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Speed of weld was held constant (92 ft/min) to determine the effect of other variables. Because a desirable heat balance could not be established by varying current, pressure and electrode position, higher speeds were indicated. By increasing speed and using unequal electrode positioning the ΔT during heating of the individual components should diverge and yield more sensitive heat control.

Conclusions from the above tests are:

1. Round wire spacer ribs were not satisfactorily welded to fuel cladding.
2. Greater speed of weld must be used to increase sensitivity for heat control.
3. Varying current, relative electrode position and speed have the greatest effect on weld formation. Contact pressure has very little effect.

Magnetic-Force Resistance Butt Welding - L. E. Mills

The thermal conductivity of the atmosphere surrounding the weld during magnetic-force resistance butt welding must be considered a welding parameter. The current required for welding in a helium atmosphere is approximately two per cent greater than welding in air, and four per cent greater than welding in a vacuum.

The following welding schedule was developed for welding closures on 0.563-inch OD by 0.030-inch wall Zircaloy clad fuel rods.

Welding time: 1/60 second

Welding Current: 20,000 amperes for 1/120 second (1/2 cycle)
24,000 amperes for 1/120 second (1/2 cycle)

Initial Force: 550 pounds

Magnetic Gap: 7/16 inch

Atmosphere: Helium

Complete evaluation was performed on fifty welds made with this welding schedule. The following table summarized the various tests and results obtained in the evaluation.

TABLE 5. 4

SUMMARY OF VARIOUS TESTS AND RESULTS

<u>Tests</u>	<u>No. of Welds</u>	<u>Results</u>
1. Helium Leak Test	50	No leaks
2. Radiograph	50	No defects
3. Autoclave Test (450 C - 1150 psi steam for 72 hrs.)	50	(a) Occasional grey corrosion areas
4. Internal Pressure Test	4	(b) Average bursting pressure - 7990 psi
5. Direct Tensile Test	3	(c) 2110 lbs average, (39, 900 psi)
6. Micro Hardness	4	DPH no. 188, cap 225, weld area 194, tube
7. Micro-Section Metallurgical Examination	10	(d) Fine grain hot forged structure

(a) The occasional grey corrosion was caused by copper contamination from the collet electrode gripping the rod. The weld upset metal must be machined to a depth of 0.002-inch below the original interface to remove all of the copper contamination.

(b) In each case the failure occurred as a split in the cladding.

(c) The tensile fracture occurred partially within the weld and partially in the base metal.

(d) Fine grain hot-forged structure is formed in both cap and the tube at the weld. The thickness of the fine grain structure is 0.050 inches. The original interface line could be distinguished occasionally within the fine grain material.

The very short welding time of 1/60 of a second requires very accurate control over all parameters which contribute to the success of the weld. Close part tolerances must be maintained and the weld surfaces must be flat and clean.

The conventional nondestructive testing methods (radiograph, eddy current and ultrasonic) do not readily lend themselves to evaluation of the resistance butt weld. Accurate monitoring of the 1/60 of a second weld current and voltage will be evaluated as a means of assuring weld quality.

The initial concept of the magnetic-force resistance welding process indicated that it would be limited to welding materials with relatively low electrical conductivity, such as steel, stainless steel and Zircaloy. Preliminary welds have been made on SAP material, with indications that the process may be applicable for making closure welds on "sintered aluminum products".

FUEL EVALUATION

Prototypic PRTR Fuel Element Irradiation: GEH-12-4 - J. L. Bates,
G. R. Horn, M. K. Millhollen

A three foot long, Zircaloy-2 clad, swaged UO_2 prototypic PRTR 19-rod fuel assembly (Figure 5.16) was irradiated in the ETR. The test was terminated at the end of 24 days because of fission product release into the coolant. The maximum heat transfer rate was approximately 130 per cent of the maximum permissible PRTR fuel element heat transfer rate. Descriptions of the fuel element and of irradiation conditions is provided in Tables 5.5 and 5.6.

TABLE 5.5

PROTOTYPIC PRTR FUEL ELEMENT, GEH-12-4

Dimensions (inches)

Total length fuel element	46.31
Diameter fuel element (without wire wrap)	3.047
(with wire wrap)	3.187
OD fuel rod	0.563
Zircaloy-2 cladding thickness	0.029
Diameter of Zircaloy-2 wire wrap	0.072
Length of fuel per rod	34

Uranium Dioxide, Fused

Bulk density, g/cm^3	9.7
Enrichment, U^{235} , per cent	0.71

TABLE 5. 6

IRRADIATION DATA FOR PROTOTYPIC 19-ROD CLUSTER
PRTR FUEL ELEMENT IN C69G7 FACILITY, ETR, GEH-12-4

Coolant - water

Inlet pressure, psig	2000
Flow rate, gpm	125
Velocity, ft/sec	11.7
pH	8-9
Temperature (F)	
Inlet	458-477
Outlet	505-520

Exposure

Total irradiation - equivalent full power days	24
Total days irradiated	32

Heat Generation BTU/hr-ft²

Maximum

Start of irradiation	~350,000
End of irradiation	~525,000

Average

Start of irradiation	280,000
End of irradiation	405,000

The 24 day irradiation included 8 scrams and startups of the reactor. Activity was detected after 23.3 equivalent full power days of irradiation. The failure occurred as the fuel element attained maximum heat generation after a sudden scram resulting from other reactor tests. After the first indication of fuel element failure, the irradiation continued at full power for 15 hours, 7 minutes of which were at maximum coolant temperature. Activity increased gradually in the recirculating loop, finally requiring discharge of the fuel element. That was the first failure of a swaged UO₂ fuel rod during the Hanford irradiation program.

FIGURE 5.16

Prototypic, Zircaloy-Clad, Swaged UO_2 , 19-Rod PRTR Fuel Element

Irradiation of Vibrationally Compacted Nonhomogeneously Enriched UO₂
Fuel Cluster, GEH-4-44 - J. L. Bates

A 4-rod cluster fuel element containing enriched UO₂ was irradiated in a reflector position of the MTR in the GEH-4 loop. Each rod contained UO₂ fuel of the same average U²³⁵ content, but differed in the method of enrichment (Table 5.7). Dimensions of the fuel cluster and the irradiation conditions are listed in Tables 5.8 and 5.9, respectively. The element is awaiting post-irradiation examination.

TABLE 5.7

UO₂ COMPOSITION IN 4-ROD CLUSTER FUEL
ELEMENT: GEH-4-44

Rod No.	Fabrication Method	Type UO ₂	Enrichment w/o U ²³⁵ in U	Composition, Weight Per Cent		
				-6+10	-35+65	-200
1	Vibration	Sintered & Crushed	1.6	60	15	25
2	Vibration	Fused	0.71	60		
		Sintered & Crushed	2.9		15	25
3	Vibration	Fused	0.71	60	15	
		Sintered & Crushed	4.15			25
4	Sintered Pellets	Sintered	1.6	--	--	--

TABLE 5.8

GEH-4-44 IRRADIATION TEST - FUEL ELEMENT DESCRIPTION

Dimensions (inches)

Over-all length of fuel element	11.38
Rod diameter	0.565
UO ₂ core length	10.5
Fuel element diameter	1.44
Zircaloy-2 cladding thickness	0.03

TABLE 5.9GEH-4-44 IRRADIATION TEST DATA

Coolant - Water	
Pressure, psig	280
Flow rate, gpm	20
Velocity, ft/sec	11.7
Temperature, ° C	
Inlet	31
Outlet	41
pH	5.5-6.5
Exposure Days	15
Heat Generation Kw/ft per rod	
Average,	19.0
Maximum	~22.0
Surface Heat Flux, BTU/hr ft ²	
Average	490,000
Maximum	570,000

Swaged UO₂ Capsule Irradiation - J. L. Bates

Swaged UO₂ capsules have attained an estimated exposure of approximately 17,500 MWD/T in MTR-ETR.

Thin-Wall Stainless Steel Clad Fuel Rods (VBWR-2) - G. R. Horn and J. J. Hauth

Nine UO₂ fuel rods vibrationally compacted in thin-wall stainless steel tubes (38-inches long by 0.563-inch OD) were fabricated for irradiation in the VBWR. Four rods have a cladding thickness of 0.015-inch, three have a cladding thickness of 0.010-inch, and two have a cladding thickness of 0.008-inch. The fuel mixture had the following particle size composition:

TABLE 5.10PARTICLE SIZE COMPOSITION

<u>Mesh Size (Tyler)</u>	<u>w/o</u>
-3+20	60
-35+65	10
-100	30

The fines (-100 mesh) were sintered UO_2 (95 per cent TD) enriched to 10 w/o U^{235} in uranium. The remainder was natural fused UO_2 (~99 per cent TD). The average bulk density of the 0.015-inch clad rods was 86.1 per cent TD. The 0.010-inch clad and 0.008-inch clad rods averaged 88.5 per cent TD. All tubes were Zyglo fluorescent penetrant tested on the outside and inside surfaces prior to compaction. The finished rods were helium leak tested and autoclaved for one hour in water at 1000 psi and 280 C.

Four 0.005-inch thick stainless steel clad rods were also fabricated by the same techniques. The fuel density in these rods averaged 84.8 per cent TD.

During autoclaving, the cladding collapsed and formed broad-based ridges approximately 1 to 2 mils high along the length of the rod. One of these rods was successfully swaged, after autoclaving, to a final diameter of 0.547-inch. The ridges were eliminated with no indication of buckling or folding of the metal. No ridges were formed when the rod was autoclaved a second time. This rod will be sectioned and examined to determine the character of this inside surface.

The low densities, especially in the 0.005-inch clad rods, were due primarily to low particle density in the sintered fines and less than optimum particle size composition in both the coarse and fine fractions. By correcting these conditions it should be possible to vibrationally compact 0.005-inch clad fuel rods to densities high enough to prevent collapse in the autoclave. Fused UO_2 with a favorable particle size composition was compacted to 90-91 per cent TD, with no "dimpling" of the 0.0075-inch stainless steel cladding. The coarse fraction of the mixture was composed of smaller particles than those of the lower density mixtures which produced small dimples in the cladding during compaction.

GEH-12-20, Irradiation of Purposely Defected, Swaged UO_2 , 7-Rod Cluster Fuel Element - M. K. Millhollen, J. L. Bates, G. R. Horn

A 7-rod, swaged UO_2 cluster, purposely defected, was irradiated in the ETR. The fuel element consisted of a center rod with six rods equally spaced around it on a 1.285-inch diameter bolt circle. Four rods were clad with Zircaloy-2 and three with Zircaloy-4. The UO_2 fuel length in each rod was 32 inches, and the UO_2 bulk density in each rod was 85 per cent of theoretical. Rod diameters were 0.565-inch and the rods were spaced apart by 0.072-inch diameter Zircaloy-2 wire wrapped on four rods on a ten-inch pitch. The entire bundle was wrapped with 0.072-inch wire which was welded to the end brackets. Rods were leak tested, radiographed and autoclaved before assembly, Figure 5.17.

Two each of the Zircaloy-2 clad and Zircaloy-4 clad rods were defected by drilling a 0.006-inch diameter hole through the 0.030-inch wall cladding. In each type of cladding one rod was defected at the center and one two inches from the lower end. The assembly was thermocoupled to measure inlet and outlet coolant temperature.

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FIGURE 5.17

Seven-Rod, Swaged UO_2 Fuel Element

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The fuel element was irradiated in a pressurized water, recirculating loop in the reflector of ETR. The irradiation conditions are summarized in Table 5.11.

TABLE 5.11

IRRADIATION DATA OF GEH-12-20 IN R33P7 LOOP ETR

Coolant - Water

Inlet pressure, psig	2,000
Flow rate, gpm	60
Velocity	12.9
pH	9-10
Temperature (17 days) (°C)	
Inlet	103
Outlet	121
Temperature (last 28 hours) (°C)	
Inlet	497
Outlet	515

Exposure

Days at low coolant temperatures	16.5
Days at high coolant temperatures	1.3
Total full power days	17.8

Heat Generation, BTU/hr-ft²

Average	225,000
Maximum	400,000

Maximum Surface Temperature Zircaloy
Cladding

750 F

Maximum fission products release occurred during the first 24 hours at full reactor power, approximately 14 hours after reaching full power. The coolant temperature was approximately 120 F.

During the remainder of the test, including the increase to coolant temperatures above 500 F, the release of fission products was an order of magnitude less. The irradiation included four startups and ten scrams.

GEH-4-54 Defect Rod Test - M. K. Millhollen

For a direct comparison of hydride formation in Zircaloy-2 and Zircaloy-4 under identical reactor conditions, a 24-inch long fuel rod was fabricated by butt welding 12-inch sections of Zircaloy-2 and Zircaloy-4 together and then vibrationally compacting sintered and crushed enriched UO_2 (3.2 w/o U^{235} in U) in the tube. This material was compacted in the rod to a bulk density of 80.6 per cent of theoretical. The fuel rod was defected at the center of the butt weld by drilling a 0.005-0.006-inch diameter hole through the 0.030-inch wall, Figure 5.18. The test irradiation will be conducted in the GEH-4 facility of the MTR early in the next quarter.

 UO_2 Fuel Element Defect Tests (HD-1, HD-2, HD-3) - M. K. Millhollen, G. R. Horn

Irradiation of purposely defected, Zircaloy clad UO_2 fuel elements has led to a better understanding of Zircaloy hydriding. Several defected, 4-rod cluster UO_2 fuel elements will be irradiated in a Hanford reactor to supplement the short term data from ETR and MTR defect tests.

The test elements are 12-inches long with the four rods spaced 90° apart on a 1.250-inch diameter bolt circle, Figures 5.19 and 5.20. Each cluster consists of two Zircaloy-2 clad and two Zircaloy-4 clad fuel rods (0.565-inch OD by 0.030-inch wall). One rod of each of the two types of cladding material is defected at its mid-point with a 0.006-inch diameter hole. Each fuel element is placed in an aluminum basket before loading in reactor. The basket centers the element in the process tube and controls coolant flow over the element.

Three clusters were fabricated for irradiation. One contains sintered UO_2 pellets (HD-1); one contains swaged, sintered and crushed UO_2 (HD-3); and one contains swaged, fused UO_2 (HD-2). All fuel rods were leak tested and autoclaved before assembly.

The element containing sintered UO_2 pellets is now being irradiated. Essentially no activity has been detected in the effluent coolant.

Irradiation of Purposely Defected, Swaged UO_2 Elements - J. L. Bates

Although Zircaloy has excellent corrosion resistance to high temperature water, the formation of brittle zirconium hydride in defected UO_2 fuel elements irradiated at high surface heat fluxes may be a serious problem.

A series of four purposely defected, swaged UO_2 fuel rods with a 0.005-0.006-inch defects clad in Zircaloy, were irradiated in the MTR. (Table 5.12) Two of the rods were clad in Zircaloy-2 and two in Zircaloy-4 (nickel free Zircaloy-2). The fuel rods (0.557-0.559-inch in diameter) and 24 inches long, were irradiated as long as two hours in 100 F coolant water at maximum surface heat fluxes as great as 758,000 BTU/hr-ft².

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FIGURE 5.18
GEH-4-54 Fuel Rod Before Irradiation

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FIGURE 5.19

Hanford Defect Element in Centering Basket

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FIGURE 5.20

Hanford Defect Test Element and Centering Basket Before Assembly

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TABLE 5.12

IRRADIATION CONDITIONS FOR PURPOSELY DEFECTEDSWAGED UO₂ FUEL TESTS

	I (GEH-4-39)	II (GEH-4-45)	III (GEH-4-52)	IV (GEH-4-53)
Heat Generation(Kw/inch)				
Average*	1.32	1.77	1.69	2.00
Maximum**	1.78	2.37	2.28	2.70
Surface Heat Flux(BTU/hr-ft ²)				
Average*	370,000	495,000	473,000	562,000
Maximum	499,000	663,000	638,000	758,000
Length of Irradiation(min)				
Total	97	96	75	134
At maximum heat generation	45	42	22.5	40
At surface heat flux greater than 500,000 BTU/hr-ft ²	45	55	30	99
Burnup,a/o x 10 ⁴				
Calorimetrically	3.8	5.3	2.7	6.2
Radiochemically***	4.0	4.1	2.0	
Maximum Temperatures (C)				
Coolant				
Inlet	30	31	32	30
Outlet	42	47	47	48
ΔT	12	16	15	18
Zircaloy				
Outside surface	122	144	134	152
Inside surface	200	248	230	260
UO ₂ (estimated)	~2200	~2750	~2750	~2700

*Determined from Calorimetric Data

** Estimated from Calorimetric Data and time averaged perturbed neutron flux

*** Cs¹³⁷ counting method

The hydriding resistance of Zircaloy-4 was superior to the hydriding resistance of Zircaloy-2 (Figure 5.21). The Zircaloy-2 became hydrided over the entire length of the fuel element (Figure 5.22). The Zircaloy-4 became hydrided near the defect and in localized areas of increased cladding temperature (Figure 5.23).

No changes were observed in the general appearance of the surfaces or outside dimensions of the fuel rod. One rod warped approximately 0.15 inch. This bowing was attributed to an unusual flux distribution during irradiation.

During the irradiation, no fission products were released until the maximum surface heat flux exceeded 150,000 BTU/hr-ft². Bursts of fission products were released during other increases in heat generation with the maximum bursts occurring above 350,000 BTU/hr-ft². At constant power generations, the fission product release decreased with time to a relatively constant value approximately 1/4 to 1/5 of the peak release. Sudden bursts of fission products were released during reductions in heat generations (Figures 5.24 and 5.25).

The amount of fission products released varied depending upon the UO₂ used in swaging. A large percentage of fine UO₂ particles and non-homogeneous U²³⁵ enrichment significantly increased the release of fission products.

No particulate matter was released during irradiation.

The structure of the swaged UO₂ changed extensively during irradiation (Figures 5.26 and 5.27). The physical changes in the fuel core were similar to changes observed in other UO₂ fuel cores irradiated to longer exposures.

Details of these irradiation tests will soon be published in HW-65465.

BASIC STUDIES

High Temperature Behavior of Irradiated UO₂ - J. A. Christensen

A correlation between the high temperature behavior of irradiated UO₂ and in-reactor exposure has been established. The change of the melting point of uranium oxide irradiated to 15,000 MWD/T was measured by high temperature microscopy, and is shown in Figure 5.28. The melting point increased with exposure to a maximum of 2915 C ± 20 (mp of non-irradiated UO₂, 2790 C ± 15) at about 0.06 a/o burnup (1.4 x 10¹⁹ fissions/cc).

Motion pictures of the irradiated UO₂ specimen during heating revealed that volatility varied inversely with the melting point. Irradiated UO₂ vaporized rapidly at temperatures near the melting point and crystal growth by vapor deposition was observed as with nonirradiated uranium oxide. However, the irradiated UO₂ which had completely vaporized and redeposited also exhibited a higher melting point and lower vapor pressure than non-irradiated UO₂.

FIGURE 5.21

Summary of Hydride Distribution Along Length of Purposely Defected, Swaged UO_2 Fuel Rods

FIGURE 5.22

Hydride Distribution and Microstructure of Zircaloy-2 Cladding
of Longitudinal Cross Section at Defect of Purposely Defected,
Swaged UO₂ Fuel Rod Test I (GEH-4-39)

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0.010 Inch from Defect

0.075 Inch from Defect

0.140 Inch from Defect

0.210 Inch from Defect

0.275 Inch from Defect

FIGURE 5.23

Microstructure of Zircaloy-4 Cladding from Longitudinal Cross Section
0.10, 0.075, 0.140, 0.210, and 0.275 Inches from Defect of Purposely
Defected, Swaged UO₂ Fuel Rod Test IV (GEH-4-53) 250X, Bright Field

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FIGURE 5.24

Relative Gamma Activity Released from Purposely Defected, Swaged UO₂
Fuel Rod, Test I (GEH-4-39)

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FIGURE 5.25
Relative Gamma Activity Released from Deliberately Defected Swaged UO₂ Fuel Rod, Test GEH-4-45

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Test I

Test II

Test III

Test IV

FIGURE 5. 26

Cross Sections of Irradiated, Purposely Defected, Swaged UO_2 Fuel Rods Cross Sections at Defect (Polished)

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0.202-Inch: Termini of Honeycomb UO_2

0.167-Inch: Termini of Voids in
Columnar Grains

0.125-Inch: Termini Columnar Grains

0.090-Inch: Grain Growth

0.050-Inch: Initial Sintering

0.020-Inch: Circumferential Crack

0.000-Inch: Zircaloy-2- UO_2 Interface

FIGURE 5.27

Microstructure of Irradiated, Purposely Defected, Swaged UO_2 Fuel
Rod from Cladding to Central Cavity, Test II (GEH-4-45)

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FIGURE 5.28

Melting Point vs In-Reactor Exposure for Irradiated UO_2

Thermal Conductivity - J. L. Daniel

Evaluation of the influence of fabrication method of UO_2 fuel on its (nonirradiated) thermal conductivity continued. Measurements on a cold-pressed and sintered specimen gave thermal conductivity values intermediate between the two curves previously established for the isostatically pressed specimens and for the extruded specimens.

Additional specimens being prepared for future measurements include a vibrationally compacted sample and a large fused UO_2 crystal. Electrical conductivity measurements are being considered for correlating and interpreting thermal conductivity data previously obtained from irradiated UO_2 specimens.

Electron Microscopy - J. L. Daniel

The JEM 5Y/6A electron microscope was received, and preliminary installation of electrical and water supplies completed. Laboratory space and supporting equipment installation was also completed. Final assembly and testing of the microscope and accessories will begin October 1, coincident with the arrival of the vendor's physicist-engineer.

REFERENCES

1. Quarterly Progress Report, Fuels Development Operation, April, May, June, 1960, HW-67904. July, 1960. (Secret)

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