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**QUARTERLY PROGRESS REPORT
FUELS DEVELOPMENT OPERATION
JANUARY, FEBRUARY, MARCH, 1962**

APRIL 16, 1962

HANFORD LABORATORIES

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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QUARTERLY PROGRESS REPORT
FUELS DEVELOPMENT OPERATION
JANUARY, FEBRUARY, MARCH, 1962

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April 16, 1962

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

Work performed under Contract No. AT(45-1)-1350 between the
Atomic Energy Commission and General Electric Company

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PHYSICAL METALLURGYIRRADIATION EFFECTS IN REACTOR METALS

A tentative group of alloys has been selected for initial study on this program. These include HY-80, A302B, 304 SS, 348 SS, 406, AM355, Inconel Alloy 600, Inconel Alloy X-750, D979, Hastelloy X-280, and Zircaloy-2.

The in-reactor creep test at 310 C and 30,000 psi on a 20 percent cold worked Zircaloy-2 specimen was continued through a third reactor shutdown. A sigmoidal strain-time curve was again observed although the total strain occurring during the third shutdown was only 1/3 of that which occurred during the first shutdown. Activation energy measurements on this sample resulted in a value of 85,000 cal/mole during temperature increases and a value of 52,000 cal/mole during temperature decreases. The specimen failed at 5 percent strain.

It has proved possible to maintain a constant specimen temperature during reactor shutdown in in-reactor creep capsules containing internal water cooling coils by controlling the water flow in conjunction with the specimen heater power.

A total of 392 specimens, irradiated from 5×10^{19} to 8×10^{20} nvt, of Zircaloy-2 and types 304 and 347 stainless steel have been discharged from the G-7, hot water loop facility. Start-up of the out-of-reactor loop to duplicate the environment of the G-7 loop was achieved.

Tensile testing of unirradiated control samples of Zircaloy-2 has indicated that appreciable recovery is occurring during the preirradiation autoclaving treatment. This is more pronounced for the rolling direction than for the transverse direction.

By varying the rate of tensile straining from 2.1×10^{-3} to 2.1 min^{-1} , it was determined that Zircaloy-2 is strain-rate sensitive. The rate sensitivity is less at 300 C than at room temperature indicating strain ageing at 300 C.

A loss of ductility was observed to occur in ingot iron, decarburized iron and decarburized-nitrogenized iron tensile specimens after an exposure of only 10^{17} nvt. The decrease was greatest for the nitrogenized specimens.

High purity iron foils, 0.003-inch thick, have been received for transmission electron microscope studies.

SWELLING OF IRRADIATED FISSIONABLE MATERIALS

Four controlled temperature irradiation capsules, containing split hollow cylinders of high purity uranium, are operating at respective temperatures of 625, 625, 575 and 400 C. These capsules were charged in tandem, two to a test hole.

Four unmonitored, NaK-filled, ambient temperature capsules, each containing six uranium-to-uranium diffusion couples, completed their irradiation. Two of the capsules were opened and the specimens recovered.

Quantitative Metallography has revealed that the pores present in 0.41 at.% burnup uranium, irradiated at intermediate temperatures and under conditions of restraint, undergo drastic changes as a result of post-irradiation annealing at elevated temperatures. On the other hand, pores present in 0.3 at.% burnup uranium irradiated at high temperature under conditions of no restraint exhibit no change after postirradiation annealing at a temperature equivalent to the irradiation temperature.

NEUTRON DAMAGE TO METALS

A capsule containing four single crystal bend test specimens of molybdenum with varying carbon contents are being irradiated. Other capsules containing polycrystalline tensile, microhardness, resistivity, stored energy, and length change specimens have been submitted for irradiation. Capsules containing foil specimens have also been submitted.

Microhardness measurements on cold-worked molybdenum after isochronal and isothermal anneals have indicated a two stage recovery process, one with an activation energy of 2.2 ev and the other about 4 ev.

SUPPORTING STUDIES

Two capsules, each containing 30 notched and un-notched tensile specimens of Zircaloy-2, were sent to the ETR for charging in Cycle 46.

IRRADIATION EFFECTS IN REACTOR METALS

As reactor concepts become more diversified, it is increasingly difficult to provide the necessary information concerning the engineering properties of the structural materials presently being used or being contemplated for future use. Some information is available for a given set of conditions, but this information is not sufficient to satisfy the demands of present reactors and there is insufficient understanding of the damaging processes involved. There is a need for information about a greater number of materials irradiated under an increased variety of conditions. Furthermore, this information must be supplemented by fundamental data in order to understand the effects that have been observed so that predictions of property changes can be safely made for irradiation conditions somewhat different than those for which measurements have been made.

A program, which is a co-ordinated effort among several sites, has been initiated by the Division of Reactor Development of the AEC to determine the irradiation effects on several reactor structural materials. The HLO studies fall into a number of categories: (1) the selection of materials of general enough interest to warrant study under this program; (2) the accumulation of in-reactor properties data on these materials under a

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variety of flux and temperature conditions; (3) the measurement of pre- and postirradiation mechanical properties of materials of interest; and (4) the investigation of the basic nature of irradiation effects.

METALLIC FUELS DEVELOPMENT

NEW FUELS DEVELOPMENT

An experimental single tube N-Reactor fuel element having a fluted outer surface is being designed. The element will have eight flutes generated by tangential arcs in which the hill-to-valley arc radii ratio will be approximately 1/10.

Pressure testing of the Zircaloy-2 cladding of a fluted fuel element resulted in a 37 percent expansion of the volume before jacket failure.

Examination of a Zircaloy-2 clad uranium rod recently irradiated in a NaK capsule has shown a brittle failure in the 30 mil cladding without an associated split in the substrate uranium.

Fatigue tests are being conducted on the Zircaloy-2 "buggy spring" supports for the NPR inner tube. These supports have withstood 10^7 cycles with an amplitude of 0.006 inch without failure. A study is underway to determine coefficients of friction associated with charging aluminum clad fuel elements into ribbed or unribbed aluminum or Zircaloy process tubes. In the work to date, it has been found that addition of a soluble oil to the coolant substantially reduced the measured coefficient of friction whenever the fuel cladding and the process tubes were fabricated from the same metals. However, the use of soluble oil actually caused a substantial increase in the measured coefficient of friction whenever the clad and process tube were fabricated from different metals.

FABRICATION DEVELOPMENT

"Self-brazed" closures on Zircaloy clad fuel elements have been successfully fabricated by welding a partially copper plated Zircaloy ring into the recessed ends of the fuel component, placing the component within a steel die lined with insulation, and subjecting the fuel to several pulses of high amperage current using a resistance welder as the current source.

In studies directed toward projection welding an end cap to the projecting ID and OD clad of a recessed fuel tube, premature fusion of the ID projection has caused expulsion of molten metal from the ID weld. It is not yet clear whether an adequate control of current distribution can be obtained to permit simultaneous welding of the ID and OD projections.

The reference closure presently employed for N fuel fabrication requires a ring of 5 Be-95 Zircaloy-2.

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In the study of alternate closure methods, unalloyed beryllium is being pre-placed on the Zircaloy end cap by vapor deposition. An electron beam heating technique has permitted the placement of adherent beryllium films varying from a few angstroms in thickness up to 0.0005 inch.

Control of the metallurgical structure of uranium ingot or billet stock intended for coextrusion is being studied by beta heat treatment of the ingot or billet. The present process requires triple beta heat treatment of the cast ingot. An ingot containing additions of iron and silicon has been subjected to a fifteen hour alpha phase soak, followed by a single beta heat treatment. The ingot was then extruded to billet dimensions. Preliminary examination of the macrostructure of the resulting billet indicated that the structure is equivalent to, or superior to, that of material processed through the triple beta heat treatment step.

The extrusion constants of zirconium alloys which vary from the standard composition of Zircaloy-2 by reduction of tin content, and which contain two different levels of oxygen, have been measured at temperatures between 500 and 750 C. These extrusion constants have been compared with that of unalloyed production uranium. A match between extrusion constants of uranium and zirconium occurs only with unalloyed low oxygen zirconium and with extrusion temperatures of approximately 520 C.

In the study of material properties affecting the quality of coextruded zirconium alloy clad uranium fuel, uranium billets with widely different grain sizes have been coextruded together with unalloyed low oxygen zirconium, and with a hardened high oxygen zirconium alloy. The assembled billets were coextruded clad in copper, in steel, or bare. In all cases, the harder clad formed a smoother clad-uranium interface than the softer clad, and the bare and steel canned extrusions have a smoother clad-uranium interface than do those canned in copper.

Zircaloy-2 wires modified from the normal Zircaloy-2 compositions by the addition of 0.5 wt % of cobalt are being fabricated. At the present time, the material has been double vacuum arc melted, extruded to one-half-inch rod stock, and cold swaged to 0.130-inch rod stock. After annealing, the stock will be cold drawn to 0.075-inch diameter wire. During cleaning and autoclaving of material samples, it was found that the pickling rate of this alloy in HF-HNO₃ is approximately one-half that of Zircaloy-2. The samples formed a black adherent autoclave film after a 12-hour exposure to 400 C, 1500 psi steam.

FUEL EVALUATION

Irradiation of a full-sized NPR fuel element has continued at the ETR with the exposure of the outer tube now being 700 MWD/T. Dimensional measurements continue to show the fuel element warping in the plane of the flux gradient. Fuel element supports on the inner component have failed as a result of fatigue induced by vibrations in the test loop. An experimental single tube dual-enrichment fuel element has completed one cycle of irradiation at the ETR. Calculated maximum fuel temperature was 750 C.

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Density measurements on NPR inner fuel component continue to show relatively small amounts of swelling as a result of irradiation in pressurized water loops. A computer program is being developed to evaluate observed swelling in terms of an empirical equation.

CERAMIC FUELS DEVELOPMENT

CERAMIC (URANIUM) FUELS RESEARCH

Fission fragment migration studies with sintered UO_2 and with vibrationally compacted UO_2 - ThO_2 confirmed earlier data indicating extensive relocation of both fission fragments and plutonium during irradiation of ceramic fuels.

Irradiated, single crystal UO_2 was examined at elevated temperatures in the electron microscope, for the first time, using reflection microscopy techniques.

Postirradiation examination of a previously characterized single crystal of UO_2 revealed: (1) no change in the general appearance, (2) an increase in hardness and wide variations in fracture surface characteristics, (3) an increase in melting point, and (4) a decrease in the sublimation rate.

Several methods of simulating nuclear fuel heating were developed. Microstructures developed during sintering and during solidification of UO_2 cores were similar to those observed in many irradiated fuel elements. Molten UO_2 flowed readily through a 0.001-inch wide crack.

Swaged UO_2 irradiated to estimated exposures exceeding 20,000 MWD/T_U with core temperatures approaching the melting point developed unusually thin radial columnar grains in portions of the fuel.

Selected UO_2 single crystals were purified, machined, and thoroughly characterized for fundamental UO_2 property studies at Hanford and at other sites.

Techniques for quantitative determination of open porosity in fused UO_2 were developed. A water elutriation method was developed for separating porous, UO_2 chunks from fused or electrodeposited UO_2 .

A new Model 1220-B Dynapak machine was installed for evaluation of the high energy impact process for preparing ultra-high density ceramic fuel materials.

Initial postirradiation examination of a nonfree-standing-clad, vibrationally compacted UO_2 fuel element revealed no distortion of the

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0.008-inch 304-L stainless steel clad rods after an exposure to 1200 MWD/T_U in the VBWR.

Hot isostatic pressing of fuel elements that are partially predensified by swaging or vibrational compaction was successfully demonstrated using several types of UO₂. Fuel densities greater than 98 percent TD were achieved with micronized UO₂ swaged at Hanford and pressed by BMI engineers.

Tests revealed that severe hot spots in nonhomogeneous UO₂ based fuel mixtures were rapidly alleviated by rapid relocation and dispersion of the enriched component into the fuel rod center.

PLUTONIUM RECYCLE PROGRAM

Uranium Fuels Development

The PRTR Mark I swaged UO₂ elements revealed no mechanical deterioration, or corrosion after irradiation to approximately 2300 MWD/T_U. Other UO₂ test elements being irradiated include a vibrationally compacted Mark I cluster (~725 MWD/T_U), a hot swaged UO₂ Mark I cluster (~630 MWD/T_U) and a vibrationally compacted, Mark II-C nested tubular fuel element (~1100 MWD/T_U).

The feasibility of cathodic etching of fuel element components to clean them prior to completing remote welding of end closures was demonstrated.

Resonant beam energy transfer techniques are being developed for remote vibrational compaction of partially decontaminated recycled fuel. The technique was demonstrated by compacting fused UO₂ to >91 percent TD in PRTR Mark I fuel element rods. A completed Mark II-C (tubular) fuel element was mechanically tested in the remote vibration mock-up and shown to be amenable to preassembly and inspections before fuel loading.

High voltage electron beam techniques were adapted to attaching ribs to PRTR Mark II fuel element cladding.

Six PRTR Mark I UO₂ fuel elements are being assembled with combination flux monitor-spacer wires for physics tests in the PRTR and the PRCF.

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

The alloys to be studied under the program to determine the irradiation effects in reactor metals must represent both the alloys being used in existing reactors and those alloys suitable for use in future reactors. Efforts this past quarter have been directed toward the selection of alloys to be investigated under the new program. The alloys listed in Table 3.1 have been tentatively selected for initial study. These alloys were selected on the basis of an extensive literature survey, personal contact with manufacturers, and consideration of types and quantities of the alloys used in existing reactors.

➤ In addition to the alloys listed in Table 3.1, one high strength zirconium base alloy and one niobium base alloy will be selected for investigation.

An extensive effort will be made to insure that all material procured for investigation under the new program is representative of commercially obtainable heats. This is being accomplished by obtaining from steel manufacturers heat analyses and data of heat chemistry variation of each selected alloy for heats produced during the past 6 to 12 months. From this data, specifications will be written to insure procurement of a representative material. Alloy development is not to be a part of this program.

To insure a homogeneous lot of material, sufficient amounts of each alloy will be procured to supply the needs of the entire program. Personal contact will be made to witness all fabrication processes of each alloy to insure complete knowledge of the fabrication and heat treatment histories of the selected alloys. Sufficient chemical analyses will be made to determine the chemical variation of both the alloy heat and of the ingots poured.

A program of this magnitude will require the testing of many specimens. Orderly coordination of such investigations can only be achieved through a program based upon statistical design. The quantity of data which must be analyzed will also make it feasible to program the information into existing and new data processing programs. The Hanford Laboratories, Operations Research and Synthesis Operation has been requested to design a suitable statistical and data analysis program. The data derived from these programs can then be placed into the data retrieval system now being initiated in Hanford Laboratories.

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TABLE 3.1
ALLOYS RECOMMENDED FOR INITIAL INVESTIGATION OF IRRADIATION EFFECTS
ON REACTOR STRUCTURAL METALS

Alloy	Nominal Composition															
	C	Mn	P	S	Si	Ni	Cr	Mo	Ti	V	Cu	W	Co	Fe	Al	B
HY-80	0.18	0.10/ 0.40	0.025	0.025	0.15/ 0.35	2.0/ 3.25	1.00/ 1.80	0.20/ 0.60	0.02	0.3	0.25			Bal.		
A 302B	0.25	1.45/ 1.50	0.035	0.04	0.15/ 0.30			0.45/ 0.60						Bal.		
304 SS	0.08	2.0			1.0	8.0/ 11.0	18.0/ 20.0							Bal.		
348 SS	0.03	2			1	11	18/ 20							Bal.		0.3
406	0.07	0.40	0.014	0.012	0.48	0.12	13.5	0.03	0.32		0.05			Bal.	3.9	
AM 355	0.13	0.95			0.25	4.3	15.5	2.75	0.10					Bal.		
Inconel Alloy 600	0.15	1.0		0.05	0.50	72	14/ 17				0.50			6/ 10		
Inconel Alloy X-750	0.08	1.0		0.01	0.5	70	14/ 17		2.25/ 2.75		0.50		1.0	5/ 9	0.4/ 1.0	0.7/ 1.20
D 979	0.05	0.50			0.50	45.0	15.0	3.75	3.0			3.75		27.44	1.00	0.01
Hastelloy X-280	0.05/ 0.15	1.0			1.0	47	20/ 23	8/ 10				12/ 1.0		<0.5	17/ 20	
Zircaloy-2						0.03/ 0.08	0.05/ 0.15					0.07/ 0.20				Bal. 1.2/ 1.7

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

The objective of the in-reactor measurements study is to define the physical and mechanical behavior of structural materials during neutron irradiation. At the present time, a study of the in-reactor creep properties of Zircaloy-2 is being conducted. Data from a prototype in-reactor creep test capsule operated between 260 and 285 C indicated that creep is markedly increased in a fast neutron flux in this temperature range. A subsequent test conducted at 310 C, in a capsule with improved strain resolution, also displayed a marked increase in total creep strain on the in-reactor specimen, but most of the actual creep occurred during the reactor shutdowns rather than during the time the specimen was being irradiated. A similar test at 600 C displayed no difference in the creep rates between the in-reactor and out-of-reactor specimens.

In-Reactor Studies

Analysis of data taken during the first reactor outage of the creep test (Capsule III-3) conducted in-reactor on a 20 percent cold worked Zircaloy-2 specimen at 310 C and 30,000 psi⁽¹⁾ is now complete. The data show that no increase in creep rate occurs until about 2-1/2 hours after shutdown. Creep rates then increase for about 7-1/2 hours after which the rates decrease (Figure 3.1). During the past quarter, the creep test was

FIGURE 3.1

Creep Data from Capsule III-3 During the First Shutdown

continued through a third shutdown. The change in creep rate immediately after this shutdown was observed by taking strain readings at one-half hour intervals. Again, as in the first shutdown, a sigmoidal strain-time curve was observed; however, the strain occurring during this shutdown was only 1/3 of that which occurred during the first shutdown.

After completion of shutdown No. 3, the stress was removed from the specimen concluding the constant stress and temperature phase of the creep test planned for this capsule. The entire creep curve for the constant stress and temperature portion of the 310 C in-reactor creep test is presented in Figure 3.2. The creep test was discontinued as the total plastic creep strain was approaching 1 percent. One percent strain is a value at which third stage creep is imminent. Third stage creep was avoided so that the next phase of the experiment, activation energy measurements, would not be conducted on a specimen undergoing either necking or internal fracturing.

FIGURE 3.2

Entire Creep Curve for the In-Reactor Test Conducted
at 310 C and 30,000 psi

Activation energy measurements were conducted in the capsule at the conclusion of the creep test. Temperatures investigated were 300 to 450 C at stresses of 30,000 to 43,000 psi. The method used for activation energy measurements was identical to that used in the preirradiation study.⁽²⁾ The creep rates established just before and just after an abrupt temperature change from T_1 to T_2 are substituted into the equation,

$$R \ln \frac{\dot{\epsilon}_1}{\dot{\epsilon}_2} = \left(\frac{1}{T_1} - \frac{1}{T_2} \right) \Delta H,$$

where R = gas constant

ΔH = activation energy

$\dot{\epsilon}_1$ = creep rate at temperature T_1

$\dot{\epsilon}_2$ = creep rate just after an abrupt temperature change from T_1 to T_2 .

and the expression solved for ΔH .

The activation energies found at various temperatures are shown in Figure 3.3. The values in Figure 3.3 can be separated into two groups. The first group has an average value near 85,000 cal/mole and is observed only upon temperature increases. The second group has an average value near 52,000 cal/mole and is observed upon temperature decreases. Specimen failure at about 5 percent strain precluded the extension of the activation study to other temperatures.

The form of the in-reactor creep curve (Figure 3.2) can be rationalized in terms of neutron produced defects and annealing of the defects. Neutron irradiation produces defects which tend to increase resistance to plastic deformation. At irradiation temperatures below 380 C neutron irradiation inhibits tensile plastic deformation. Further, above 280 C, annealing of the deformation inhibiting defects can take place.⁽³⁾ The accumulation of irradiation defects at 310 C inhibiting the plastic deformation process of creep can then be considered to be the cause of the low in-reactor creep rate under reactor on conditions. Upon a reactor shutdown, these defects are no longer generated and those which exist from the previous irradiation slowly anneal out with time producing the increased creep rate observed a few hours after shutdown. The creep strain is greater during the shutdown than for normal primary creep of 20 percent cold worked material. This can only mean that, in addition to producing defects, irradiation alters the cold work substructure in such a manner that its creep resistance is reduced.

FIGURE 3. 3In-Reactor Activation Energies for Zircaloy-2

The difference in activation energies for temperature increases and decreases can also be rationalized in terms of defect production and annealing. Since irradiation damage in Zircaloy-2 can be annealed at temperatures greater than 280 C, a steady state concentration will be established when defect production rates are equivalent to defect annealing rates; hence, a particular concentration will be established for each temperature and flux intensity. The mechanism of Zircaloy-2 creep above 300 C has been established out-of-reactor as dislocation climb and is associated with an activation energy of 58,500 cal/mole. Since in-reactor the number of irradiation produced defects and the rate of dislocation climb depends on temperature, an activation energy greater than 58,500 cal/mole will be observed upon temperature increases. This assumes that significant defect annealing takes place during the temperature change. In contrast, if the temperature is

lowered, the defect level of the higher temperature is essentially frozen and only the activation energy for climb is observed. The fact that the activation energy for temperature decreases is only 52,000 cal/mole, rather than 58,500 cannot be explained at the present time. More data points from future tests are required to complete the spectrum of activation energies.

Capsule Development

The incorporation of water cooling in the creep capsule created the problem of maintaining specimen temperature during a reactor shutdown. The first capsule with water-cooling required the manual operation of water flow and dummy electrical loads to maintain constant temperature through reactor shutdowns. This is a function which must be automatic if capsules are to perform properly through unscheduled reactor outages.

A recent in-reactor capsule has been operated in the temperature range of 250 to 350 C through reactor cycling by controlling water flow. The capsule demonstrated the capabilities of water cooling as a mode of specimen temperature control. Water control is obtained by using two proportional controllers, one sensing the effluent water temperature and the second sensing the temperature of the creep specimen. The first controller is set to maintain an effluent water temperature of 80 C by operating a valve in the water line. The second controller, which controls both electrical power to the specimen heater and a second valve in parallel to the first, is set to the desired specimen temperature. During reactor shutdown, the first controller acts to hold the water at 80 C while the second controller adjusts to minimum water flow and maximum heater power to maintain a set specimen temperature. As the reactor is brought to its operating power level, gamma heat is generated in the capsule. The first controller continues to control for an 80 C water temperature. Due to nuclear induced heating in the capsule, the power to the heaters must be decreased and the cooling capacity increased. Both of these functions are performed simultaneously by the second controller. Essentially, the two controllers provide a temperature range available to the specimen even though they act independent of each other. The upper temperature limit is determined by the maximum power level of the heaters and the minimum water flow. During reactor shutdown, the first controller supplies the required amount of water while the second controller supplies none. The lower temperature limit is determined by the maximum water cooling and no power to the heaters. At minimum temperature, the first controller supplies no water while the second supplies full flow.

Presently, the electrical power requirements of the capsule during a reactor shutdown are such that the manual adjustment of dummy loads is still necessary. However, the replacement of the magnetic amplifiers with signal controlled rectifiers in the power control circuit will render the system fully automatic.

As previously mentioned, the range of temperatures obtainable in the present capsules with water cooling is limited to about 100 C. Mock-up studies have been started to determine the best cooling coil arrangements to increase the range of operating temperatures.

Irradiation and Environmental Effects - A. L. Bement and A. J. Lovell

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

With the conclusion of ETR Cycles 42 and 43, six quadrants containing both bend test and tensile specimens of Zircaloy-2 were discharged from the G-7, hot-water loop facility. The estimated neutron fluxes and exposures for these quadrants are given in Table 3.2. From the start of loop operations with Cycle 39, a total of 392 irradiated specimens of Zircaloy-2 and types 304 and 347 stainless steel have been discharged, representing exposures ranging from about 5×10^{19} to about 8×10^{20} nvt. Of these specimens, 288 have been shipped to HAPO for testing.

At the end of Cycle 44, a new sample holder and central flux monitor were installed in the loop. The new monitor consists of 1/8-inch lengths of iron, nickel, titanium, and Al-0.1% cobalt flux wires positioned at each of the seven quadrant levels. These wires are enclosed in an aluminum sheath, which is in turn contained in a 0.350-inch OD stainless steel capsule with a threaded end cap. The stainless steel capsule fits within the central bore of the specimen holder, and is easily removed and replaced. Upon discharge, the aluminum sheath is removed from the stainless steel capsule under water. The flux wires can then be removed from the aluminum sheath behind nominal lead shielding within the operations area of the Engineering Test Reactor (ETR).

The operating temperature and history for the G-7 loop for Cycles 42 and 43 are shown in Table 3.3. Here it is seen that better than 80 percent of the loop operation time is controlled at the set-point temperature of 540 F. Furthermore, the temperature records reveal that deviations from this set point during full reactor power are within ± 3 F.

Start-up of an out-of-reactor loop to duplicate environment of the ETR G-7 loop facility except for the neutron flux was achieved. This loop contains three test sections fabricated of 2.300-inch ID heavy-wall tubing. The water is pressurized by steam dome, and feed and bleed makeup is

TABLE 3.2QUADRANT DISCHARGES FOR ETR CYCLES 42 AND 43

Quadrant Number	Composition	Estimated Flux, nv	Estimated Exposure, nvt
25	18 Zircaloy-2 Tensile Specimens	1.3×10^{14}	7.8×10^{20}
26	18 Zircaloy-2 Tensile Specimens	1.3×10^{14}	7.8×10^{20}
45	12 Zircaloy-2 Bend Test Specimens	6.9×10^{13}	5.9×10^{19}
46	12 Zircaloy-2 Bend Test Specimens	6.9×10^{13}	5.9×10^{19}
47	12 Zircaloy-2 Bend Test Specimens	1.2×10^{14}	1.8×10^{20}
48	12 Zircaloy-2 Bend Test Specimens	1.2×10^{14}	1.8×10^{20}

Total specimens discharged, Cycles 42 and 43 = 84

Total specimens discharged to date = 392

TABLE 3.3TEMPERATURE AND OPERATING HISTORY OF ETR G-7 FACILITY

Cycle	42	43
1. <u>Reactor</u>		
Start-up ⁽¹⁾	11-25-61	1-12-62
End of cycle	1-2-62	2-4-62
Megawatt days	4270	2447
Effective days at 175 Mw	24.4	14.0
Number of scrams ⁽²⁾	21	6
Number of shutdowns	4	1
2. <u>G-7 Loop</u>		
Maximum temperature	543 F	543 F
Effective days above 200 F	23.1	14.3
Effective days at operating temperature	19.2	12.2
Operating efficiency ⁽³⁾	94.7%	102.0%
Temperature efficiency ⁽⁴⁾	83.2%	85.5%

(1) Includes flux run

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

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

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

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available, but optional. Additional facilities to adjust and maintain water quality and to record and control operation variables are a part of the loop. Two test sections accommodate 336 tensile specimens and 56 corrosion blanks, and the third test section accommodates 84 bend test and notch-tensile specimens.

Zircaloy-2 surveillance tensile specimens placed in the ETR G-7 loop are autoclaved as part of their preparation. To determine the extent of recovery due to autoclaving on the tensile properties of Zircaloy-2, two series of tensile specimens were tested. The first series was autoclaved at 425 C and the second was vacuum annealed at 360 C. The duration of the treatments was 72 hours. The average results from these tests are listed in Table 3. 4.

In general, the amount of recovery is greater for the rolling than for the transverse direction. This is particularly true for the 360 C treatment where practically no change is observed for the yield strength, uniform strain, necking strain, and plastic work in the transverse direction; whereas moderate changes in these properties occur for the rolling direction. After a 425 C treatment, the tensile strength recovers about 10,000 psi for either orientation at each level of cold work (10 to 70 percent). Significant recovery at this temperature also occurs in ductility parameters for the longitudinal direction.

Of singular interest is the change brought about by annealing in the plastic work required for fracture in tension. This quantity represents the area under the load-deflection diagram after correction for work expended in elastic deformation. Referring to Figure 3. 4, which is a plot of plastic work versus cold work, it is seen that the work expended in the rolling direction is nearly doubled after autoclaving at 425 C. This reflects the marked increase in total elongation for this direction resulting from cold work recovery. In contrast, the transverse direction exhibits little or no change in work as a result of either autoclaving treatment.

The strength and ductility of metals are affected, sometimes markedly, by the rate of load application. Since stress relaxation is pronounced in Zircaloy-2 at room temperature, it would be expected that the strain-rate sensitivity would be appreciable. In the present study, the crosshead speed of the Instron testing instrument was varied from 0. 005 to 5 in. /min, corresponding to strain rates of about 2.1×10^{-3} to 2.1 min^{-1} . In the course of these studies, two possible sources of error in the test data were investigated. The first was adiabatic heating, and the second was load-rate sensitivity of the Instron machine.

TABLE 3.4
EFFECTS OF 360 AND 425 C THERMAL TREATMENTS
ON TENSILE PROPERTIES OF ZIRCALOY-2
(AVERAGE OF THREE TESTS)

Cold Work	Condition		Strength, ksi		Percent Strain		Reduction in Area, %	Plastic Work, ft-lb
	Orientation	Treatment	0.2% Yield	Tensile	Uniform	Necking		
0	RD	As-Prepared	47.3	66.4	14.0	20.6	50.1	86.1
		360	47.4	66.5	14.2	14.5	48.0	71.3
		425	49.9	71.1	14.4	16.8	49.3	78.0
	TD	As-Prepared	54.4	63.5	11.2	27.3	52.2	81.1
		360	54.9	63.3	11.9	20.5	56.7	69.5
		425	54.8	63.1	10.8	19.9	56.4	73.0
10	RD	As-Prepared	73.8	82.2	2.6	8.8	47.2	33.8
		360	67.3	75.6	4.7	12.1	48.0	47.3
		425	60.4	72.5	7.4	13.4	48.2	56.2
	TD	As-Prepared	70.6	80.0	2.7	9.0	52.3	31.7
		360	71.4	76.3	2.6	8.0	56.8	29.0
		425	66.8	71.2	4.7	11.7	56.4	42.9
20	RD	As-Prepared	80.1	89.0	2.6	7.0	42.8	30.6
		360	74.1	82.8	4.4	9.5	43.4	42.8
		425	63.6	76.7	6.9	13.1	47.6	55.9
	TD	As-Prepared	77.4	92.8	2.7	7.2	53.2	31.0
		360	76.7	81.2	2.7	8.2	55.8	28.0
		425	71.4	76.0	3.5	7.6	56.9	25.3
40	RD	As-Prepared	86.1	96.3	3.0	5.7	39.4	29.2
		360	78.6	89.4	4.7	9.1	43.1	45.9
		425	72.7	89.1	6.7	9.7	39.4	55.3
	TD	As-Prepared	80.4	92.4	2.6	5.7	52.7	24.3
		360	81.3	86.6	1.9	6.7	55.2	23.8
		425	76.9	81.3	3.6	6.3	58.0	27.1
70	RD	As-Prepared	92.8	109.1	3.4	5.3	34.2	32.7
		360	87.1	102.2	5.1	6.6	38.0	43.0
		425	79.9	99.0	6.8	7.8	38.0	53.3
	TD	As-Prepared	86.7	106.0	3.9	4.1	40.7	26.1
		360	91.7	103.1	2.2	3.5	47.9	20.2
		425	93.3	97.2	1.9	5.5	49.6	25.3

FIGURE 3.4
Recovery in Plastic Work for Autoclaved Zircaloy-2

Because of anelasticity and internal friction, part of the work imparted during a tensile test is converted to thermal energy. In the absence of heat transfer from the specimen to the surroundings, the temperature increases, causing a significant change in the tensile properties. This can be observed by comparing tests conducted in still air with those conducted in water at the same ambient temperature. Strength values for the latter case, where heat is more rapidly dissipated from the specimen by conduction and convection, are usually a few psi higher. It is important to determine whether the maximum temperature rise varies significantly with strain rate; that is, whether the heat generation is significantly greater than heat dissipation in normally circulated air. Notch tensile specimens were employed for which plastic strain is confined between the notches, thus removing uncertainty in the placement of the thermocouple. A chromel-alumel couple constructed of 0.010-inch wire was spot welded to the specimen, and the voltage signal was fed to a Sanborn, oscillograph recorder. Two crosshead speeds were used; namely, 0.5 and 20 inches per minute. The corresponding maximum surface temperatures recorded were 45 and 69 C. It was concluded that the temperature rise for either strain rate was only moderate, and that errors resulting from adiabatic heating could be neglected in view of the normal scatter expected in the data. This may not be true in general, since in the present case the specimen was 0.050-inch thick and had a high surface-to-volume ratio.

Another important source of error at high strain rates is the limitation of the testing machine in tracking load application. Because of time lags in the recording system, major errors can be introduced, depending upon specimen gage length, modulus of elasticity, crosshead speed, and the percent of scale employed. The error is manifested in three ways. First, the modulus of elasticity is markedly reduced; secondly, the maximum load can be greatly underestimated by signal suppression with load reversal associated with necking; and thirdly, overshooting may occur with rapid tracking speeds exhibiting a pseudo-yield point. All of these errors were observed in testing 2-inch gage specimens at crosshead speeds exceeding 5 inches per minute. Three types of tests were conducted to determine the nature of these errors. These included zero-point suppression of the load signal, spring constant measurements of a helical spring, and modulus of elasticity measurements on iron wire of various lengths. These tests revealed that the Instron is capable of a higher rate sensitivity than the 0.5 second per 10 percent of chart specified by the manufacturer, and that crosshead speeds up to 5 inches per minute with the present specimen geometry give accurate results so long as a sufficiently high load scale is employed. The experience gained in these tests suggests that other data reported in the literature, where the Instron is employed at higher crosshead speeds with subsize specimens may be in error.

The results obtained from tensile tests of Zircaloy-2 conducted at various strain rates confirmed the mechanical equation of state, which predicts a linear plot of stress at constant strain as a function of log strain rate. The variation of the ultimate tensile stress with log strain rate is shown in Figure 3.5 for various amounts of cold work in the transverse direction. The slopes of these curves (calculated on the basis of crosshead speed) are plotted in Figures 3.6 and 3.7 for the rolling and transverse directions, respectively. In making these plots, the question arises whether the constant strain condition is obeyed in plotting tensile strength versus log strain rate. Figure 3.8, which plots the uniform strain (associated with the tensile strength) as a function of log strain rate, reveals that this condition is met both at room temperature and 300 C for the cold worked specimens, but is not met for the annealed specimens. The reduction in uniform strain with strain rate for the annealed specimens could account for the comparatively low value of the strain-rate sensitivity plotted in Figures 3.6 and 3.7.

Referring to Figures 3.6 and 3.7, it is observed that the rate sensitivity for the tensile strength is lower at 300 C for both the rolling and transverse directions than at room temperature. This behavior suggests strain aging at 300 C, which is consistent with other manifestations of aging at this temperature. It would appear from these plots that the transverse direction is more strongly affected by aging, but the curves require better definition; particularly, at the intermediate levels of cold work.

FIGURE 3. 5

Effect of Strain Rate on Tensile Strength of Zircaloy-2 Transverse Direction

FIGURE 3. 6

Strain-Rate Sensitivity Versus Cold Work for Zircaloy-2
(Rolling Direction)

FIGURE 3. 7

Strain-Rate Sensitivity Versus Cold Work for Zircaloy-2
(Transverse Direction)

FIGURE 3.8

Effect of Strain Rate on Uniform Elongation for Zircaloy-2

Damage Mechanisms in Iron - T. K. Bierlein, J. E. Irvin,
H. E. Kissinger, J. J. Laidler, and B. Mastel

Previous studies of pure iron at Hanford Laboratories have demonstrated the pronounced effect of solute atom content on the properties of neutron irradiated "pure metals". Vacuum-melted electrolytic iron containing approximately 500 ppm total impurities was used as specimen stock in a study recently completed. The specimens were machined from three ingots, which varied in interstitial impurity content. The substitutional impurities were nearly constant for the three ingots. The dose dependence of uniform strain was a function of the interstitial impurity content; the ingot of highest purity retained appreciable uniform ductility to exposures greater than 10^{19} nvt. However, as interstitial content increased, the tendency for radiation embrittlement increased. Damage recovery in iron, as indicated by electrical resistivity and microhardness changes was also found to be dependent on interstitial impurity content.

The purpose of the present program is to establish the nature of interactions between defects present prior to irradiation and defects introduced by irradiation, and to investigate the possibility of neutralizing the deleterious effect of interstitial impurity atoms by chemical stabilization.

Arrangements have been made to purchase from Battelle Memorial Institute (BMI) ten pounds of zone-refined iron of purity at least 99.99 percent (100 ppm total impurity). Of this, 20 ppm or less are interstitial elements. Delivery of the first one pound lots of this material will commence in May, 1962.

In addition, 100 pounds of 99.95 percent pure iron has been ordered to serve as starting material for electron-beam zone-refining. It is not expected that this material will be refined to the purity obtained by BMI, but it will suffice for preliminary experiments and for less sensitive measurements.

Iron foils, 0.003-inch thick, have been received. These foils, 99.992 percent pure with respect to metallics, will be subjected to further purification before examination by electron transmission microscopy.

Controlled levels of interstitial impurities will be added to zone-refined iron. The effect of these impurities on radiation damage will be studied. The effect of additions of a stabilizing agent and the resulting stability of the carbides and nitrides formed will also be studied as a function of irradiation dose. Irradiations will be conducted at temperatures between 20 and 100 C.

Carbon and nitrogen, in amounts varying from 10 to 200 ppm, will be added to zone-refined iron. These additions will be made in a glass vacuum system, already constructed, which can be evacuated to pressures

of less than 10^{-6} mm of mercury. Additions of carbon will be made by means of a controlled CO-CO₂ atmosphere, the amount of carbon added being a function of the composition of the gas mixture. Nitrogen will be added in an ammonia-hydrogen atmosphere. After addition of interstitials, the material will be annealed at high temperatures for homogenization, cold-worked, and then recrystallized to a common grain size. In this way, the effects of variable grain size will be eliminated.

Titanium has been selected for initial study of stabilizing agents, as it is a strong nitride- and carbide-former. The titanium will be introduced by electron-beam floating zone melting techniques.

Initial studies of radiation damage will be made on iron with the following compositions: (1) iron, zone refined; (2) iron plus 50 ppm carbon, (3) iron plus 50 ppm nitrogen; (4) iron plus 500 ppm titanium; (5) iron plus 50 ppm carbon and 50 ppm nitrogen; (6) iron plus 50 ppm carbon and 500 ppm titanium; (7) iron plus 50 ppm nitrogen and 500 ppm titanium; and (8) iron plus 50 ppm nitrogen, 50 ppm carbon, and 500 ppm titanium. Further adjustments of the relative compositions may be necessary as techniques evolve.

Experimental Tests

A number of specific tests will be conducted under this program. Tensile tests will be performed on all compositions utilizing Instron testing machines capable of numerous strain rates and load ranges. The strain-aging behavior of both irradiated and unirradiated iron will be the prime method for evaluating the state of damage and will be supplemented by the ordinary tensile test. Test data will be processed by the 7090 computer. A FORTRAN program has been written which will yield all the common tensile parameters as well as true-stress true-strain values. X-ray diffraction examination will be limited initially to lattice parameter and line broadening measurements. Fourier analyses of line shapes will also be made by means of a program for the 7090 computer. A Philips EM-200 double-condenser electron microscope will be employed in studies of irradiated thin foils by transmission techniques. Replica techniques will also be used. Release of stored energy from irradiated iron will be measured by a differential thermal analysis method. Measurements will be conducted on all compositions.

Kinetics of damage recovery will be characterized by microhardness, electrical resistivity, and lattice parameter measurements. The results will be combined with stored energy data and used to interpret the recovery processes occurring. Additional functional study areas being considered include: internal friction measurements, field ion microscopy, small-angle X-ray scattering studies, and growth of high-purity iron single crystals and iron whiskers for subsequent irradiation studies.

Preliminary experiments on introducing interstitial impurities in iron have begun. Ingot iron, the starting material, was partially decarburized by heating at 1000 C for 16 hours in wet hydrogen. A portion of this material was then "nitrogenized" at 450 C in an ammonia-hydrogen atmosphere calculated to introduce 100 ppm nitrogen. The entire lot of material was then cold-drawn and recrystallized. Tensile specimens were machined from the ingot iron, decarburized iron, and decarburized-nitrogenized iron rods. The specimens were irradiated to fast neutron exposures of 10^{17} , 5×10^{17} , 10^{18} , 2×10^{18} , and 5×10^{18} nvt. To date testing has been completed on the lowest exposure specimens only. Strain-aging effects were observed in all specimens and were greatest in the nitrogenized specimens. A loss of ductility occurs after an exposure of only 10^{17} nvt. The decrease is greatest for the nitrogenized specimens.

SWELLING OF IRRADIATED FISSIONABLE MATERIALS

The irradiation of uranium at elevated temperature frequently causes large increases in the fuel volume. This increase is thought to be due to the accumulation of fission product gases into pores and to the formation of voids and cracks in the fuel. The majority of the knowledge on uranium swelling applies to prototype fuel elements irradiated under conditions intended to simulate reactor operation. This information is highly important, but does not provide adequate insight into the basic mechanisms of the swelling phenomenon. Consequently, the present investigation is directed toward establishing an improved empirical and fundamental understanding of swelling in irradiated uranium. The influence of metallurgical variables such as structure and composition, and irradiation variables such as temperature, thermal cycling, thermal gradients, external restraint (including pressure), burnup, and rate of burnup, are being studied.

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

The implementation of the above studies has required the design, development and testing of suitable capsules to evaluate the effects of the various parameters on swelling. At the present time, four capsules are in general use. These capsules are: (a) a general swelling capsule in which small, bare uranium specimens are maintained at a constant temperature regardless of the operating condition of the reactor; (b) a capsule for irradiating metallographic uranium specimens at a constant temperature only when the reactor is operating; (c) a capsule for irradiating clad uranium rods at an elevated temperature (See Section 4 for additional information concerning this capsule); and (d) a capsule that does not contain thermocouples for irradiating uranium specimens at or near ambient reactor temperatures. Future plans involve the design, development and testing of a capsule containing bare uranium specimens that will be irradiated at a constant temperature at elevated pressures.

Up to the present time, a major effort has been devoted to the construction of constant temperature capsules. These capsules have behaved in-reactor according to the design specifications and all of the components have shown extremely high integrity. Five of these capsules have been successfully irradiated. Most of the desired postirradiation information on the specimens from three of these capsules is complete. Further details follow in subsequent sections. The other two capsules have been shipped to Radiometallurgy for recovery of the specimens. Four additional capsules are presently being irradiated. These capsules are connected in tandem two to each test hole. Three of these capsules are operating at constant temperatures of 625 C, 625 C, and 575 C, respectively. The temperatures are being maintained through all conditions of reactor operations, using the control and record instrumentation of the new instrument facility (CG 785). A momentary temperature drop of 10 to 15 C is experienced when the reactor undergoes a normal shutdown and about a 40 C drop when the reactor scrams. The temperature fully recovers with no overshoot in a matter of minutes. The fourth capsule is operating at about 400 C when the reactor is up, due to the resistance to heat flow built into the capsule, but cools to ambient when the reactor goes down. The heater in this capsule had been broken during capsule assembly and although it had been repaired and successfully tested in the laboratory, it did not possess sufficient integrity to withstand the reactor environment. Hence, the heater failed soon after the capsule was inserted into the reactor. The samples in this capsule will provide valuable information concerning the behavior of uranium in the temperature range (350 to 500 C) in which growth (dimensional instability) disappears but swelling (increase in fuel volume) begins. In this transition temperature range, an abnormal swelling has, on occasion, been observed that appears to be accounted for by grain boundary tearing rather than by the more classical nucleation and growth of fission gas pores.

An additional capsule has been assembled and successfully bench tested and another capsule is about 50 percent completed. It is anticipated that these two capsules will be irradiated in a tandem configuration in a single test hole. Assembly of additional capsules will be delayed until heaters are delivered, around the end of June, 1962. Meanwhile, metallographic capsules will be assembled and irradiated. These capsules will contain specimens that have had a surface very carefully precharacterized to study the change in grain structure as a function of high temperature irradiation. The source of specimens for the initial capsules will be high purity ingot uranium and uranium received from the United Kingdom containing several hundred parts per million of iron and aluminum additions.

Four unmonitored NaK filled capsules, GEH-14-334, 335, 336, and 337, each containing six uranium-to-uranium diffusion couples were discharged after receiving their goal exposures. Two of these were opened in Radiometallurgy and the specimens recovered. The remaining two capsules are being stored for future use. The next section, "Postirradiation Examination", discusses these specimens in more detail.

Specifications have been written and a call for bids has been issued for a data logging system to be used to assimilate the temperature and pressure data generated by the general swelling capsules during irradiation.

Control instrumentation previously used with general swelling capsules has been returned to the vendor for re-work and replacement of defective and malfunctioning components. This instrumentation will be used in conjunction with the data logging system with future general swelling capsules in order to release the CG 785 instrumentation currently being used.

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

The radiometallurgy examination of uranium specimens from three controlled temperature capsules is now complete. Specimen 4-2, which was irradiated at 615 C to a burnup of 0.28 at.% and then vacuum annealed at 650 C for 100 hours, was dropped during handling. The specimen broke into small fragments and could not be used for metallographic study or density measurements. The chemical burnup analysis of Specimen 4-3, also irradiated at 615 C to a nominal burnup of 0.28 at.%, was reported last quarter as being 1.66 at.%. This sample was rerun and the value obtained was 0.43 at.% which agrees somewhat better with the nominal burnup values and the analysis of the other sample in Capsule 4. The measured value is still believed to be somewhat high.

Preparations are being made to open Capsules 7 and 8 and recover the specimens. It is planned to measure the density of the samples, to complete metallographic analyses, and finally measure the burnup with chemical analysis.

It is generally believed that the size and number of fission gas pores in irradiated uranium control the over-all swelling that is observed. Very little is known concerning the distribution of pores and even less is known concerning the influence of various swelling parameters on this distribution. It is of the utmost importance, therefore, that pore-size distributions be examined in great detail to achieve a better understanding of the basic mechanisms involved. Two groups of data are currently being examined. The first group was obtained from specimens irradiated at an intermediate temperature in a high pressure loop and subjected to postirradiation annealing while the second group was obtained from specimens irradiated at elevated temperatures in controlled temperature capsules. The details of the treatment of these specimens, the observed microstructural and density changes, and the statistical treatments employed have been reported in previous quarterly reports. All the pore diameter measurements that have been made on electron micrographs have been processed through the computer and this great mass of information is currently being analyzed and interpreted.

The changes in the pore volume fraction distribution observed in the high burnup samples (0.41 at.%) subjected to postirradiation annealing are much more pronounced than were the changes produced in the lower burnup samples (0.29 at.%). An example of this can be seen in Figure 3.9 which displays the as-irradiated condition and an annealed condition (600 C for 1 hour) for the two burnup levels. It is interesting to note that, although the 0.29 at.% burnup uranium exhibited more swelling in the as-irradiated state (probably due to a slightly higher irradiation temperature, lower external restraint, and/or lower burnup rate), the 0.41 at.% burnup uranium exhibits much more swelling after annealing at 600 C. The changes that occur in the pore volume fraction distribution during vacuum annealing of 0.41 at.% burnup uranium specimens are shown more completely in Figure 3.10. Also tabulated in this figure are the swelling values (total volume fraction), obtained from density measurements and both the swelling values and pore density values (per μ^3) obtained from the statistical analyses. As can readily be seen, the total volume fraction obtained by density measurements and statistical analyses agree quite well up to about 600 C and then the statistics leads to values that are much too low. This is due to the fact that there is a large contribution by pores that are too big to be included in the size range studied. No satisfactory method has yet been developed to handle these large pores. The possibility of using lower magnification electron micrographs (15,000X is presently being used) to permit the large pores to be included is being considered. Attention is also called to the fact that the number of pores per unit specimen volume first increases over the as-irradiated value and then decreases rapidly as the annealing temperature is increased. The initial increase can probably be attributed to the growth of pores that were too small to be measured in the as-irradiated state rather than to the nucleation and growth of new pores. The decrease at the higher temperatures is believed to be real and unequivocal. The mechanism at higher temperature may be an agglomeration process where several small pores come together forming a larger one or it may be a solution-reprecipitation process where small pores dissolve in the matrix and the gas migrates to growing large pores. A concerted effort is being made to determine which process is predominating.

Interpretation of the computer data from the second group of specimens (irradiated at elevated temperatures in controlled temperature capsules) is still somewhat preliminary but several facts appear particularly significant. It was mentioned last quarter, ⁽¹⁾ that Specimen 4-4, irradiated at 575 C to a burnup of 0.28 at.% and annealed at 575 C for 100 hours after irradiation, contained a large number of small pores and hence the pores on micrographs at both 15,000X and 35,000X, were measured. Figures 3.11 and 3.12 illustrate the pore volume fraction distributions obtained for the specimen in both the as-irradiated state and the annealed state. The advantage of the high magnification micrographs is quite obvious as a peak in the volume fraction distribution is now observed that was not evident from measurements made

FIGURE 3.9

Influence of Postirradiation Annealing and Burnup Level
on the Pore Volume Fraction in Irradiated Uranium

FIGURE 3.10

Influence of Postirradiation Annealing
on the Pore Volume Fraction in Uranium Irradiated to 0.4 at. % Burnup

FIGURE 3.11

Influence of Annealing Uranium
at the Same Temperature at Which It Was Irradiated
(35,000X Micrographs)

FIGURE 3.12

Influence of Annealing Uranium
at the Same Temperature at Which It Was Irradiated
(15,000X Micrographs)

at the lower magnification. Table 3.5 lists the contribution of pores to the over-all pore volume fractions and pore densities portrayed by the curves shown in Figures 3.11 and 3.12, and includes composite values derived from high magnification and medium magnification micrographs.

TABLE 3.5

SWELLING IN IRRADIATED URANIUM BEFORE AND AFTER ANNEALING

	<u>As-Irradiated</u>		<u>After Annealing</u>	
	<u>Pore Volume, Pore Density,</u> <u>Fraction</u>	<u>No. /μ^3</u>	<u>Pore Volume, Pore Density,</u> <u>Fraction</u>	<u>No. /μ^3</u>
High Magnification (Figure 3.11)	0.029	1.36×10^2	0.018	1.36×10^2
Medium Magnification (Figure 3.12)	0.029	0.11×10^2	0.029	0.14×10^2
Composite	0.051	1.38×10^2	0.039	1.46×10^2

The similarity in the distribution curves before and after annealing, and the similarity in the pore volume and pore density estimates listed in Table 3.5 leads to the conclusion that annealing of this specimen at its irradiation temperature has not caused any marked change in its state of swelling. Similar analyses of pore size distribution information on specimens subjected to different burnups, different irradiation temperatures, and different postirradiation annealing treatments are being made.

An approach to studying mobility of fission gases in uranium currently being pursued involves use of irradiated uranium (enriched)-to-uranium (depleted) diffusion couples. Such specimens irradiated at low temperatures, <300 C, have been subjected to postirradiation anneals to develop measurable pores near the uranium-to-uranium interface. Such a specimen, GEH-14-281, showed no porosity in the as-irradiated state and also none after vacuum annealing at 400 and 500 C for one hour. The electron micrographs of Figure 3.13 show that porosity does develop in the enriched uranium shell (irradiated to a nominal burnup of 0.3 at. %) after annealing at 650 C for 100 hours. Pores ranging from 0.05 to 0.5 μ in diameter are located primarily at the grain boundaries of newly formed, recrystallized grains. All of the enriched layer has not recrystallized and hence, pores are distributed nonuniformly. (These observations are in agreement with those made on specimen GEH-14-36, which is discussed in the last quarterly report.) The depleted uranium (irradiated to a nominal burnup of 0.015 percent) shows no porosity. Because pores are distributed nonuniformly in the

FIGURE 3.13

Formation of Fission Gas Pores in the Enriched U^{235} Portion of Enriched-Depleted Irradiated Uranium-to-Uranium Diffusion Couple GEH-14-281 After a 100 Hour Anneal at 650 C.
Pores are Located Primarily at Boundaries of Grains Which Have Recrystallized.

enriched shell, it is impossible to determine whether or not the number and/or size of pores change as the uranium-to-uranium interface is approached. There is no indication on this sample that diffusion of fission products into the depleted uranium core has occurred. This annealing of diffusion couples will be continued and particular attention will be placed on sweeping of gases by moving grain boundaries.

Four other capsules, GEH-14-334, 335, 336, and 337, each containing six uranium-to-uranium diffusion specimens similar to the one, GEH-14-281 already examined, have been received in Radiometallurgy. Two of these capsules, GEH-14-334 and 336, have been opened and the specimens successfully recovered. The other two capsules, GEH-14-335, and 337, are being stored until the samples are needed for examination. The irradiation history of the capsules and the calculated burnup in the 0.030-inch thick, 3 percent enriched shell and in the 1/2-inch diameter, 0.15 percent depleted core are summarized in Table 3.6.

TABLE 3. 6

IRRADIATION HISTORY OF CAPSULES GEH-14-334, 335, 336 AND 337

<u>Capsule Number</u>	<u>Cycles*</u>	<u>Exposure, nvt</u>	<u>at. % Burnup</u>	
			<u>Shell</u>	<u>Core</u>
334	2	1.31×10^{20}	0.234	0.011
335	2	1.31×10^{20}	0.234	0.011
336	4	2.47×10^{20}	0.442	0.022
337	4	2.42×10^{20}	0.431	0.021

* Irradiated in the Materials Testing Reactor (MTR) starting with Cycle No. 165 in a flux of 5×10^{13} nv.

The burnup values indicated in Table 3.6 represent averages for the entire capsule estimated from reactor operations data. Flux gradients will cause some variation in exposure between specimens. Sampling techniques have been developed in Radiometallurgy that will permit the enriched shell to be analyzed for burnup to determine the extent of the exposure gradients along the length of the capsules.

The samples, in general, were in excellent condition, exhibiting the changes that were fully anticipated. The appearance of part of the low and high exposure samples is illustrated in Figure 3.14. The shell enriched to

GEH-14-334 (1.31×10^{20} nvt) GEH-14-336 (2.47×10^{20} nvt)FIGURE 3.14

Macro Appearance of Zircaloy-2 Clad Uranium-to-Uranium Diffusion Couples GEH-14-334 and 336 After Irradiation 2X
The cylindrical shell which is enriched in U^{235} appears dark because the surface has become roughened due to low temperature irradiation growth. Growth of the enriched layer in a longitudinal direction is greater in the lower set of three specimens which received twice the exposure of the upper set.

3 percent U^{235} can be clearly seen as a black ring just inside the Zircaloy-2 cladding. This ring appears dark because of the bumping and growth that occurred in the enriched shell due to the low temperature of irradiation (<300 C). This growth can be seen in the side view of both sets of specimens in Figure 3. 14. Note that the growth of the enriched layer was appreciably greater in the high burnup specimens than in the low burnup ones. The amount of growth observed with these Zircaloy-2 clad specimens is considerably less than the amount observed previously with the copper clad (unbonded) specimens GEH-14-281 and 282. This is thought to be due to the restraining effect of the cladding on the growth phenomena.

A sample from each capsule is being prepared for metallography and an additional sample is being vacuum annealed at 650 C for 100 hours. Other samples will be annealed at 700 and 800 C, respectively, in an effort to produce porosity in the depleted uranium by virtue of fission gas diffusion.

Related Investigations - T. K. Bierlein, B. S. Kosut, R. D. Leggett, and B. Mastel

There is speculation that a finely dispersed second phase may be effective in increasing the number of fission gas pores formed during irradiation. Hopefully, these pores would be quite small, retain the fission gas at higher pressure because of the higher surface tension forces of small pores and, hence, result in less total swelling. It is of interest to the swelling program, therefore, to examine finely dispersed second phases as to their size, distribution and stability as a function of composition and thermal treatment. If desirable distributions are observed, selected specimens will be irradiated at elevated temperatures.

Samples from three uranium alloys (varying primarily in the iron and silicon content) are being examined in each of ten heat treated conditions. This includes as-received samples; samples that were quenched from the high alpha, the beta, and the gamma fields; and quenched samples that were given a precipitation treatment at low and medium alpha temperatures, respectively. The presence, morphology and distribution of second phase precipitates will be established from electron micrographs and statistical analysis. The spectrographic and chemical analyses of the three uranium alloys employed in this study are indicated in Table 3. 7.

Fourteen of the thirty specimens have been polished and etched and examined by light microscopy. The remaining samples are in process. No precipitate could be positively identified with the light microscope. Efforts to produce high quality positive replicas from cathodic vacuum etched surfaces have not yet been successful. If positive replication proves unsatisfactory, negative replicas will be employed.

TABLE 3. 7
ANALYSES OF THREE DINGOT URANIUM ALLOYS
USED FOR SECOND PHASE STUDIES

<u>Element</u>	<u>High Purity</u> <u>Dingot, A</u>	<u>Dingot Alloy, B</u>		<u>Dingot Alloy, C</u>	
	<u>Hanford*</u>	<u>Hanford</u>	<u>MCW**</u>	<u>Hanford</u>	<u>MCW</u>
Iron	45***	107	86	185	150
Silicon	20	52	69	93	100
Aluminum	~50	~20	40	~35	13
Chromium	~5	~50	--	~2	--
Copper	~20	~10	3	~20	2
Nickel	~5	~1	10	~10	22
Carbon	18	9	--	17	---
Nitrogen	12	11	12	11	34
Hydrogen	2. 5	2. 4	--	2. 8	4. 4

* Average of analyses from the top and bottom of a core blank

** Analyses supplied by Mallinckrodt Chemical Works (MCW) for the original dingot from which the core blanks were cut.

*** All values in parts per million

NEUTRON DAMAGE TO METALS

The objective of this program is to establish the extent of change in the properties of a given metal subjected to neutron bombardment, and to determine whether impurity atoms present in the metal alter the damage state. This information together with damage recovery information will be used to deduce a model describing the damage state of the metal.

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

High purity molybdenum is a suitable metal for initial study because a large proportion of the neutron induced damage is retained at room temperature, and because transmutations and residual radioactivity are slight. The impurity selected for investigation is carbon.

A capsule containing four single crystal bend test specimens is being irradiated to 10^{19} nvt (fast). One crystal containing 10 to 30 ppm C, has polished perpendicular faces of the form $\{110\}$ and $\{100\}$; the second, also containing 10 to 30 ppm C, has faces of the form $\{110\}$; and the third and fourth, containing 400 to 500 ppm C, have polished perpendicular faces of the form $\{110\}$. This capsule is a precursor of similar capsules to be irradiated to other exposure levels.

Single crystal rods of molybdenum, representing three controlled carbon contents, were cut into 1-1/2-inch lengths for tensile specimens. Each length was ground to a cylinder, heavily electropolished, and inspected for grain boundaries and other defects. A 3/8-inch long reduced section was ground on acceptable crystals. The gage lengths were again electropolished to a smooth mirror finish. A total of 112 specimens have been prepared in this way and consist of 37 low-carbon (10 to 20 ppm), 42 medium carbon (100 to 200 ppm) and 33 high-carbon (400 to 500 ppm) samples.

The crystallographic direction of the axis of each rod was determined from back-reflection Laue photographs. The ground and polished rods were mounted in the Laue fixture with the rod axis vertical and parallel to the film. In the resulting diffraction photograph, all spots lying on a horizontal line through the center of the film are a result of reflections from planes lying parallel to the rod axis. By indexing the Laue spots on the equatorial line, the orientation of the rod axis with respect to the crystal was determined.

The resulting orientations, as shown in Figure 3.15, are plotted on the unit stereographic triangle. The numbered areas in the figure indicate deformation modes predicted by Maddin.⁽⁴⁾ The total number of crystals in each deformation mode are tabulated in Table 3.8. This information will be used to determine the distribution of individual samples among the four proposed irradiation levels, 0, 10^{18} , 10^{19} , and 10^{20} nvt, to yield the maximum amount of useful data.

TABLE 3.8
NUMBER OF SINGLE-CRYSTAL TENSILE SPECIMENS
WITH ORIENTATIONS FALLING IN THE FOUR PREDICTED
DEFORMATION MODES

<u>Carbon Level</u>	<u>Deformation Mode</u>				<u>Total</u>
	<u>1</u>	<u>2</u>	<u>3</u>	<u>4</u>	
Low	13	2	8	14	37
Medium	25	3	13	1	42
High	28	2	3	0	33

(a) High Carbon 400 to 500 ppm

(b) Medium Carbon 100 to 200 ppm

FIGURE 3. 15

Orientations of Tensile Axes
of Single Crystal Specimens of Molybdenum Containing Carbon.
Numbered Zones Indicate Predicted Deformation Modes.

(c) Low-Carbon 10 to 30 ppm

FIGURE 3.15 (contd)

Single-crystal X-ray diffraction specimens having ground and polished (110) and (100) faces are being prepared. Two specimens from each carbon level will be irradiated to each of the four irradiation levels. It is anticipated that six capsules, each containing 16 single crystal tensile specimens and three single crystal X-ray specimens per capsule, will be used. Prior to encapsulation of the tensile and X-ray specimens, all tensile specimen lengths will be measured precisely.

A series of capsules have been submitted for irradiation in a low neutron flux environment to exposures of 10^{18} , 10^{19} , and 10^{20} nvt (fast). These capsules contain polycrystalline tensile, microhardness, resistivity, stored energy, and length change specimens. Capsules to be irradiated in a high neutron flux environment are being assembled. Six capsules contain four polycrystalline tensile specimens per capsule; three capsules contain two stored energy and two length change specimens per capsule; and three capsules contain nine polycrystalline resistivity specimens per capsule.

One hundred forty-four annealed polycrystalline foil specimens, 0.003-inch thick, representing four carbon impurity levels, were sealed in a total of 24 capsules. These capsules have been submitted for irradiation to various exposure levels.

As-rolled 0.003 and 0.005-inch-thick foil specimens, stock for optical hot stage deformation studies and electron microscope studies, have been subjected to a 1/2 hour annealing treatment at 1900 C to achieve grain growth and carbon homogenization. Equiaxed grain growth was observed, and carbides in the medium and high carbon stock (100 to 200 ppm C and 400 to 500 ppm C, respectively) became redistributed on a fine scale and decreased in size. An example of this is given in Figure 3.16. Electron microscope studies of these annealed foils after electrolytic thinning showed many of the carbide particles to be present on dislocation lines and at boundaries. Examples of the interaction between carbides and dislocations are given in Figure 3.17. Carbide particles in many instances showed a striated structure which was insensitive to tilting of the specimen foil. The striations may arise from crystallographic solution of the particles into the matrix during the annealing or from selective crystallographic etching of the carbides during the electrolytic thinning operation. As shown in Figure 3.17, only an occasional prismatic loop is present.

Swaging of polycrystalline molybdenum rod has been investigated for the purpose of (1) introducing sufficient cold-work damage for study of cold-work recovery by stored energy release measurements, and (2) obtaining an increased dislocation density for study of irradiation effects on cold-worked material. The stock used for this study was obtained in two conditions: (1) 1/8-inch diameter rod, hot-swaged; (2) 1/2-inch diameter rod, swaged and then annealed. Since equipment characteristics dictate the use of specimens nearly 7/16-inch in diameter, the 1/2-inch rod is required for starting material. Although the 1/8-inch diameter material was easily reduced by swaging to reductions as great as 25 percent, cold swaging of the 1/2-inch rod was impossible. Attempts to obtain a wrought structure by initial swaging at 600 C followed by cold-swaging have been unsuccessful. It is concluded that a unique annealing texture present in the 1/2-inch rod is responsible for the observed resistance to deformation. The observed brittleness cannot be attributed to grain-boundary precipitation effects, as the microstructure of the 1/2-inch material is similar to that of the 1/8-inch material after annealing. The chemical compositions of the two materials are nearly identical. Additional experiments are underway to ascertain optimum conditions for primary breakdown of the annealing texture.

Recovery of cold-work damage in molybdenum has been characterized by means of microhardness measurements. An isochronal anneal was conducted for one-hour periods and for temperature increments of 25 C over the range of 25 to 1200 C. Recovery occurred in a single stage, centered at 1025 C. An isothermal anneal at 1000 C was also conducted. The two respective annealing

FIGURE 3.16
Micrographs of Molybdenum Foils Containing 400 to 500 ppm Carbon in (a) As-Rolled, As-Chemically-Etched State, (b) As-Annealed, As-Thermally Etched State, and (c) Repolished, As-Chemically-Etched State Showing Change in Carbide Precipitate and Development of Carbide Network at Subgrain Boundaries
250X

75, 000X

(400 to 500 ppm Carbon)

60, 000X

(100 to 200 ppm Carbon)

FIGURE 3. 17

Transmission Electron Micrographs of Molybdenum Containing Two Levels of Carbon as Impurity. After an Annealing Treatment at 1900 C, the Foils Were Thinned Electrolytically. Note Interaction Between Carbides and Dislocation Segments.

curves, given in Figure 3.18, were used to calculate the recovery kinetics. The recovery stage was determined to consist of two separate processes, one occurring with an activation energy of 2.2 ev and the other about 4 ev. The latter process is most certainly recrystallization. Reaction orders were not determined, but the calculation has been programmed for the 7090 computer and will be made shortly. The computer program has been written to accommodate future recovery studies.

SUPPORTING STUDIES

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

The mechanical properties of Zircaloy-2 clad metal fuel elements are being investigated. In support of these studies notched tensile samples taken from Zircaloy-2 rolled sheet are being tested in the irradiated and unirradiated condition. The influence of notches on the mechanical properties of unirradiated control samples was reported previously. A knowledge of the combined effects of notch strengthening and radiation damage on mechanical properties will be obtained from notched material scheduled for an exposure of approximately 1×10^{20} nvt (fast) in the Engineering Test Reactor (ETR). Un-notched tensile specimens formed from coextruded Zircaloy-2 cladding are also included for study. The irradiation vehicle is tubular in shape, open at both ends to allow the reactor coolant to flow around the specimens. This permits an irradiation temperature which is essentially bulk water temperature. Calculations indicate a film ΔT of about 1 F and a specimen surface temperature of about 121 F. The specimens, 30 per capsule, are positioned in a slotted holder with their width dimension extending in a rosette pattern in the capsule annulus between the external body and the central tube. A spacer permits postirradiation capsule cutting without danger to specimens. Welded end straps prohibit longitudinal motion of the specimen holder. Figure 3.19 illustrates the capsule components and assembly. Two capsules, irradiation number GEH-14-354 and 355, will be charged in the ETR Cycle 46.

FIGURE 3.18
Cold-Work Recovery in High-Purity Molybdenum

FIGURE 3.19

Assembly and Exploded View of Tensile Specimen Irradiation Capsule

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4. 1

METALLIC FUELS DEVELOPMENT

NEW FUELS DEVELOPMENT

Shaped Single Tube Element - K. R. Merckx and D. P. O'Keefe

Design was started on an experimental N-reactor element in the form of a single tube with an outer surface shaped to accommodate uranium swelling without stressing the cladding in tension. The design concept of the fluted outer surface for fuel elements has been described in previous reports of this series. The principle will be incorporated in the design of a single-piece element that will meet operating requirements for the N-reactor. The preliminary design has been established on the basis of a 5 to 10 percent expansion capability. The element will have eight flutes generated by tangential arcs. The hill-to-valley arc radii ratio will be about 1:10. The possible cladding deformation that could occur adjacent to the brazed end caps may be a limiting factor. Hence, an end cap with a tapered transition between the uranium and the zirconium cap will be employed to lessen the displacement discontinuity between the uranium and the cap.

Mechanical models are being developed which describe the conditions in the cladding during various stages of the deformation. Elastic studies of a cladding deformed by a fuel material exerting a uniform pressure⁽¹⁾ have been previously described. These analyses were made to estimate the initial deformations of the cladding. Since shaped fuel elements would be used to extend the irradiation life by allowing for sizeable volume increases, the cladding would deform plastically. The limiting deformation that would be possible without extending the cladding surface is obtained when the corrugated surface becomes a circular cylinder. If the initial corrugations are formed by tangential circular arcs, a criteria of equal bending strains upon reaching the limiting circular geometry can be used to establish a ratio of arc radii. Calculations based on this criteria for cladding of uniform thickness have been completed for 6, 8, and 10 corrugations for various ratios of corrugation arc-length to base circle arc length (base circle is the circle tangent to the valley of the corrugations). These arc radii ratios are plotted in Figure 4. 1. These hill-to-valley radii ratios are larger than those predicted for minimum elastic stress conditions.

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4. 2

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FIGURE 4. 1

Ratio of Tangential Arc Radii
Based on Equal Limiting Bending Strains

Corrugated Fuel Element - R. G. Wheeler

The employment of a fluted or corrugated fuel element jacket shape to provide for core swelling during irradiation has been described previously. Since test fuel elements of this shape are scheduled for irradiation, a bench test was made to determine how much the volume contained in a corrugated jacket could increase before the jacket failed.

The solid line in Figure 4. 2 represents one-sixth of the cross-section through a corrugated fuel element. The coextruded core was removed chemically from the Zircaloy-2 jackets and Zircaloy-2 end caps were brazed into each end. Hydraulic fluid was pumped into the space formerly occupied by

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fuel and the shape of the jacket was measured as a function of both percent increase in volume and hydraulic pressure. The results are represented by the broken lines in Figure 4.2. The jacket finally failed at a junction with the end cap at a volume increase of 37 percent and an internal pressure of 650 psi.

FIGURE 4.2

Sketch Represents One-Sixth of a Section Through a Container Made from the Cladding of a Coextruded Uranium Cored Fuel Element. Broken Lines Represent the Change in Shape Caused by Application of Internal Pressure.

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Capsule Irradiations - J. W. Weber

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

Cladding Performance

Thinning or "necking" and fracture of Zircaloy-2 cladding have occurred on a rod from each of two 7-rod clusters and on the outer cladding of the tubular fuel element irradiated in pressurized water, and on several rods irradiated under a variety of conditions in NaK filled capsules. The characteristics of the failures are similar and have been described in previous quarterlies of this series. Examination of one uranium rod coextrusion clad with 0.030-inch Zircaloy-2 and recently irradiated in the MTR shows a defect on the cladding outer surface (Figure 4.3), that appears to be the same as the defects found on the above rods and tube. However, a transverse section through the area of the defect shows it to be a split in the cladding apparently resulting from bending. Figure 4.4 is the transverse cross-section with the surface defect indicated by the arrow. There is no uranium crack associated with the split shown in Figure 4.4 in the higher magnification picture, Figure 4.5.

Cladding Failure

FIGURE 4.3

Failure in Zircaloy Cladding is Shown Below the Orientation Line
3X

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Cladding
Failure

FIGURE 4.4

Transverse Cross Section Through the Failure Shown in Figure 4.3. Several cracks in the cladding are indicated, some associated with cracks in the uranium core. The failure shown by the arrow has no uranium cracks near it. 3X

FIGURE 4.5

Cladding Failure as Indicated by the Arrow in Figure 4.4
The uranium shows no indications of cracking associated with the
50X

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It is concluded that the failure started at the fuel cladding interface and propagated outward to the surface. Figures 4. 6 shows that little plastic deformation has taken place at the failure except possibly where it terminates at the surface, suggesting that the Zircaloy-2 was brittle under the fracture conditions. Cracks in the cladding at other locations also show brittle characteristics. Brittle failures have been found in other rods with 0. 030-inch thick cladding irradiated in NaK capsules but it is not clear if the failures started at the inner or outer surface of the cladding. The damage to the fuel rod on the opposite side from the failure resulted from a large split in the cladding and oxidation of the uranium after the capsule was opened.

To study further the effects of cladding thickness variations on the susceptibility to failure, a program of irradiating Zircaloy-2 clad uranium rods in NaK capsules with intentional striations in the cladding is being conducted. Four capsules were discharged the middle of March with 1500 MWD/T exposure. The temperature of the cladding surface in three of the capsules was 360 C and 420 C in the fourth capsule. It is planned to begin examination of the fuel rods by early May. Another series of capsule irradiations is being prepared in which the intentional striations are coextruded on the inside of the cladding, rather than being machined on the outside. In addition to investigating the effect of thickness variations, consideration will be given in this series of irradiations to the influence of striation geometry and multiple striations. The fuel rods have been coextruded and are now being heat treated. Machining of the capsules is complete and welding of the capsule components has been started.

Fuel Performance

To investigate the relative swelling of uniformly enriched and "dual" enriched uranium fuel elements operating at equivalent power generation and surface temperatures three capsule type experiments are being prepared for irradiation in the MTR.

The cross-section of the dual enriched rods is divided into a 0. 475-inch diameter core of depleted uranium of 0. 15 percent enrichment and a shell of 3. 006 percent enriched uranium with an OD of 0. 530-inch. Average enrichment of the sample is 0. 72 percent. The uniformly enriched rod has a constant 0. 72 percent enrichment over the cross-section. Both types of rods were coextruded with 0. 025-inch Zircaloy-2 cladding to a final OD of 0. 580-inch. Each capsule will contain one swelling comparison sample and one temperature measurement sample of each type of rod. All capsule and fuel components have been machined. Measurement and assembly of the fuel samples and initial assembly of the capsules has started. The neutron flux distribution in both types of rods has been calculated using the P-3 physics program. A computer program was used to determine the temperature distribution in the samples.

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FIGURE 4.6

Failure in Zircaloy-2 Cladding

Little plastic deformation has occurred at the failure except near the
outer surface.

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Thermal Expansion in NPR Fuel Elements - L. E. Kuhlken

The gross coefficient of thermal expansion for NPR fuel elements was obtained on an apparatus which measured the change in length of these elements in a constant temperature oil bath up to 100C. The length of fuel element was registered on a dial gage which read to 0.1 mil. Due to temperature changes in the oil and irregular surfaces on the ends of the elements, the readings were only reproducible to 0.5 mils. This amounted to a 2 percent error in the 20 to 25-mil length changes observed.

Measurements were made on 54 inners and 59 outers of a group of loop test elements. The average coefficient of thermal expansion was 13.055×10^{-6} in./in./°C and 12.723×10^{-6} in./in./°C for the inner and outer elements, respectively. These measured values are below the calculated coefficient of 16.0×10^{-6} in./in./°C based on theoretical considerations of cladding restraint.

Figures 4.7 and 4.8 show the thermal coefficient of expansion values plotted against the position in the coextruded fuel tube from which these elements were cut. The low thermal expansion of the beta-heat treated elements at the center of the tube can be explained by the accompanying increase in clad restraint caused by thickening of the clad at this position as also shown in Figure 4.7 by the dashed line. Figure 4.9 is the variation of thermal expansion in an as-extruded inner fuel tube showing that the effect of uranium texture overrides the effect of clad thickness variation.

Dimensional Measurement of Tubular Fuel Element - K. R. Merckx

Previous documents of this series and a report⁽²⁾ have described a computer program associated with the analysis of punched tape measurement data from tubular fuel elements. Such experimentally obtained data will contain random errors (noise). The analysis was so formulated that a noise analysis could be run when sufficient data had been collected. This noise analysis was programmed and run. The analysis indicated that the noise level was below 1 mil, and that approximately eight Fourier coefficients are larger than the noise level.

Fatigue Strength of Supports - R. G. Wheeler

The fuel centering supports on the inner fuel tube of NPR fuel assemblies may be compressed 0.020 to 0.030 inches, elastically. An exciting force can cause the fuel mass resting on its spring supports to vibrate in one or more characteristic modes. An investigation of the fatigue properties of the inner-fuel tube supports was started because of the possibility of this cyclic stressing in service.

A stress analysis* of the support detailed in Figure 4.10 gave a maximum tensile and compressive stress of 1300 and 1800 psi at points A and B, respectively, per 0.001-inch deflection.

* This analysis, made by R. L. Call, accounted for anticlastic bending and used thick beam theory.

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FIGURE 4.7

Variation of Coefficient of Thermal Expansion and Clad Thickness with Position in the Coextruded Tube for Beta Heat Treated Inner Tube

FIGURE 4.8

Variation of Coefficient of Thermal Expansion with Position in the Coextruded Tube for Beta Heat Treated Outer Tube

FIGURE 4.9

Variation of Coefficient of Thermal Expansion with Position in the Coextruded Tube for "As Extruded" Inner Tube

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FIGURE 4.10

Schematic Representation of Compressed Inner Fuel Tube
Used in the Stress Analysis

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Supports in the autoclaved and unautoclaved condition were fatigue tested through a constant cyclic amplitude at a rate of 3200 cycles per minute. Any desired mean support deflection and alternating deflection could be selected by using the fixture illustrated in Figure 4.11. This fixture was cycled in a Sontag tester.

Failure, and even incipient failure, of the supports was detected by making the support serve as two halves of a bridge circuit, Figure 4.12, so that when either half of the support starts to crack the resulting change in resistance of that half of the bridge actuates a relay which terminates the test.

Results of the fatigue tests are summarized in Table 4.1. The autoclaved supports produce less scatter in fatigue life. Neither the bright etched nor the autoclaved supports failed when tested to 10^7 cycles through an amplitude of 0.006 inches.

Mechanical Tests on Strain-Cycled Zircaloy - K. R. Merckx

An apparatus for strain-cycling tubular specimens with increasing strain increments every cycle has been described in previous reports of this series. The apparatus is designed to set a given displacement between the two specimen grips with a jack loading head. The load is determined with strain gages mounted to the base plate, the specimen strain is measured with a dial gage mounted on an extensometer attached to the inner surface of the specimen, and the specimen is heated in an oil bath with a furnace. Initial tests were run on Zircaloy-2 specimens at 265 C undergoing strain cycles of ~ 1 percent or ~ 0.5 percent with the mean strain increasing ~ 0.1 percent or ~ 0.5 percent per cycle. Errors in the test procedure resulted from the manual control of the loading rate and from relaxation of the load on the specimen causing an additional strain of ~ 0.1 percent. Since the additional strain is of the same order of magnitude as the proposed strain increments per strain cycle, the data which has been obtained to date cannot be used in evaluating material models for fuel element cladding materials. Measurements of base plate and loading head movement during the strain holding periods are being made to determine the source of the additional straining.

Mechanical Properties of Zircaloy-2 Cladding - D. P. O'Keefe

The mechanical behavior of Zircaloy-2 cladding material is being investigated by means of tests on irradiated cladding and unirradiated tubular specimens. In this work the primary emphasis is on the plastic strain behavior of the cladding for various cladding thickness variations. In coextruded fuel elements, thickness variations in the cladding are associated with the degree of roughness at the uranium-cladding interface. Generally, this roughness is in the form of longitudinal strain on the inner surface of the Zircaloy cladding. Therefore, the effect of longitudinal notches on the strain behavior of tubular specimens is being studied.

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FIGURE 4.11

Fixture for Fatigue Testing

Supports Through a Constant Amplitude at a Preset Mean Deflection

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FIGURE 4.12

Schematic Representation of the Circuit Used
to Detect Fatigue Failure of Supports

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TABLE 4.1

FATIGUE PROPERTIES OF SUPPORTS

Support Number	Condition	Mean Compression, in. /1000	Alternating Compression, ± in. /1000	Mean Tensile Stress, psi	Alternating Stress, psi	Cycles to Failure, Cycles/10 ⁶
7	Bright Etched	20	10	26,000	13,000	0.246
32	Bright Etched	14	10	18,200	13,000	0.132
35	Bright Etched	20	10	26,000	13,000	0.240
46	Bright Etched	23	10	30,000	13,000	3.14
54	Bright Etched	13	10	16,900	13,000	0.141
60	Bright Etched	16	10	20,800	13,000	7.6
62	Bright Etched	20	3	26,000	3900	> 11.4
97	Bright Etched	20	10	26,000	13,000	0.396
98	Bright Etched	16	10	20,800		0.1
38	Autoclaved	20	10	26,000		0.18
41	Autoclaved	20	10	26,000		0.510
68	Autoclaved	20	10	26,000		0.420
92	Autoclaved	20	10	26,000		0.170
24	Autoclaved	20	10	26,000		0.250
81	Autoclaved	26	3	33,900	3900	> 10
12	Autoclaved	26	3	33,900	3900	> 10

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Some test results have been obtained on extruded and annealed 0.75-inch in diameter by 0.035-inch wall tubes tested at room temperature with hydraulic pressure. The initial test results for large radius and sharp notches are plotted in Figure 4.13. Additional data will be obtained at room and elevated temperature in order to establish the degree of notch sensitivity or notch strengthening associated with the two notch geometries.

FIGURE 4.13

Circumferential Elongation Versus Notch Fraction for 0.75 Inch Diameter x 0.035-Inch Wall Annealed Zircaloy-2 Extruded Tubing Notched Longitudinally on the ID. Tested at room temperature with hydraulic pressure.

The properties of irradiated coextruded Zircaloy-2 cladding are being sought by tests on 0.593-inch diameter rods which have been irradiated as cluster fuel elements. The uranium in 4-inch long sections of the rods was removed by dissolution in a 5M H_2SO_4 , 6M HNO_3 , 0.3M $CuSO_4$ solution.

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Unpredictably, the dissolving process produced pin holes in some of the Zircaloy-2 sheaths. Although some of the specimens did not have visible holes, the intended procedure of burst testing with hydraulic pressure did not appear feasible. Therefore, an expanding mandrel was developed that performed adequately on unirradiated specimens. This mandrel will be used for room temperature tests of the irradiated cladding specimens.

Fuel Charging Force Studies (Present Reactors) - J. P. Pilger

The first phase of fuel element charging force testing is complete. Particular emphasis has been placed on evaluating charging conditions associated with the "overbore" fuel element. The "overbore" fuel element weighs approximately twice as much per unit length as the present fuel element. Unit loading at the point of support in the overbore process tube is higher and warrants examination of the compatibility of this system. Fuel elements and process tubes used for these experiments were in reactor charging condition.

Charging forces were plotted on an x-y recorder for distances of 3 feet. Process water, or a 10 percent solution of A-60 soluble oil were used as lubricant. Each test series consisted of charges of one, two, and four fuel elements. The charge moved in a stick-slip action through the test section. Charging speeds of 6 feet per minute and 18 feet per minute did not change the stick-slip action, and no difference could be seen in peak force required to set the element in motion. This would be the predicted behavior since the coefficient of static friction should remain constant. All force peaks in each test were averaged to determine an average coefficient of static friction. Experiments were performed in a test tank under a static fluid, and no nozzle mock-up was used in the process tube. Test conditions are not, therefore, identical to those experienced at a reactor site. The calculated coefficient of friction from each test was used as the basis for comparison. The calculated coefficient rather than total charging force provides a common basis for comparison, and compensates for differences in element weight and support orientation. The comparison of friction coefficients was used to quantitatively evaluate the effects of support orientation and lubricants. The extent of process tube damage was evaluated qualitatively by visual inspection.

In summary, the following general observations were made:

- 1) In all tests involving self-supported fuel elements in ribless process tubes, the average coefficient of friction was reduced from 16 to 40 percent by orienting the supports in the 45° planes as opposed to vertical orientation.

- 2) Evaluation of the influence of charging oil indicates that in all cases of similar metal contact (aluminum-aluminum); a 10 percent solution of A-60 soluble oil in the water decreases the coefficient of friction from 13 to 50 percent.

- 3) Tests where water was the lubricant in the aluminum-aluminum system showed galling of the process tube and fuel element in all cases. Addition of soluble oil reduced the galling to incipient scratching.

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4) Tests involving dissimilar metals, (aluminum-Zircaloy-2), (nickel plate-aluminum), (nickel plate-Zircaloy-2), indicate that water lubrication is superior. Friction coefficients were 10 to 30 percent lower using water lubrication than with the soluble oil solution.

5) Present self-supports on the "bumper" fuel elements are not optimum in contact contour. Contact between supports and the ribless process tube approximates one or two "point" contact areas with associated high unit loading. This is particularly true in the overbore geometry. Table 4.2 gives a summary of average friction coefficients.

Measurement of Core Distortion During Heating - F. B. Quinlan

As previously reported, a device has been built to observe the behavior of and to measure the distortion of a fuel element during a temperature excursion. This device has been given a preliminary test which indicates that it will function as predicted.

The fuel element being tested was heated to 150, 200, and 250 C and in each case the light beam was adjusted to produce a bright spot of light at the center of the reticle. In order to produce a stress which would tend to distort the fuel element, a damp cloth was touched to one side of the element. Upon doing so, the light beam immediately indicated a bowing of the element in the direction of the chill and an immediate return to zero upon removal of the cloth. This was repeated in numerous directions and its behavior proved to be quite consistent.

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TABLE 4.2

SUMMARY OF AVERAGE FRICTION COEFFICIENTS

Test Series Number	Support	Support Orientation	Lubrication	Process Tube and Material	Average Coefficient of Friction
1	None	Ribbed Tube	Water	"0" Size Aluminum	0.533
6	None	Ribbed Tube	10% Soluble Oil	"0" Size Aluminum	0.416
2	None	Ribbed Tube	Water	Overbore Size Aluminum	0.523
7	None	Ribbed Tube	10% Soluble Oil	Overbore Size Aluminum	0.256
4	Aluminum	0 - 180°	Water	Overbore Size Aluminum	0.550
15	Aluminum	0 - 180°	10% Soluble Oil	Overbore Size Aluminum	0.323
16	Aluminum	45°	Water	Overbore Size Aluminum	0.296
9	Aluminum	45°	10% Soluble Oil	Overbore Size Aluminum	0.256
18	Aluminum	45°	Water	Overbore Size Aluminum	0.243
10	Aluminum	45°	10% Soluble Oil	Overbore Size Zircaloy-2	0.330
13	Nickel Plate Aluminum	0 - 180°	Water	Overbore Size Zircaloy-2	0.240
12	Nickel Plate Aluminum	22 - 202°	10% Soluble Oil	Overbore Size Zircaloy-2	0.350
13	Nickel Plate Aluminum	22 - 202°	Water	Overbore Size Aluminum	0.275
14	Nickel Plate Aluminum	22 - 202°	10% Soluble Oil	Overbore Size Aluminum	0.305

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FABRICATION DEVELOPMENT

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

For some time we have held the conviction that the production of satisfactory closures on NPR fuel elements by the self-brazing method is a matter of: 1) assembling the components under chemically clean conditions; 2) sealing the closure zone against atmospheric contamination by electron-beam welding the cap to the jacket walls; 3) applying sufficient heat and pressure simultaneously to the closure zone for sufficient time to bring about diffusion-bonding and self generated braze-bonding.

The mechanics of simultaneously applying heat and pressure have been rather baffling. Induction-heating the piece is simple, but applying pressure at the same time is difficult. Metallic restraining dies are not feasible since the induction coil generates most of the heat in the die rather than in the work piece, and the die metal loses its strength at the pressing temperature. A ceramic die constructed to eliminate this difficulty was unsatisfactory in that the ceramic liner cracked due to the thermal shock when the work piece was heated to the desired temperature.

Another approach involved lining a steel tube with a thin electrically and thermally insulating coating which was then smoothed by honing to provide a restraining die of desired diameter. With a suitable mandrel in the bore, the test element was placed in this restraining die and clamped under a five ton load between two special electrodes in the Sciacky heavy-duty spot-welder. Several pulses of heavy current were then passed through the element longitudinally. The first attempt was partially successful; the caps at both ends of the element were partially bonded, but it appeared that not quite enough heat had been generated to effect a complete bond. The insulating lining was damaged in removing the element.

A new die was provided, having a slightly larger bore, thus permitting a thicker lining layer to be applied. After honing, the liner was lubricated with Aquadag and another fuel element with welded-in, but unbonded, end caps was processed as described above. This time both closures appeared satisfactory. As judged by the nondestructive ultrasonic test they were more completely sound than any heretofore produced in the experimental closure program. However, removal from the restraining die again gave trouble and the insulating lining was damaged. The experiment was repeated with similar results.

It is now planned to make separable restraining die with a flame-sprayed inner coating. This is expected to permit removal of the work piece after processing without damage to the lining.

Some experimental work was carried out in an attempt to form a sound closure by restraining the element in a longitudinal direction by means of a stainless steel drawbolt of a diameter to provide the desired

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bore diameter. The closure end was heated by induction and immediately passed through a rotary swage to press the components intimately together while hot. This procedure did not produce sound bonds, apparently for two principal reasons:

- 1) The drawbolt had insufficient hot strength to restrain the longitudinal flow of the element metal under the swaging stresses, and
- 2) the pressure of the swage blocks was not applied for sufficient time in any given spot to permit generation of a diffusion bond between the components.

Since the spot-welder technique appears most promising, this method is now being pursued to the exclusion of other approaches.

Projection Welding of Recessed Closures - L. R. Vancott

Weld tests on the Projection Welded Braze Closure were designed to study each variable of the welding machine and to determine an effective weld schedule for a reactor grade closure.

Variables which are controlled on the welder such as cycles per impulse, number of impulses, pressure and pressure timing, quench, and others have little effect on bonding or welding. The primary controlling variable is phase shift (phase shift controls the magnitude of welding current and is calibrated in percent of full capacity). Outside diameter projection welds are readily formed at phase shift setting between 74 to 80 percent. Inner diameter welds in this current range collapse or are expelled. The maximum phase shift setting that may be used without expulsion of the ID projection is 70 percent. Seventy percent or lower produces a joint that has a definite interface line on both the ID and OD as shown in Figure 4. 14.

A difference in heating rate between the ID and OD projection is the cause of expulsion on the ID. When fusion temperature is reached on the ID, projection on the OD is still relatively cold and since the OD projection is rigid, it retards follow-up pressure on the ID.

Unbalanced heating was evident from the outset of these closure studies. All machine variables were not fully understood which made it necessary to perform several series of tests in an attempt to reach a weld schedule that would yield reactor grade closures. A satisfactory weld schedule was not achieved, however, correlation of machine variables and limits of parameters were established.

It appears that achieving a reactor grade closure will be dependent upon getting proportional current distribution between the inner and outer projection during welding. Future efforts will be directed towards this goal

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FIGURE 4.14

Projection Weld at 68 Percent Phase Shift Cap Wedge Removed
Macro taken at 10X and is one side of N inner fuel

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Brazed Closure Development - R. N. Johnson

The present brazing practices on NPR fuels utilize braze rings of the Zircaloy-5 wt% beryllium brazing alloy. This alloy is very brittle and difficult to fabricate and, consequently, the braze rings are quite expensive. Alternate closure methods are being investigated in the hopes of producing a better and/or more inexpensive closure. Two of these methods involve the vapor deposition of pure beryllium on zirconium end caps, thus requiring the development of an adequate vapor-deposition process. The first application would be the preplacement of beryllium on the caps to form the Zircaloy-5 wt% beryllium brazing alloy by diffusion in a high temperature operation. The second application would be the use of thin films of beryllium as an initial bonding agent in the projection welding of end caps to hot-headed fuel elements. The vapor deposition process chosen utilizes an electron-beam heating technique which makes possible the plating of

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beryllium films from a few angstroms thick to several mils thick. Initial plating difficulties have now been overcome and high-quality tightly-adherent beryllium films up to 0.0005-inch thick can be deposited consistently. Thicker films are possible and should be obtained shortly.

Hot-Heading Coextruded NPR Inner Tubes - P. A. Ard

Approximately 100 short lengths (4-3/4 inches long) of Zircaloy-2 clad uranium NPR inner coextruded tubes were hot-headed on one end for evaluation of closure welding techniques on the hot-headed ends. The hot-heading operation extrudes the uranium core from the end of the tube while forming the Zircaloy-2 cladding over the end of the tube. The final closure is made by welding a Zircaloy-2 cap to the Zircaloy-2 surface formed over the end of the tube. A cross-section of a hot-headed NPR inner tube is shown in Figure 4.15. The thin wall extrusion is machined flush with the end of the tube prior to welding.

FIGURE 4.15

Cross-section of Hot-headed Coextruded NPR Inner Tube
3X

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The tubes were contained in a full length heading container and the ends were back-extruded through an extrusion annulus located over the tube wall center. The elements were plated with a chemical copper plate and coated with a thin graphite lubricant to prevent the Zircaloy-2 from seizing to the steel dies. The tubes were induction heated to 640 C for heading.

The hot-heading operation produced very heavy shoulders on the OD of the tube ends which is desirable for the closure welding. The ID Zircaloy-2 shoulders did not thicken above the ID cladding thickness as can be seen in Figure 4. 15. The heading produces a "fanning out" effect on the 0.030-inch thick ID cladding moving from the tube ID to the annular extrusion orifice at the center of the tube wall. A "gathering" effect is produced on the 0.040-inch thick OD cladding moving in to the orifice, thus producing a very heavy shoulder. The thinner ID shoulder, while not as desirable as the heavier OD shoulders, may not prove to be restrictive to the closure welding.

Beta Heat Treatment of Ingot and Billet Stock - R. S. Kemper

Nine NOE (N outer enriched) and two NIE (N inner enriched) uranium coextrusion billets having greater than 2 ppm hydrogen by ingot analysis were given an alpha phase vacuum treatment to reduce the hydrogen level. One NOE billet was sampled before and after heating to determine hydrogen gradients. The cycle used was 8 hours at 600 C at a pressure of approximately 2×10^{-5} mm Hg. Three hours of this cycle were required to bring the billets to temperature. Two NOE, or two NOE and one NIE billets were heated in one furnace load. Four samples were taken across three wall thicknesses before and after heating. The hydrogen content was 2.0 to 2.5 ppm as received and was reduced to 0.9 to 1.1 ppm at the inner and outer quarters and to 1.8 to 2.0 ppm at midwall. These data indicate that the hydrogen was reduced to within specification but that the cycle time should be increased to reduce the concentration gradient.

A total of 59 uranium coextrusion billets have been beta heat treated during the past quarter as a final fabrication step before coextrusion. Pronounced improvement in inner clad thickness control was observed on subsequent coextrusion. The treatment consists of heating to 730 C in chloride salt, holding for 30 minutes total immersion time, and quenching with 30 ± 2 seconds transfer time to a circulated 1 percent salt solution at 25 ± 6 C. Six NIE billets have been processed with the same treatment. These coextrusions are currently being evaluated by the Fuels Preparation Department.

Two 11-inch diameter, 19-inch long NOE castings of iron-silicon additive material were received for heat treatment, primary fabrication, and coextrusion. One ingot was given a 15-hour alpha phase soak at 540 C (1000 F) followed by a single beta heat treatment, consisting of 1 hour 15 minutes immersion in 730 C chloride salt, and 50 second transfer to

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20 C circulated water quench. Primary extrusion, No. PU-65, was completed by Coextruded Products Engineering, sectioned for macro-examination, and two coextrusion billet lengths machined. Examination of the macrostructures and of nondestructive billet tester results indicate that the structure is at least equivalent, or superior, to those billets processed by the standard production process. Coextrusion of this material has been completed but evaluation is not complete.

Welding NPR Fuel Element Supports - W. I. Steinkamp

Support welding processes have been furnished for N fuel plant by Fuel Fabrication Development Operation for the outer fuel tube supports, the locking clips, and the inner fuel tube supports for the N elements. A topical report on the work done is in draft form. This completes the Fuel Fabrication Development Operation's portion of the project and future responsibility has been assigned to Fuels Production Development engineers.

Briefly, the weld schedules which have been furnished are: locking clips: (on the weld Model 10 spike power welder), weld time 2 cycles, heat control 70 percent, electrode pressure 17 psi air, transformer tap switch 3, and capacitor tap switch 2.

Inner supports: (on magnetic force welders), weld time 1 cycle, heat control unbalanced first half cycle 40 percent, last half cycle 52 percent, magnetic gap 1-1/4-inch, transformer tap switches low -3- low.

Outer supports: (magnetic force welder), weld time 1 cycle, heat control 60 percent, magnetic gap 1-1/4-inch, and transformer tap switches low -3- low.

A ± 5 percent is allowed on any one of these machine settings. Changes in support dimensions, tooling, etc., will require a revision in machine settings.

The tooling of primary interest is that used in the attachment of outer supports. This tooling positions the support for welding and adjusts the support to the proper height to get the required support circle independent of fuel element diameter. Figure 4.16 shows this tooling in operating position.

The tooling for inner supports consists of the same fuel element carrier mechanism with a means of placing inner supports in the proper position for welding.

The tooling for locking clips is a cantilever electrode and clip holder which spaces and welds the clip. This is shown in Figures 4.17 and 4.18.

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FIGURE 4.16

Locking Clip Tooling with Fuel Element in Welding Position

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FIGURE 4.17

Locking Clip Tooling with Fuel Element Withdrawn

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FIGURE 4. 18

Outer Support Sizing Tooling with Support in Jaws and Electrode in Welding Position

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Component Fabrication: Extrusion Behavior and Properties of Zirconium Alloys - P. A. Ard, R. S. Kemper, and R. G. Nelson

A series of zirconium alloys have been prepared by double vacuum arc melting that will be used to determine extrusion constant versus temperature data, resulting structure, mechanical properties, and corrosion behavior. The object of the work is to determine whether a corrosion resistant alloy more nearly matching the extrusion behavior of uranium and with greater ductility than Zircaloy-2 can be developed by altering the tin and oxygen content of a nominal Zircaloy-2 composition. Two zirconium sponge blends are being used to yield a high (1300 ppm) and low (560 ppm) oxygen content and for each blend the tin content is being varied from 0 to 1.50 wt%. The iron, nickel, and chromium contents are being held at the nominal Zircaloy-2 composition.

The fourth quarter, 1961 Quarterly Report, describes the melting and primary extrusion of these alloys. The pronounced effect of oxygen on the room temperature hardness of Zircaloy-2 was shown along with the hardening effect of tin. The effect of tin was less pronounced on the high oxygen series.

During the first quarter, 1962, each of the 16 alloy compositions were extruded at four different temperatures: 500, 600, 640 and 750 C. Four additional uranium control billets were extruded at 500, 600, and 640 C under identical conditions. At 640 C, two different extrusion ratios were used on the uranium. Normal production uranium with no additives was used. This material had been alpha extruded from 7-inch cast ingot at a 14 to 1 reduction ratio.

Listed in Table 4.3 are all of the alloys and their extrusion constants along with the temperatures and reduction ratios used. Figure 4.19 shows the stiffening effect of tin on each of the two oxygen levels at the 600 C extrusion temperature. It is interesting to note that at this temperature, it takes about 0.75 wt% tin in the low oxygen material to bring the extrusion constant up to the same value as the pure high oxygen material. Similarly, 1.5 wt% tin added to the low oxygen material has a greater hardening effect than the same amount added to the high oxygen material.

Figure 4.20 shows the change in extrusion constant with temperature of several of the alloys and uranium. In the 600 to 640 C temperature range where current coextrusion takes place, the Zircaloy-2 compositions have a 6 to 7 tons/in² greater "K" value than uranium. Even the low oxygen pure zirconium is still 1-1/2 to 2 tons/in² higher than uranium in this temperature range.

The stiffness of uranium increases rapidly with lower extrusion temperature and at 500 C the uranium has an extrusion constant of 17.1 tons/in² almost exactly matching that of 17.2 tons/in² of the low oxygen pure zirconium.

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TABLE 4.3
EXTRUSION CONSTANTS OF ZIRCONIUM AND ZIRCALOY-2

		Extrusion Temperature, °C							
		500 (13:1)*		600 (20:1)*		640 (13:1)*		750 (20:1)*	
	wt % Tin	"K" Run	"K" Start	"K" Run	"K" Start	"K" Run	"K" Start	"K" Run	"K" Start
700 ppm Oxygen	Zr	17.2	18.4	14.8	16.2	13.7	14.8	13.6	14.7
	0.1	18.4	20.1	15.6	16.8	15.4	16.7	13.9	14.9
	0.25	18.6	20.2	16.2	17.2	15.7	16.7	14.4	15.3
	0.50	20.0	20.9	16.5	18.1	16.1	17.3	14.8	15.9
	0.75	20.8	19.4	17.4	19.2	16.8	17.3	15.1	16.1
	1.00	20.9	22.7	17.8	19.4	17.1	18.8	15.5	16.4
	1.25	21.2	22.8	18.5	19.1	17.4	18.7	16.4	17.4
	1.50	21.8	23.3	18.7	20.7	18.1	19.6	16.8	18.2
1350 ppm Oxygen	Zr	19.9	21.3	17.5	18.9	16.9	18.3	14.3	15.7
	0.1	19.9	21.8	17.5	18.7	16.3	17.3	15.2	16.0
	0.25	20.6	22.1	17.7	19.1	16.9	17.8	15.7	17.2
	0.50	21.7	24.5	18.3	20.7	17.3	18.6	16.1	17.9
	0.75	22.0	23.5	18.7	19.9	18.1	19.2	16.5	17.6
	1.00	22.2	23.7	19.3	20.2	18.4	19.6	16.8	17.9
	1.25	22.6	25.2	19.9	21.0	19.0	20.2	17.7	19.6
	1.50	23.2	25.2	20.0	22.3	19.3	21.6	17.7	19.5
Uranium		17.1	18.4	12.8	13.8	12.5	13.9		
						12.0	12.9**		

* Reduction Ratio

** 20:1 Reduction Ratio

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FIGURE 4.19

Effect of Tin Content on Extrusion Constant of Zircaloy-2 at 600 C

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FIGURE 4. 20

Effect of Temperature on Extrusion Constants

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Twelve corrosion coupons of each alloy were prepared and have been started in test. Half of the samples are being run in 680 F water and half in 750 F - 1500 psi steam. Weight gain data will be taken at intervals up to approximately 130 days. A total of 128 tensile specimens, eight of each alloy, have been machined for tests from room temperature to 350 C. Transverse and longitudinal metallographic sections from the front and rear of each extrusion have been cut and the group extruded at 600 C have been examined for grain size, second phase distribution, and extent of recrystallization. The balance of the samples are in process.

Coextrusion Studies - G. S. Allison

A study of the effects of billet component composition and history, and can material on the coextruded clad-core interface roughness has been initiated. Nine billets of varying composition were coextruded from a 2.25-inch container to 0.600-inch rod at 13 to 1 extrusion ratio. Two clad materials were used--unalloyed, low oxygen zirconium, and a 3 percent niobium, 2 percent tin, high oxygen zirconium alloy. The Brinell Hardness Numbers were 117-128 and 220-230, respectively, which are below and above the 170-190 BHN hardness of Zircaloy-2. The uranium cores were cut from 14:1 alpha extruded normal ingot and from rolled and gamma heat treated dingot. The latter was used to simulate the large grained as-cast structure which produces extremely rough interfaces in coextrusion. Five of the billets were canned in 0.035-inch wall pure copper, two in 0.020-inch wall 1010 steel, and two were extruded with no can. The zirconium on all billets was sealed by vacuum electron beam welding. The following table shows the combinations of the components used:

TABLE 4.4

COEXTRUSION BILLET COMPONENT VARIABLES

<u>Number</u>	<u>Clad</u>	<u>Core</u>	<u>Can</u>	<u>Code</u>
1	Pure	Extruded	Copper	P-E-Cu*
2	Alloy	Extruded	Copper	A-E-Cu
3	Pure	Extruded	Copper	P-E-Cu
4	Pure	Dingot	Copper	P-D-Cu
5	Alloy	Dingot	Copper	A-D-Cu
6	Pure	Extruded	Steel	A-E-St
7	Alloy	Extruded	Steel	A-E-St
8	Pure	Extruded	Bare	P-E-Bare
9	Alloy	Extruded	Bare	A-E-Bare

* 500 C preheat; all others 620 C preheat

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Figure 4.21 shows the transverse sections of the coextrusions arranged for comparison of interface roughness. In all cases, the harder clad has a smoother interface than the softer. In all cases, the bare and steel canned extrusions have a smoother interface than those canned in copper. At 500 C preheat, where the pure zirconium and uranium have equal extrusion constants (equal stiffness), the interface is rougher than that of the same billet extruded at 620 C. Longitudinal sections have not yet been made, but could show different relative roughnesses or clad thickness variations than do the cross-sections.

FUEL EVALUATION

Irradiation of a Prototypic N-Reactor Fuel Element - R. L. Call

Irradiation of a full-sized NPR fuel element has continued in the GEH-M3 Loop of the ETR at the National Reactor Test Station in Idaho. This fuel element is of a tube-in-tube geometry. The outer tube has an OD of 2.404-inch and an ID of 1.758-inch. The inner tube has an OD of 1.253-inch and an ID of 0.439-inch. Zircaloy-2 cladding thickness on all surfaces is 25 mils except on the outer surface of the inner tube where it is 40 mils thick. The end closures were made by chemically milling uranium from the ends of the tubes and vacuum brazing a Zircaloy-2 ring into the recess using a Zircaloy-5 wt % beryllium braze alloy.

Early in January, the fuel element was discharged from the reactor for examination. The total average exposure at that time was 550 MWD/T. This was the second time the fuel element had been discharged for examination, the first having occurred after 300 MWD/T exposure. Warp measurements made during the first examination showed that both the inner and outer tubes had warped toward the center of the reactor core, or in a direction opposite to the horizontal flux gradient. When the test was recharged, the fuel element was rotated 180 degrees to determine whether or not the warp would reverse itself. When the fuel element was measured in January, the warp was found to have reversed.

Zircaloy-2 supports were spot-welded to the OD of the tubes to center the outer fuel tube in the basket and to center the inner tube within the outer tube. During the January examination of the fuel elements, three of the supports were found to be missing from the inner tube, two from the midlength of the tube and one from the lower end. It is believed that the damage to the supports was the result of fatigue induced by vibrations in the test loop.

The damaged inner tube was discarded and a spare inner tube charged in its place. The fuel element was again rotated 180 degrees so that it was in the same orientation as when it was first charged.

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P-E-Cu (500 C)

P-D-Cu

A-D-Cu

P-E-Cu

P-E-St

P-E-Bare

A-E-Cu

A-E-St

A-E-Bare

FIGURE 4.21

Macrographs of Cross Sections of Coextruded Rods, 3X

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817, 818, 0818, 0820.

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After an additional cycle of irradiation the test was again discharged for examination. The average total exposure for the outer tube at this time was 700 MWD/T. The average increases in OD of the outer tube was 0.004-inch. The inner tube, which had an average exposure of 150 MWD/T, had an average increase in OD of 0.001 inch. The warp of the outer tube had reversed again, thus confirming the theory that the direction of warp in a fuel element depends on the flux gradient across the fuel.

One of the supports on the inner tube had failed during the one cycle of irradiation. This support was at the upper end of the fuel element. A new inner tube will be charged for the next cycle of irradiation. The new inner tube will have "suit case" handles, rather than the "buggy spring" supports used on the previous two inner tubes.

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

An experimental irradiation of a single tube, dual enrichment fuel element is being conducted in the M-3 pressurized water loop in the ETR. The element has completed one cycle of reactor operation (27.3 days). Peak power generation in the element during this period was 180 kw/ft with a corresponding maximum metal temperature of 570 C. In keeping with the planned procedure for this test, the element was removed from the reactor after one cycle of operation. It will be examined and measured before recharging for additional exposure.

Tubular Test Element (KSE) - J. W. Goffard

Progress in the irradiation testing and postirradiation evaluation of a single component coextruded tubular fuel element has been reported in the previous four quarterlies. The test element (KSE) was conceived as a "stand-in" for the NPR outer fuel tube for testing purposes in the unmodified KER Loops.

Analysis of the KSE fuel swelling data has provided an empirical expression which relates total fuel swelling to the 1.65 power of burnup and temperature in the temperature range of 400 to 470 C.

The expression is:

$$\% \Delta V = 3.0 (\text{at. \% burnup}) + 0.0145 [(T_m - 365)(\text{at. \% burnup})]^{1.65}$$

T_m = Volume mean fuel temperature, C

1 at. % burnup ~ 8650 MWD/T

The experimental KSE swelling data for three exposures and calculated swelling values using the above expression are plotted in Figure 4.22, and illustrate the transition from the theoretical minimum to the theoretical maximum swelling. The theoretical minimum swelling is that which is

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required to accommodate the net gain of atoms resulting from fuel fission and may be expressed

theoretical minimum $\% \Delta V = 3.0$ (at. $\%$ burnup).

FIGURE 4.22

Fuel Swelling Versus Volume Mean Fuel Temperature for KSE Elements

The theoretical maximum swelling includes that which results from the agglomeration of gaseous fission atoms to form bubbles in the fuel that are at pressure equilibrium with the effective restraining forces. An expression for the theoretical maximum swelling assuming the loop coolant pressure (1600 psi) to be limiting restraining force is

theoretical maximum $\% \Delta = \text{theoretical minimum} + (\text{at. } \% \text{ burnup})$

$$(550) \left(\frac{T_{ma}}{273} \right) \left(\frac{14.7}{1600} \right)$$

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burnup = Percent of uranium atoms fissioned

T_{ma} = Volume mean fuel temperature, absolute

550 = (100)(5.50 cc xenon and krypton/cc uranium/at. % burnup)

It is seen in Figure 4.22 that the experimentally observed swelling did not attain the calculated theoretical maximum based upon an effective restraining force of 1600 psi loop coolant pressure. At points A, B, and C in Figure 4.22, the maximum observed swelling for the three exposures, the calculated effective total restraining forces are 2900, 2200, and 2100 psi, respectively. If points A, B, and C are actual limits, as may be suggested by the clustering of the data, then the total effective restraint is greater than that of the loop coolant pressure alone. The additional restraint may be that offered by both cladding and surface tension restraint within the fuel.

NPR Inner Fuel Tubes (NIN and NIE) - J. W. Goffard

The fabrication and irradiation testing of prototype inner tubes of the NPR fuel element assembly, as described in the previous quarterly, is continuing. The NIN elements contain natural uranium and were used in the earliest KER irradiation tests. The NIE elements contain 0.95 percent enriched uranium with iron and silicon additions for grain refinement and have been used in the recent irradiation tests.

A total of 35 NIN and NIE elements irradiated to various exposures have been weighed in the K-reactor basin for the determination of fuel swelling. Table 4.5 summarizes the data obtained.

The NIN elements irradiated to the 1000 and 2100 MWD/T exposure levels have been examined in the Radiometallurgy facility. The performance of these elements has been excellent, with no observed inadequacies or deficiencies in the Zircaloy-beryllium eutectic brazed closures or in the braze heat-affected zones of the fuel, clad, or bonds. Outside diameters increased an average of +0.002-inch for the 1000 exposure pieces and +0.012-inch for the 2100 exposure pieces. Postirradiation warp has not been severe with the maximum single throw warp being 0.042-inch on the 2 foot long elements.

The NIE elements irradiated to 900 and 2300 MWD/T exposure levels have been examined in the Radiometallurgy facility. Their performance has been comparable to that observed with the NIN elements. There have been no observed inadequacies or deficiencies in the braze closures or in the braze heat-affected zones. No observable effects, either desirable or detrimental, resulting from the iron and silicon additions in the NIE fuel material, have been observed.

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TABLE 4.5

NIN AND NIE FUEL SWELLING DATA
AT NPR INNER FUEL TUBE OPERATING CONDITIONS

Approximate Exposure, MWD/T	Number of Elements	Average Fuel Swelling, + vol %	Remarks
1000	4	0.4	NIN
2100	5	2.7	NIN*
500	3	0.3	NIE
750	5	0.8	NIE
750	4	0.3	NIE*
900	1	1.1	NIE
1000	5	0.2	NIE*
2300	3	2.4	NIE
3000	5	1.7	NIE
	35		

* Data obtained with refined techniques and equipment. Results accurate to approximately ± 0.1 percent. Other data accurate to approximately ± 0.5 percent.

Fuel Element Swelling Model - K. R. Merckx

Test data on fuel element irradiations should provide a means of evaluating parameters in a model for swelling of uranium. Since various fuel element geometries have different temperature and burnup distributions while only mean volume expansions of the fuel element can be determined, empirical models for statistical evaluation of fuel element data must contain some procedure for averaging the irradiation conditions. An empirical model was formulated based upon the concepts that the volume must increase at least as much as the volume of the new atoms formed by fissioning, the volume should not increase more than the volume of the fission gas products at the coolant pressure and the self-restraint of the fuel is a power function of the temperature and the burnup. The resulting equation for the mean percentage volume increase of the fuel element is:

$$\bar{v} = \frac{2}{r_o^2 - r_i^2} \int_{r_i}^{r_o} \left[\frac{300 + p}{b^{\frac{1}{2}} 2 T^{0.3}} \right] r \, dr$$

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where $\bar{v}$ = average volume increase
 r_o = outer fuel element radius
 r_i = inner fuel element radius
 b = burnup [%] as a function of radius
 T = absolute temperature [C]/273 as a function of radius
 p = coolant pressure in atmospheres
 $\theta_1, \theta_2, \theta_3$ = parameters

A computer program "NELLY" already existed which could statistically evaluate the parameters in such a model based on weighted mean squared errors. The additional programming and punch card data preparation are presently being completed for numerical evaluation of existing experimental data.

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

The production brazed irradiation test, GEH-4-63 and 64, brazed with the Zircaloy-5 wt % beryllium brazing alloy, has completed six cycles of irradiation at the MTR and had been returned to Hanford for radio-metallurgical examination. The elements operated at a specific power of 62 kw/ft and accumulated a total burnup of approximately 1390 MWD/T. Initial radiometallurgical examination has shown no indications of "incipient" failure on the surface of the elements; however, markings which appeared to be stressed areas in the cladding were noticed around the two end-caps (one end-cap on each element) which operated in the high-flux region of the GEH-4 loop. These areas are being sectioned to determine the integrity of the brazed closure and the cause of the stressed-appearing areas.

The variable-brazed-thickness test, GEH-4-68, 69, and 70, has been discharged after completing five cycles of irradiation at the MTR. A sixth cycle has been requested and approved and the elements are to be re-inserted for the next MTR cycle (Cycle 172). The elements operated at a specific power of 80.7 kw/ft and have accumulated a burnup of approximately 1140 MWD/T.

Thermocoupled Fuel Element Tests - R. L. Call

In the recent past, three different thermocoupled fuel rods have been charged into one of the high temperature recirculating water loops in a Hanford reactor. These rods were charged as part of a program to study "crud film" formations on fuel elements. Each of the three tests was terminated by a failure, two by failure of the thermocoupled rod and one by failure of a test fuel element.

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In each of the first three tests, the thermocoupled rod had three thermocouples in it, two just under the Zircaloy-2 cladding and one in the center of the core. The failures in the thermocoupled rods were believed to be associated with the thermocouples which were just under the cladding.

A new Zircaloy-2 clad thermocouple probe was designed. The new probe has a Zircaloy-2 core inside a ring of enriched uranium. A single thermocouple is installed in this Zircaloy-2 core.

At the present time, a probe of the new design has accumulated 770 MWD/T of irradiation exposure in a high temperature loop. Measured core temperatures ranged from 605 to 640 C during equilibrium operating periods. Current plans call for a decontamination of the loop with the probe in place during the next outage in order to study "crud film" formation following loop decontamination.

PRTR Flux Wire Fabrication - R. G. Nelson and F. B. Quinlan

The Ceramic Fuels Operations has requested the melting and fabrication of approximately 2500 feet of 0.073-inch diameter wire, the composition of which is to be 1.50 wt % tin, 0.10 wt % chromium, 0.15 wt % iron, 0.50 wt % cobalt, balance zirconium. Two 3.8-inch diameter 24 pound ingots of this composition have been double vacuum arc melted and processed down to 0.125-inch diameter.

Melting

Electrodes were prepared by compacting mixtures of the alloying elements and zirconium into 1-1/2-inch octagons 12 inches long. These bars were electron beam welded together end to end to form first melt electrodes. Two primary ingots were made for each final ingot produced. The primary ingots were scalped, quartered longitudinally, and the eight quarter sections welded together to form the electrode for the final melt. Melting conditions are shown in Table 4.6.

TABLE 4.6

	<u>Volts</u>	<u>Amperes</u>	<u>Vacuum μ</u>
Primary	37 Start 31 Finish	2100	10 to 20
Secondary	34	3800	3 to 7

Ingot Evaluation

Figure 4.23 schematically represents the hardness and cobalt distribution in the two ingots produced. There is an apparent increase in cobalt from bottom to top of the ingots.

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FIGURE 4.23
Ingot Evaluation

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The analyses shown in Table 4.7 are from the cast sample taken at the top of the ingot. The cobalt distribution samples were taken from the ends of the 1/2-inch diameter secondary extrusions and represent the approximate locations shown in Figure 4.23.

Cursory autoclave tests made on the 1/2-inch diameter rod stock show a black tight adhering autoclave film with no evidence of detrimental corrosion. Test conditions were 12 hours, 400 C 1500 psi steam.

TABLE 4.7
INGOT ANALYSIS

		Heat Number 113	Heat Number 114
Cobalt	(wt %)	0.50	0.52
Iron	(wt %)	0.105	0.096
Chromium	(wt %)	0.101	0.100
Tin	(wt %)	1.49	1.49
Hydrogen	(ppm)	6	6
Oxygen	(ppm)	730	820
Nitrogen	(ppm)	19	29
Copper	(ppm)	31	32
Silicon	(ppm)	46	43
Uranium	(ppm)	0.5	0.5
Aluminum	(ppm)	58	57
Boron	(ppm)	<0.2	0.3
Cadmium	(ppm)	<0.3	<0.3
Hafnium	(ppm)	60	58
Magnesium	(ppm)	<10	<10
Manganese	(ppm)	15	15
Molybdenum	(ppm)	<10	<10
Nickel	(ppm)	<10	<10
Sodium	(ppm)	<20	<20
Lead	(ppm)	15	15
Titanium	(ppm)	<20	<20
Vanadium	(ppm)	<20	<20
Tungsten	(ppm)	<40	<40

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Fabrication

Processing of the ingots to wire is being done by three basic steps: hot extrusion, cold swaging, and cold drawing with interim vacuum annealing.

The cast ingots were machined and upset bare at 750 C from 3.6 to 4 inches diameter, machined lightly and extruded bare at 750 C to 2-1/8-inch diameter. Four billets from each ingot were machined, canned in copper, and extruded at 750 C to 1/2-inch diameter. The copper was removed by pickling in HNO_3 . The stripped rods were then pickled in the standard Hf- HNO_3 Zircaloy-2 bath. It is interesting to note that the pickling rate of this alloy is about one-half that of Zircaloy-2.

The 1/2-inch diameter hot-extruded and pickled rods were cold-swaged to 130 mils in diameter.* After each 40 to 50 percent reduction in cross-section area, the swaged rods were vacuum annealed at 750 C for one hour. As successive swaging reductions were made, the surface of the rods improved till a 16 to 32 microinch finish was attained at the final pass. From this point on, the rods will be drawn into wires by use of the draw-bench and carboloy wire drawing dies.

REFERENCES

1. Merckx, K. R. Deformation of Shaped Cylindrical Reactor Fuel Elements, HW-70501. May 5, 1961.
2. Merckx, K. R. Personal communication. November 10, 1961.

* First centerless ground to remove the ridges formed during extrusion.

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CERAMIC FUELS DEVELOPMENTCERAMIC (URANIUM) FUELS RESEARCHFission Fragment Migration in UO_2 - J. L. Bates, J. A. Christensen and W. E. Roake

Nonuniform concentration of fission fragments and plutonium in irradiated uranium oxide effectively nullifies any correlations of properties and performance with nuclear burnup. Earlier quarterly reports of this series discussed experiments that revealed extensive relocation of fission fragments from the points at which the kinetic energy of fission was expended and of plutonium from its location of genesis. New data from a high-rated, sintered UO_2 pellet, and a vibrationally compacted UO_2 - ThO_2 mixture parallel those reported earlier for a vibrationally compacted UO_2 fuel rod.

Microradiochemical analyses for fission fragments and plutonium in an irradiated, sintered UO_2 pellet (9/16-inch diameter, 0.27×10^{20} fissions per cc, 31 kw/ft) show approximately the same correlation between fission fragment concentration and the microstructure of the UO_2 that was observed in the vibrationally compacted element (1.44-inch diameter, 0.08×10^{20} fissions per cc). Results of both sets of analyses are shown in Figure 5. 1. In each element the principle feature of the distribution is depletion of Zr-Nb⁹⁵, Cs¹³⁷, Ru¹⁰⁶, Ce¹⁴⁴, and Pu in the high density, large columnar grains. The region of Cs¹³⁷ depletion in the sintered pellet is somewhat broader than for the other isotopes and the minimum Cs¹³⁷ concentration occurs 0.05-inch farther from the thermal center of the fuel. The greater mobility of the cesium is thought to be related to its relatively high volatility.

Swaged UO_2 , irradiated to 10×10^{20} and to 14×10^{20} fissions per cc (as reported by reactor operations) and a vibrationally compacted mixture of UO_2 -10 wt % ThO_2 (0.09×10^{20} fissions per cc) were sampled at 0.05-inch intervals along diameters of cross sections of the fuel rods. Direct counting of the samples before dissolution yielded approximate distributions of Zr-Nb⁹⁵ and Ru¹⁰⁶, as normalized to Ce¹⁴⁴. The distribution in the UO_2 - ThO_2 element correlates well with those previously obtained for other irradiated fuel cores, but both high burnup elements show relatively less segregation, as was also apparent from autoradiographs. These observations support the hypothesis that much of the fission fragment segregation is a consequence of the same process by which the equilibrium structure of the fuel is reached, i. e., a vapor phase zone refining process that forms the large columnar grains and transports vaporized impurities toward the center of the fuel. If a stable fuel structure is formed and not later modified, no further gross segregation by that mechanism would be expected and bulk diffusion should tend to reduce that which has occurred.

Vipac 1.44-Inch OD 300 MWD/T_U Sintered Pellets 0.560-Inch OD
1000 MWD/T_U

FIGURE 5. 1

Fission Fragment Segregation in UO_2 Fuels

Sampling techniques were improved by using strips of cellulose acetate, thoroughly wet with acetone, to remove UO_2 particles from a scratch, also dampened with acetone. The cellulose acetate, when dry, is pulled from the surface and many small particles of loosened UO_2 adhere to the cellulose acetate. The line of fine powder has a UO_2 concentration of approximately 3 mg/inch of scratch length. The strip is cut into segments that are analyzed radiochemically for fission fragments and plutonium.

A facility for scanning polished fuel cross sections with a gamma scintillation crystal was installed by Radiometallurgy Operation. Lead collimators (7 inches long and 0.030 to 0.060-inch diameter holes), a 2-x 2-inch NaI crystal, and a 200 channel pulse analyzer permit measurement of the relative concentrations of eight radioactive isotopes in small selected areas of a fuel rod cross section. The first irradiated sample studied was a 1/16-inch thick wafer of the 1.44-inch diameter vibrationally compacted fuel element used for the initial radiochemical determination of the fission fragment distribution. The resulting data were similar to those obtained radiochemically, but the sample activity was too low for reasonable counting times. More active specimens are being prepared.

Fission fragment depletion by recoil loss at the boundaries of UO_2 fuel cores was investigated by measuring the concentration distribution of fission fragments in aluminum that had been in contact with a UO_2 single crystal during irradiation to 1.44×10^{20} fissions per/cc. One micron thick layers were removed from the aluminum surface by chemical milling in a 12.5 percent NaOH solution. The plot of concentration versus depth of penetration showed a maximum at a depth of approximately 13μ for each isotope.

Electron Microscopy - J. L. Daniel and J. O. McPartland

The microstructures of uranium dioxide crystal surfaces are being studied in detail, to assist in interpreting effects of irradiation on physical and chemical properties. A crystal of irradiated UO_2 was examined directly in the electron microscope, for the first time, using reflection microscopy techniques. The material was a piece of single crystal UO_2 , irradiated to 1.44×10^{20} fissions/cc. Temperature of the sample surface at the point of observation during microscopy is estimated to be between 500 and 1000 C, due to heating by the electron beam. The fracture surface examined was similar to that of a nonirradiated crystal examined earlier; initial examinations showed a wide variation in surface characteristics, but no unique features have been identified. Examination of additional specimens is in progress, and an arc image furnace for elevated temperature observations is under construction.

Comparison of similar areas of the irradiated UO_2 examined by reflection and by a conventional positive replica technique (Figure 5.2)

Direct Reflection

Transmission Replica

FIGURE 5.2

Electron Micrographs of Irradiated UO_2 Single Crystal
(Similar Fracture Surfaces Examined by Direct Reflection
Electron Microscopy and by Positive Transmission Replication)
Irradiation: 1.44×10^{20} Fissions/cc

5000X

shows the similarity and differences in the two methods. Replicas provide better resolution, although they are difficult to prepare from small or very smooth samples. Reflection samples emphasize surface contours and vertical structure, and allow sample treatments such as heating and gas reaction during examination. Reflection image shapes are somewhat distorted because of the sample position during reflection observations.

Postirradiation Examination of UO_2 Single Crystal - J. L. Bates,
J. A. Christensen, and J. L. Daniel

Although uranium dioxide is relatively stable under irradiation, changes occur in many properties. Little is known of the damage processes that cause these property changes or the limitations that they impose. The basic processes of irradiation damage are being studied intensively, by characterizing single crystals of UO_2 before irradiation and repeating all preirradiation measurements after irradiation.

A single crystal of UO_2 , 0.89 cm square cross section x 3.8 cm long, was machined from a large, fused boule and sliced transversely into 0.14-inch thick sections and irradiated in 1.44×10^{20} fissions/cc. Two adjacent sections were used for chemical analyses, hardness measurements, high temperature observations of melting and vaporization behavior, and intensive electron and light microscopy of polished and fractured surfaces.

One section was metallographically polished on one face, precharacterized and encapsulated in aluminum for irradiation in the Engineering Test Reactor under conditions summarized in Table 5. 1.

TABLE 5. 1

IRRADIATION CONDITIONS FOR UO_2 SINGLE CRYSTAL

<u>Exposure</u>	1.44×10^{20} fissions/cc 62.5 equivalent full-power days
<u>Unperturbed Flux</u>	
Thermal	$\sim 5 \times 10^{14}$ nv
Fast	$\sim 1 \times 10^{14}$ nv
<u>Maximum Heat Generation</u>	
U^{235} fissioning	78 watts/gram of UO_2
Gamma heating	12 watts/gram of UO_2
<u>Calculated UO_2 Temperatures</u>	
Surface	~ 325 C
Maximum centerline	~ 400 C

No detectable (less than 0.03 percent) amounts of xenon or krypton were released from the 0.966 gram crystal, although a calculated 0.13 cc/gram of UO_2 was produced during irradiation. Approximately 0.2 cc of hydrogen was released into the capsule void during irradiation (Table 5.2).

TABLE 5.2

MASS SPECTROGRAPHIC ANALYSIS
OF GAS REMOVED FROM CAPSULE

(GEH-14-242)

CONTAINING IRRADIATED UO_2 SINGLE CRYSTALS

	<u>Xenon</u>	<u>Krypton</u>	<u>Helium</u>	<u>Argon</u>	<u>H₂</u>	
Volume Percent	<0.01	<0.01	24.7	0.07	71.8	
Gas Volume (STP) x 10 ² cc	<3.3 x 10 ⁻³	<3.3 x 10 ⁻³	8.15	0.023	23.7	
	<u>N₂</u>	<u>O₂</u>	<u>CH₄</u>	<u>H₂O</u>	<u>CO₂</u>	<u>Other</u>
Volume Percent	1.92	0.61	0.52	0.29	0.08	0.01
Gas Volume (STP) x 10 ² cc	0.634	0.20	0.17	0.096	0.076	0.0033

The helium in the capsule gas was introduced during encapsulation and welding. The other gases were presumably released during irradiation. Hydrogen, nitrogen, argon, water, and carbon dioxide are commonly present in nonirradiated, fused UO_2 crystals. However, the maximum bulk UO_2 temperature of 400 C during irradiation is significantly lower than the 1000 C generally used to extract chemisorbed gases from nonirradiated UO_2 . Localized high temperatures due to fission events and the subsequent alterations of the UO_2 lattice are thought to have caused the release of these gases at the lower bulk temperatures.

The general appearance of the crystal was not changed by irradiation. The crystal broke into five pieces during the irradiation or during the postirradiation handling. Subsequent intentional fracture indicated a highly stressed and brittle crystal. Hardness of the UO_2 was significantly increased from its preirradiation KHN values between 660 and 760, (Figure 5.3). Both the preirradiation and postirradiation hardness differed as the orientation of the Knoop indenter axis was varied with respect to the crystal surface (Figure 5.3).

Crystal A-7-B

Crystal A-7-D

Specimens Irradiated to 1.44×10^{20} Fissions/cm³. Preirradiation Hardness Was 686 ± 12 . Arrows Indicate Direction of Long Axis of Knoop Indenter.

FIGURE 5. 3

Microhardness of Irradiated UO₂ Single Crystals
Showing Dependence on Crystal Orientation

The melting point of the UO_2 was increased approximately 50 C from the preirradiation value of 2790 C, and the sublimation rate of UO_2 was noticeably suppressed. No gross fracturing or spalling was observed during heating.

Electron microscopy of the irradiated single crystal was conducted by both replication and direct (reflection) techniques. Irradiated polished surfaces, repolished surfaces, and fracture surfaces were examined. A wide variation in surface characteristics was found over relatively small areas. However, no unique features definitely attributed to irradiation were identified. No differences were observed by light microscopy of the repolished surfaces after irradiation. Examination of samples and study of micrographs are continuing.

Single Crystal UO_2 Studies - H. J. Anderson

Large single crystals of UO_2 are found in quantity in bulk fused UO_2 supplies. Spherical and rectangular specimens have been purified and characterized for fundamental studies, both at Hanford and at other sites. The specimens were machined from large, single crystals of UO_2 that had been heat treated in hydrogen containing ~1000 ppm H_2O at 1750 C to (1) eliminate very small inclusions, (2) assure stoichiometry, and (3) relieve stresses. Characterization studies (Table 5.3) upon the bulk materials included: O:U atom ratio, density, porosity, carbon content, gas evolution, microstructure, and impurity levels (determined spectrographically). Radiographic examinations of the final specimens confirmed the absence of porosity and inclusions. X-ray diffraction measurements were made to confirm crystallinity of specimens.

Additional property measurements and irradiation performance tests are in progress.

TABLE 5. 3

UO₂ SINGLE CRYSTALSI. BULK MATERIALS

<u>Property</u>	<u>A</u>	<u>B</u>
O:U Atom Ratio (Coulometric-Titration)	2. 00 <u>1</u>	2. 00 <u>7</u>
Carbon, (ppm) (Combustion Train)	24	34
Density, (g cm ⁻³) (Water Displacement)	10. 8 <u>4</u>	10. 9 <u>5</u>
(Mercury Displacement)	10. 8 <u>4</u>	10. 9 <u>0</u>
Porosity - Open, (%) (Mercury Porosimetry)		

<u>Pressure</u> (psi)	<u>Pore Diameter</u> (Microns greater than)		
1. 77	100	0	0
19. 1	9	0	0
3000	0. 06	0	0

Impurity, (ppm)
(Spectrographically) less than 200 parts per million

Gas Content, (cm³ g⁻¹)
(Vacuum Extraction at 1000 C) 0. 08 0. 10

II. SPECIMENS

	<u>No. 1</u>	<u>No. 2</u>	<u>No. 3</u>
Weight, (gram)	1. 407 <u>8</u>	1. 384 <u>0</u>	10. 552 <u>2</u>
Volume, (cm ³)	0. 131 <u>4</u>	0. 127 <u>2</u>	0. 961 <u>4</u>
Density, (g cm ⁻³)			
Mensuration	10. 71	10. 88	10. 98
CCl ₄	10. 9 <u>6</u>	10. 9 <u>1</u>	10. 9 <u>4</u>
H ₂ O	10. 9 <u>4</u>	10. 9 <u>3</u>	10. 9 <u>5</u>
Dimensions, (inch)	0. 449 OD x 0. 41	0. 0534 OD x 0. 039	0. 249 OD x 1. 201

Simulated Nuclear Fuel Heating - J. A. Christensen and L. E. Mills

Out-of-reactor experiments are valuable adjuncts to irradiation tests if they enable better understanding of the mechanisms of the changes that occur in fuel structure during irradiation. Nuclear fuel heating was simulated by self-resistance heating of UO_2 in a closed capsule, the intent being to provide a simple method of studying fuel relocation, fission product migration, fuel cracking, and void formation in various nuclear fuel materials. Electrical connection was provided at each end of the fuel through insulated tungsten electrodes. The outer surface of the fuel section was cooled with a water jacket. Microscopic examination of the fuel revealed evidence of central UO_2 fusion. The fused core was surrounded by nonfused UO_2 adjacent to the cladding. Both circumferential and radial cracks were evident. The section appeared similar in many respects to sections of irradiated fuel rods.

Nuclear fuel heating of tungsten-clad UO_2 was simulated at cladding temperatures between 2000 and 2800 C, to anticipate problems during in-reactor gas cooled experiments. Test capsules consisted of sintered pellets of UO_2 enclosed in self-resistance heated tungsten vessels. The first test capsule contained approximately 4 grams of UO_2 in a 1/4-inch diameter x 1-inch long container. The capsule was re-radiographed after thirty minutes at 2000 C. The only observable effect of the heating was fracturing of the UO_2 pellets by thermal shock during heating and cooling. The capsule was subsequently cycled ten times between room temperature and 2400 C (15 seconds heating and 20 seconds cooling), held at 2400 C for thirty minutes, and cycled an additional ten times. Radiography again showed no damage to the capsule except for additional thermal fracture of the UO_2 .

A second capsule containing 1.5 grams of UO_2 in a 1/8-inch diameter x 1-inch long tungsten vessel was heated to 2800 C, at which time the UO_2 was melted. The molten UO_2 flowed readily through 0.001-inch wide cracks in the wall of the container. A radiograph showed that the UO_2 had fused and resolidified to form a deep meniscus and a large shrinkage cavity. The capsule was sectioned longitudinally and given a rough metallographic polish. The UO_2 appeared much like UO_2 fused in-reactor. Large equiaxed grains containing many large spherical voids had formed on solidification and no columnar grain growth was apparent.

Induction Melting of UO_2 - L. E. Mills

Atmosphere contamination of UO_2 during arc fusion and cooling led to evaluation of alternate methods of fusion. Tests were performed to determine the effect of atmosphere on melting of UO_2 contained in a tungsten susceptor. Gross UO_2 vaporization occurred in a helium atmosphere, and no pool of UO_2 was obtained. The vapors deposited on a small area adjacent to the induction coil. The UO_2 vapor traveled about 1-1/2 inches, passing a cool tungsten zone to deposit on the hot tungsten surface. Relatively little UO_2 was vaporized in an argon atmosphere. However, the pool of molten UO_2 flowed readily through a 0.005-inch wide gap in the tungsten susceptor

and solidified into spheres and fingers as it dropped through the surrounding argon atmosphere. Shrinkage during solidification produced large central voids in the spheres and fingers of UO_2 .

Fused UO_2 Quality - D. W. Brite and L. P. Murphy

Uranium dioxide core material for vibrationally compacted fuel elements must be free of pores if high bulk densities are to be attained. HAPO purchase specifications for fused UO_2 prescribe a minimum UO_2 density of 10.80 g/cc, as measured by mercury displacement after evacuating air from the sample and back-filling with mercury to one atmosphere pressure. By this technique, open pores larger than 10 microns diameter are filled with mercury, and their volume is excluded from the density determination. Recent measurements with a mercury porosimeter revealed that some lots of fused UO_2 have a significant volume of open pores with diameters in the range 9-100 microns. Average measured density of two lots of fused oxide decreased from 10.84 g/cc to 10.72 g/cc when the minimum mercury-filled pore diameter was increased from 9 to 100 microns. Ceramographic examination revealed that these two lots of UO_2 consisted primarily of polycrystalline material with grain diameters of about 500 microns, as is illustrated in Figure 5.4. A large number of voids, each approximately 15 microns wide and 200-300 microns long, can be seen at the grain boundaries. Open porosity which is undetected by normal liquid displacement techniques can thus be quantitatively determined by mercury porosimetry.

Porous, foamed layers are occasionally formed during fusion of UO_2 . The porous material is sometimes inadvertently included with solid, fused UO_2 at the vendor's plant. An elutriation method for separating impurities and relatively porous material from fused or electrodeposited UO_2 was investigated. Water, passing through a fluidized bed of UO_2 particles of narrow size distribution, removes the lighter particles. Figure 5.5 reveals the difference in appearance of the two materials separated by elutriation from a minus eight, plus ten mesh size fraction of fused UO_2 . Bulk densities of the light and heavy fractions were 4.5 and 5.8 g/cc, respectively. The technique is thus effective for upgrading fused UO_2 which is of marginal average particle density because of contamination by a few highly porous particles.

FIGURE 5.4
Polycrystalline Fused UO_2 50 X

Light Particles

Heavy Particles

FIGURE 5.5
Variations in Porosity of Fused UO_2 10 X

High Energy Impact Forming - D. W. Brite and L. E. Mills

The feasibility of high energy impact forming of ultra-high density (99 percent TD) UO_2 fuel material with a Dynapak machine has been reported in previous quarterly reports of this series. The technique comprises impaction of UO_2 powder in a heated, stainless steel container within an expendable steel ring, between Bridgman anvils, at an impact pressure of approximately 500,000 psi. A tungsten carbide punch was recently used to impact UO_2 at approximately 500,000 psi directly in a die without using Bridgman anvils or expendable steel rings. Direct impaction of ultra-high density UO_2 fuel material thus appears to be economically more attractive than previously thought.

Uranium dioxide powder packaged in stainless steel foil, rather than in cans, was successfully heated under conditions required for subsequent compaction in the Dynapak machine. A significant reduction in the expense of high energy impaction should result if foil-wrapped powder packages are used. Behavior of foil-wrapped powder packages during high energy impaction will be determined.

The new Model 1220-B Dynapak machine was installed and trial operated. The machine has a vertical operation with a maximum rated energy of 145,000 foot-pounds. The machine will be used for further evaluation of the high energy impaction process for preparing ultra-high density ceramic materials.

Hot Isostatic Pressing - J. J. Hauth

A program is being conducted jointly with BMI to evaluate hot isostatic pressing of fuel elements that are partially predensified by swaging or vibrational compaction, or both. Variables being investigated include cladding material and dimensions, and fuel material and particle-size distribution. Approximately 60 UO_2 rods (~16 inches long) were fabricated and hot isostatically pressed in exploratory experiments to define the areas of greater interest. Results of these experiments, although not yet complete, are summarized as follows:

1. Micronized UO_2 , hot-swaged in Inconel-X (0.020-inch wall) to 70 percent TD, was compacted to >98 percent TD by hot isostatic pressing at 2100 F, 10 kpsi for 3 hours. Rod straightness and surface condition are good (Figure 5.6, Rod No. 1).
2. Fused UO_2 , either cold swaged to 88 percent TD or hot swaged to 92 percent TD in 0.030-inch wall Zircaloy, was increased in density by ~2 percent TD during hot isostatic pressing at 1550 F, 10 kpsi for 4 hours. Rods were straight, with smooth surfaces (Figure 5.6, Rods 8 to 24).

FIGURE 5.6
Hot Isostatically Pressed UO_2 Rods Fabricated by Swaging or Vibrational Compaction
and Sealed in 0.022-Inch Wall Inconel-X (Rod No. 1) or 0.030-Inch Wall Zircaloy

3. Electrodeposited UO_2 , vibrationally compacted in 0.030-inch wall Zircaloy to 89 percent was increased to 94 percent TD under the same conditions. The rods were straight and smooth (Figure 5. 6, Rod No. 6).
4. Mixtures of fused UO_2 and micronized UO_2 , vibrationally compacted in 0.015-inch wall stainless steel tubes, were pressed (2100 F, 10 kpsi, 3 hours) to densities up to 96 percent TD. Cladding was distorted to a degree dependent upon particle-size distribution and initial density (Figure 5. 7).

To achieve high densities by hot isostatic pressing of mixtures of fused UO_2 and micronized UO_2 , approximately 30 wt % micronized UO_2 powder is required. To prevent gross distortion of the cladding, however, requires initial fuel densities of ~75 percent TD, and the maximum particle size in the fused- UO_2 component must be less than 20 mesh in rods having thin-wall (0.015-inch) cladding. Fuel mixtures that reconcile the need for relatively high initial density with the required high surface area and small particle size are, therefore, more suitable for precompaction by swaging than by vibrational compaction.

Cross sections of typical rods showing the effects of fabrication technique and fuel material are reproduced as Figures 5. 8 to 5. 14.

High Power Laser - L. E. Mills

Evaluation of a high power laser as a welding heat source is being sponsored off-site. Preliminary tests demonstrated capability to melt 0.01-inch diameter holes in thin stainless steel. The high energy laser beam that was optically focused on the stainless steel was generated in a 0.6-inch OD x 8-inch long ruby crystal, activated by a 200 kw power pulse of a few milliseconds duration. A mirrored surface was used on only one end of the ruby crystal to provide laser operation threshold at a lower input energy than had previously been required with a laser having a mirrored surface on each end.

Irradiation of Heterogeneously Enriched Fuel - G. R. Horn

Concern has often been expressed that inhomogeneity in mixed fuel, such as UO_2 - PuO_2 , might cause hot spots and burn-through of the cladding during irradiation. Tests were performed to evaluate this possibility. Continuing examination of a vibrationally compacted fuel element (GEH-4-62)⁽¹⁾ having severe preirradiation enrichment inhomogeneity in the UO_2 has provided additional evidence that a high rate of fuel homogenizing occurred during the early stages of irradiation.

Each 0.565-inch OD rod of the four-rod cluster initially contained one large particle (approximately 1/8-inch diameter) of 93 percent enriched UO_2 situated adjacent to the cladding near the fuel midpoint. The remaining fuel in each rod comprised natural UO_2 particles mechanically blended with 93 percent enriched UO_2 fines (-10 mesh) to achieve an average enrichment of 1.6 percent. The element was successfully irradiated for 16.6 equivalent full-power days (maximum surface heat flux, excluding hot spot, was 410,000 Btu/(hr)(ft²).

FIGURE 5. 7

Hot Isostatically Pressed Rods Fabricated by Vibrational Compaction
of UO_2 in 0.015-Inch Wall Stainless Steel

8X

100X

100X

FIGURE 5. 8

Micronized UO_2 , Hot Swaged to ~70 Percent TD and Hot Isostatically Pressed at 2100 F, 10 kpsi for 3 Hours. Final Density ~98 Percent TD, Inconel-X Cladding (Rod No. 1)

Reduced 30 % when printed.
REC-GE RICHLAND WASH

UNCLASSIFIED

8X

100X

100X

FIGURE 5. 9

Electrodeposited UO_2 , Vibrationally Compacted to 89 Percent TD
and Hot Isostatically Pressed at 1550 F, 10 kpsi for 4 Hours.

Final Density, 94 Percent TD, Zircaloy-4 Cladding (Rod No. 6)

Particle Size Distribution:

55 wt % -6 +10, 12.5 wt % -10 +20, 12.5 wt % -35 +65, 20 wt % -200

Reduced 30 % when printed.

ATC GE RICHLAND WASH

UNCLASSIFIED

8X

100X

100X

FIGURE 5. 10

Fused UO_2 (-20 Mesh) Cold Swaged to 88 Percent TD
and Hot Isostatically Pressed at 1550 F, 10 kpsi for 4 Hours.
Final Density, 91 Percent TD, Zircaloy-2 Cladding (Rod No. 8)

Reduced 30 % when printed.

AE. GE RICHLAND WASH

UNCLASSIFIED

8X

100X

100X

FIGURE 5. 11

Fused UO_2 (-20 Mesh) Hot Swaged to 92 Percent TD
and Hot Isostatically Pressed at 1550 F, 10 kpsi for 4 Hours.
Final Density, 94 Percent TD, Zircaloy-2 Cladding (Rod No. 18)

Reduced 30 % when printed.
AEC-GE RICHLAND, WASH.

UNCLASSIFIED

8X

100X

100X

FIGURE 5. 12

90 wt % Fused, 10 wt % Micronized UO_2 , Vibrationally Compacted to 81 Percent TD and Hot Isostatically Pressed at 2100 F, 10 kpsi for 3 hours
Final Density 92 Percent TD, 304 L Stainless Steel Cladding (Rod No. 25)
Particle Size Distribution: 70 wt % -10 +20, 20 wt % -65 +100

Reduced 30 % when printed.
AEC-GE RICHLAND WASH

UNCLASSIFIED

8X

100X

100X

FIGURE 5. 13

80 wt % Fused, 20 wt % Micronized UO_2
Vibrationally Compacted to 76 Percent TD
and Hot Isostatically Pressed at 2100 F, 10 kpsi for 3 Hours
Final Density, 91 Percent TD, 304 L Stainless Steel Cladding (Rod No. 28)
Particle Size Distribution: 65 wt % -10 +20, 15 wt % -65 +100

Reduced 30 % when printed.

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FIGURE 5. 14

60 wt % Fused (-6 +10), 40 wt % Micronized UO_2
Vibrationally Compacted to 64 Percent TD
and Hot Isostatically Pressed at 2100 F, 10 kpsi for 3 Hours
Final Density, 93 Percent TD, 304 L Stainless Steel Cladding (Rod No. 31)

Reduced 30% when printed.

Rod No. 2, (fused UO_2) was sectioned transversely near the particle of interest, then longitudinally through the axis of the rod. Photographs of the cross sections and an autoradiograph of the longitudinal cross sections are shown in Figure 5. 15. Dark spots in the autoradiograph indicates the location of U^{235} in the fuel. A photomosaic of the transverse cross section is shown in Figure 5. 16 (reduced from a 75 X original).

Rod No. 3 (sintered and crushed UO_2) was sectioned transversely near the subject particle. The cross section and its autoradiograph are shown in Figure 5. 17. A longitudinal cross section and autoradiograph of Rod No. 4 (high energy impact formed UO_2) is also shown in Figure 5. 18.

In general, the photographs show localized microstructural characteristics typical of UO_2 irradiated to produce high core temperatures; the primary differences are found in the shape and eccentric location of the heat affected zone. The autoradiographs show continued presence of discrete particles of enriched UO_2 in the cooler, outer portion of the fuel. However, the average concentration of U^{235} in the homogenized, columnar grain growth region is considerably greater than elsewhere, illustrating the spreading of highly enriched UO_2 into that region.

Irradiation of UO_2 - ThO_2 Fuel Mixture - G. R. Horn

Homogenizing of mixed fuels at high heat ratings is being studied to determine whether special precautions need to be taken to prevent segregation during fabrication of fuel elements containing mixtures. Ceramographic examination of an irradiated heterogeneous mixture of UO_2 and 10 wt % ThO_2 (GEH-4-61)⁽¹⁾ revealed structural characteristics similar to those observed in UO_2 irradiated under similar conditions. Extensive homogenizing occurred in the columnar grain region. Because of the extreme difficulty in etching this irradiated fuel mixture, the examination was performed on an as-polished, unetched cross section.

Irradiation of Swaged UO_2 - G. R. Horn

Swaged UO_2 irradiated to high exposures at central temperatures near the melting point sometimes develops structures different from those observed in UO_2 irradiated to low exposure at similar heat ratings. For example, examination of a swaged UO_2 capsule irradiated to a reported 37,000 MWD/ T_U (GEH-14-177) revealed that the radially oriented columnar grains are very long and extremely thin and that the voids are relatively more uniformly distributed throughout the fuel. Similar characteristics were observed in another capsule (GEH-3-52) irradiated to a reported 53,000 MWD/ T_U (mass spectrographic determinations of burnup are in progress). Cross sections of these two capsules are shown in Figure 5. 19. Fabrication and irradiation data are shown in Table 5. 4.

Section A-A

Longitudinal Section

Autoradiograph of Longitudinal Section

FIGURE 5. 15

Cross Sections of Heterogeneously Enriched UO_2
(GEH-4-62, Rod 2)

FIGURE 5. 16

Photomosaic of Heterogeneously Enriched UO_2
(GEH-4-62, Rod 2)

Longitudinal
Cross Section

Autoradiograph

Rod 3

FIGURE 5. 17

Cross Sections of Heterogeneously Enriched UO_2
(GEH-4-62)

Rod 4

FIGURE 5. 18

Cross Sections of Heterogeneously Enriched UO_2
(GEH-4-62)

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HW-72347

37,000 MWD/T_U
(GEH-14-177)

53,000 MWD/T_U
(GEH-3-52)

FIGURE 5. 19

Swaged UO₂ Irradiated to High Exposure

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TABLE 5.4
FABRICATION AND IRRADIATION DATA FOR HIGH EXPOSURE,
SWAGED UO₂ CAPSULES

	<u>GEH-14-177</u>	<u>GEH-3-52</u>
UO ₂ Form	Sintered and Crushed	Sintered and Crushed
Enrichment, wt % U ²³⁵ in uranium	2.42	2.44
Preirradiation Bulk Density	~ 85 % TD	~ 85 % TD
Preirradiation Particle Density	~ 95 % TD	~ 95 % TD
Cladding OD, inches	0.565	0.770
Cladding Thickness, inches	0.030	0.050
* Surface Heat Flux Btu/(hr)(ft ²)	600,000	400,000
* Exposure, MWD/T _U	37,000	53,000

* Based on reported thermal neutron flux values. Not yet confirmed by postirradiation burnup analyses.

Extensive ceramagraphic examination of GEH-14-177 revealed remarkable differences between the structural characteristics of different parts of the same fuel rods. Photomosaics of these cross sections are shown in Figure 5.20. These two cross sections were separated in the fuel rod by approximately 5/8-inch.

Fission gas release data for both high exposure fuel capsules are summarized in Table 5.5.

TABLE 5.5
FISSION GAS RELEASE DATA

	<u>GEH-14-177</u>	<u>GEH-3-52</u>
Total Gas Collected, ml (at STP)	19.2	190
Xenon Released, % of Total Formed	15 %	63 %
Krypton Released, % of Total Formed	8 %	33 %
Xe/Kr ratio	9.95	9.57

Fission gas release from the fuel in GEH-3-52 was sufficient to rupture the 0.050-inch thick Zircaloy cladding even though the capsule was contained in a shrink-fitted, thick wall aluminum can during irradiation.

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5. 30

HW-72347

FIGURE 5.20

Photomosaics of UO₂ Irradiated to 37,000 MWD/TU
These Two Cross Sections were ~5/8 inch Apart in the Fuel Capsule
(GEH-14-177)

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Thin-Wall Cladding Performance - J. J. Hauth, G. R. Horn and
M. K. Millhollen

An irradiation test of a nonfree-standing-clad element was completed in the Vallecitos Boiling Water Reactor (VBWR). The thin-wall stainless steel clad, vibrationally compacted UO_2 fuel element (HAPO-2) was discharged from the VBWR after an estimated exposure of 1200 MWD/ T_U at a peak surface heat flux of 280,000 Btu/(hr)(ft²). The 9-rod cluster comprised rods clad in 0.008-inch, 0.010-inch, and 0.015-inch walls of 304L stainless steel.

Preliminary examination in the VBWR basin revealed: (1) no bowing of any of the rods; (2) "handling marks" (scratches) on the outer rods; and, (3) several wrinkles and dimples near the ends of the four rods (minor deformation was visible on some rods before irradiation). The cluster will be returned to Hanford for extensive examination.

A cluster (GEH-4-71) comprising two rods clad in 0.009-inch thick AISI 406 stainless steel and two rods clad in 0.015-inch thick 304L stainless steel was irradiated in the MTR. The test ended because of intermittent, high gamma activity in the effluent coolant. The element operated for approximately 20 hours at an average surface heat flux of 400,000 Btu/(hr)(ft²) (determined calorimetrically) and a maximum of 520,000 Btu/(hr)(ft²). The element will be returned to Hanford for destructive examination.

PLUTONIUM RECYCLE PROGRAM

PRTR UO_2 Fuel Element Inspection - M. K. Millhollen

The PRTR is fueled with swaged UO_2 and spike-enrichment PU-Al fuel elements. Periodic evaluation of fuel element performance is combined with pressure tube inspections during refueling outages. Three UO_2 19-rod cluster fuel elements were discharged from the reactor and visually examined in the reactor storage basin. These elements included a cold swaged cluster with an exposure of 1700 MWD/ T_U , a vibrationally compacted cluster with an exposure of 330 MWD/ T_U , and a hot swaged cluster with an exposure of 300 MWD/ T_U . There was no evidence of wire-wrap loosening, crud accumulation, or corrosion on any of the three assemblies. All of the elements were recharged into the PRTR.

Vibrational Compaction Studies - J. J. Hauth, R. F. Klein, M. K. Millhollen, L. P. Murphy, and R. C. Smith

One product of the Plutonium Recycle Program is physics data on plutonium behavior in thermal reactors. Six PRTR Mark-I UO_2 fuel elements are being assembled using cobalt-zirconium alloy flux monitor wire spacing members for correlating the evolution of plutonium isotopes and fission products with fuel burnup. The fuel rods were made by vibrational compaction. Commercially fused UO_2 was crushed, and the (-4 +20) mesh fraction was vibrationally milled to eliminate porous aggregates. Particle densities, as determined by mercury porosimetry, were 10.6 to 10.8 g/cc, excluding pores having diameters greater than 100 microns.

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Average amounts of sorbed gases in two lots of fused oxide, before outgassing, were 0.045 and 0.081 cc/g. Carbon contents were 28 and 24 ppm, and the oxygen-uranium ratios were 1.988 and 2.004, respectively. After crushing and screening, the oxide was vacuum outgassed four hours at 750 C.

The fuel rods were fabricated using the resonant-beam technique (described in the previous Quarterly Report) illustrated in Figure 5. 21. Density of the compacted UO_2 varied from 87 to 89 percent TD, corresponding to compaction efficiencies of 89 to 91 percent. Further investigations are being performed to correlate the effects of vibrational milling of the fuel, top-coupling of the cladding, and dimensions of the resonant beam, in order to increase the compacted densities and process efficiency.

An unusually high rejection rate of the end closures was encountered during radiographic testing of the rods. An intensive review of the testing preparation and welding of the cladding tubes failed to disclose a cause of the faulty closures. However, rejection rates similar to that encountered in radiographic testing resulted from the autoclave test of both as-received tubes and assembled fuel rods. The cause was traced to surface contamination, probably by nitrogen, which apparently occurred during manufacture of the Zircaloy tubes. Both inside and outside surfaces of the tubes were affected, to a depth of 0.003-0.004 inch. (2)

Remote Fabrication Studies - R. C. Smith

Rapid development of the electrodeposition process for recovery and partial decontamination of irradiated oxide fuels provides additional incentive to develop remote fuel element fabrication technology. Construction of a room designed for remote fuel element fabrication studies was completed, and vibrational compaction equipment was installed. Vibrational energy is transmitted to the fuel rods through a 15-foot long steel beam to which the fuel rods are attached. As a performance test, fuel rods were satisfactorily compacted for seven Mark-I PRTR fuel elements for various physics experiments.

One-step fabrication of a complete fuel element by remote vibrational compaction is being evaluated. The results of initial experiments were encouraging. A completed Mark II-C, nested tubular PRTR fuel element (161 pounds), suspended from the remote-operated compaction-beam, was vibrated at frequencies between 200 and 3600 cps at accelerations (applied to the beam) as great as 60 grams. Disassembly and inspection of the element's components revealed only minor scuffing of cladding surfaces at nodal points. The pin that positions the center rod in the bottom end fitting was broken. Even though this allowed more rattling of the center rod, no appreciable damage occurred (a benefit may have occurred because no single point of contact was maintained).

FIGURE 5.21
Vibrational Compaction Using Resonant-Beam Coupling
of Fuel Cladding to Source of Vibration

The element was reassembled for further destructive testing at accelerations to 100 grams.

Equipment and facilities are being assembled for other remote fabrication studies. A precision speed-controlled turntable and a time-sequence welding power supply were ordered. Check prints on a controlled atmosphere welding box are being reviewed and modified before being issued for construction. Shielding systems for the remote enclosure were designed and a potential vendor contacted for a budget estimate. Preliminary cost analyses were received for other remote fabrication equipment.

Five sets of Mark II-C, PRTR nested tubular, outer cladding components were sent off-site to have ribs attached by electron beam welding.

Modified end hangers for remote welding studies were designed. Several hangers were built that complement the end closures completed last quarter and are ready for remote welding experiments. Remotized cathodic etching techniques were developed.

Approximately 150 pounds of electrodeposited UO_2 were prepared. Some of this material must be heat treated to remove volatile impurities before use in remote-control fabrication experiments.

Fuel Cladding Fabrication by Electron Beam Welding - R. F. Klein

High voltage electron beam welding can be used to weld a part to the inside of a tube by melting through the tube wall, or to tangentially weld a rib or one thin walled tube to another without filler metal. It allows unique shapes to be fabricated from simple part designs. For example, it provides a method of fabricating an internally cooled only (inverted cluster) fuel element. Also, it allows construction of an integral fuel element process tube assembly. Short sections of these two fuel element designs are being fabricated. They have not been fabricated by other means.

Spacer ribs on five sets of cladding for the eight-foot long nested tubular fuel elements are also being welded by this process.

Stud Welding of Small Diameter Wire - R. F. Klein

Special test elements for PRTR experiments required cobalt monitors to be situated within Zircaloy fuel rod spacer wires. A stud welding process was developed as part of the fabrication sequence.

Stud welding is a percussion joining process, in which the two pieces to be welded are brought to contact, usually by spring or pneumatic force, and capacitor-stored energy is concurrently discharged through the joint to produce fusion of the two parts. In the monitor wire fabrication process, a small cobalt pin is inserted into a small, axially drilled hole in the end of a 0.072-inch diameter, Zircaloy wire. After the hole is sealed by fusion welding, another wire is joined, end to end, to the first to form a continuous wire. The stud welding method has the advantage of low heat input except at the joint, and atmospheric contamination of the Zircaloy can be easily minimized by flooding the joint with inert gas.

Hydriding of Zircaloy - G. R. Horn

A continuing investigation of the causes and effects of hydriding of failed zirconium alloy, UO_2 fuel element cladding included several deliberately defected test elements irradiated in Hanford reactors. Examination of a third Hanford defect test element (HD-5)⁽¹⁾ revealed no appreciable quantity of zirconium hydride in the cladding of any of the rods. The element was irradiated for 8 days at a heat generation rate that was expected to cause catastrophic hydriding.

The four-rod cluster comprised two UO_2 fuel rods clad in Zircaloy-2 and two rods clad in low-nickel Zircaloy-2. One each of the Zircaloy-2 and low-nickel Zircaloy-2 clad rods was deliberately defected (0.006-inch diameter hole) at its midpoint. The defected Zircaloy-2 clad rod failed at a very low rod power during initial startup, probably because of water-logging, and all the fuel was subsequently washed out of the rod. Therefore, no hydriding occurred in the defected Zircaloy-2 cladding. As shown in Figure 5.22, only small quantities of hydride were observed in the cladding of the other three rods.

This test demonstrated that defected, low-nickel Zircaloy-2 became hydrided at no greater rate than nondefected low-nickel Zircaloy-2 or Zircaloy-2 during short-time irradiation at high heat flux. No comparison of the hydriding characteristics of defected Zircaloy-2 and low-nickel Zircaloy could be made because of the complete loss of fuel from the defected, Zircaloy-2 clad rod.

REFERENCES

1. Cadwell, J. J. Quarterly Progress Report-Fuels Development Operation-October, November, December, 1961, HW-72346. January 15, 1962.
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2. Haws, K. M. Personal Communication.

Nondefected Zircaloy-2

Nondefected Low Nickel Zircaloy-2

Defected Low-Nickel Zircaloy-2

FIGURE 5. 22

Zircaloy Cladding from a Third Hanford Defect Test, HD-5

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