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DEVELOPMENT OF TUBULAR UO_2 FUEL ELEMENTS FOR POWER REACTORS

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DEVELOPMENT OF TUBULAR UO_2 FUEL ELEMENTS FOR POWER REACTORS

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ABSTRACT

A process was developed for fabricating tubular fuel elements with compacted cores of fused-and-crushed UO_2 in Zircaloy sheaths. Final core densities of greater than 90% of theoretical were obtained by a combination of vibration plus cold swaging.

Irradiation of more than 135 tubes with compacted urania cores at test conditions that ranged up to 17,000 MWD/MTU peak exposure and 68 watts/cm peak thermal rating ($\int_{Ts}^{Tc} k d\theta$) indicated that such tubes made by the best available techniques should operate satisfactorily in a full-scale heavy-water-moderated power reactor to peak exposures of at least 20,000 MWD/MTU and peak thermal ratings of 30 watts/cm. Examination of 45 tubes irradiated in the Heavy Water Components Test Reactor indicated that:

1) Less than 10% of the fission gas produced during irradiation was released in fuel tubes operating at low $\int k d\theta$ (below about 25 watts/cm); onset of columnar grain growth in the UO_2 core occurred at about 25 watts/cm and coincided with a sharp rise in the fractional release of fission gas.

2) Release of nitrogen from impurities in fused UO_2 was substantially reduced by outgassing the UO_2 at high temperature in a vacuum or in hydrogen before irradiation.

3) The outer sheath wrinkled in tubes with relatively low-density cores (82-87% of theoretical density) when operated at $\int k d\theta$ of 40-45 watts/cm; tubes compacted to 90% of theoretical density did not wrinkle when irradiated at 50 watts/cm.

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INTRODUCTION

The development program on heavy-water-moderated power reactors sponsored by the U. S. Atomic Energy Commission had as its goal the formulation of a full-scale design upon which a valid estimate of power cost could be made. The Du Pont study was mainly directed toward heavy-water-moderated and-cooled reactors. This program was terminated by the AEC at the end of FY-1965 in favor of a heavy-water-moderated, organic-cooled reactor.

Tubular fuel elements of UO_2 received heavy emphasis in the Du Pont study because of the potentially low fabrication costs,[1] high melting point, resistance to attack by hot water, and relative dimensional stability under irradiation. UO_2 can be arc-fused and crushed to produce a powder of high particle density that can be compacted into an annulus between concentric tubes of Zircaloy to form tubes of high bulk density. Tubes produced by these techniques were irradiated to permit correlation of in-reactor performance with fabrication variables, and ultimately to determine the safe thermal rating and exposure limit of fuel elements.

This report summarizes the fabrication development and irradiation tests of compacted urania tubes and gives the status of the development when the Du Pont study was terminated. Irradiation of UO_2 tubes in the Heavy Water Components Test Reactor (HWCTR) at Savannah River is discussed in a companion report.[2]

SUMMARY

A process was developed to fabricate relatively massive tubes of UO_2 for service in power reactors. More than 300 tubes containing compacted powder cores were fabricated in sizes ranging from 1.7 to 3.7 inches in diameter and from 14 inches to 10 feet long. In this process, fused-and-crushed UO_2 was outgassed, sized and blended, loaded into the annulus between Zircaloy sheaths, and compacted by vibration to a density of about 83% of theoretical.

The "oxide" tubes were then dried, and swaged over a mandrel in two passes to at least 90% of theoretical density. The tubes were cut to desired length, and end plugs with void chambers, to accommodate gases released during irradiation, were welded to the sheath tubes.

More than 135 uranium dioxide tubes were irradiated at SRP in production reactors at low coolant pressures and sheath temperatures, and in the HWCTR at power reactor conditions. Test irradiations proceeded concurrently with fabrication development and, in several cases, led to modifications in the fabrication process. When the program was terminated, two assemblies had attained peak exposures of 4000 and 17,000 MWD/MTU* at average peak** ratings*** of 36 and 19 watts/cm, respectively. The most highly exposed elements in these assemblies were examined; results of the examination were interpreted to indicate that these test elements would have operated satisfactorily to the goal exposures of 20,000 and 30,000 MWD/MTU, respectively.

Examination of more than 45 fuel tubes irradiated under power reactor conditions and data from the irradiation of 10 other assemblies (not examined) indicated that:

1) In fuel tubes operating at low $\int k d\theta$, less than 10% of the fission gas produced during irradiation was released into the core voids.

2) The release of fission gas increased sharply at an $\int k d\theta$ of about 25-30 watts/cm, coincident with the onset of columnar grain growth in the UO_2 core.

---

* MWD/MTU - Megawatt days per metric ton of contained uranium (numerically equivalent to watt-days per gram of uranium).

** Average peak refers to the time-weighted average value at the axial location of the highest integrated exposure.

*** Thermal rating - $\int_{Ts}^{Tc} k d\theta$, an index of fuel temperature that is useful when precise values of the thermal conductivity are unknown.[3]

3) Columnar grain growth was extensive at 35 to 40 watts/cm, and central core melting occurred at about 60 watts/cm in swage-compacted tubes.

4) Release of gases from contaminants in fused UO_2, and consequent buildup of pressure in the fuel element, was substantially reduced by high-temperature vacuum outgassing or hydrogen treatment of the UO_2 before irradiation.

5) The tubes resisted wrinkling of the outer sheath by the high coolant pressure, even at thermal ratings of 50 watts/cm, when the bulk density of the UO_2 core was 90% or higher. The outer sheaths of four tubes with relatively low density cores (82-87%) wrinkled at thermal ratings of 40-45 watts/cm.

These results were interpreted to indicate that Zircaloy-sheathed UO_2 tubes fabricated by the best available techniques should operate satisfactorily in a full-scale heavy-water-moderated and cooled power reactor to peak exposures of at least 20,000 MWD/MTU and peak thermal ratings of 30 watts/cm.

FABRICATION DEVELOPMENT

REFERENCE PROCESS

A process was developed for fabricating Zircaloy-clad fuel tubes containing powdered UO_2 cores compacted by vibration and cold swaging. Fused-and-crushed UO_2 of selected particle sizes was loaded into the annulus between Zircaloy sheath tubes that had been etched and fitted with temporary end plugs, and was compacted by vibration to a density of about 83% of the theoretical value. After drying the oxide cores, the tubes were sealed and further densified by cold swaging over a steel mandrel to at least 90% of the theoretical value. The tubes were next cut to desired length, counterbored, and permanent end plugs with void chambers, to accommodate gases released during irradiation, were welded to the sheath tubes. After nondestructive testing, the tubes were etched

and autoclaved before final inspection. The fabrication steps are summarized in Figure 1 and presented in Appendix A in more detail.

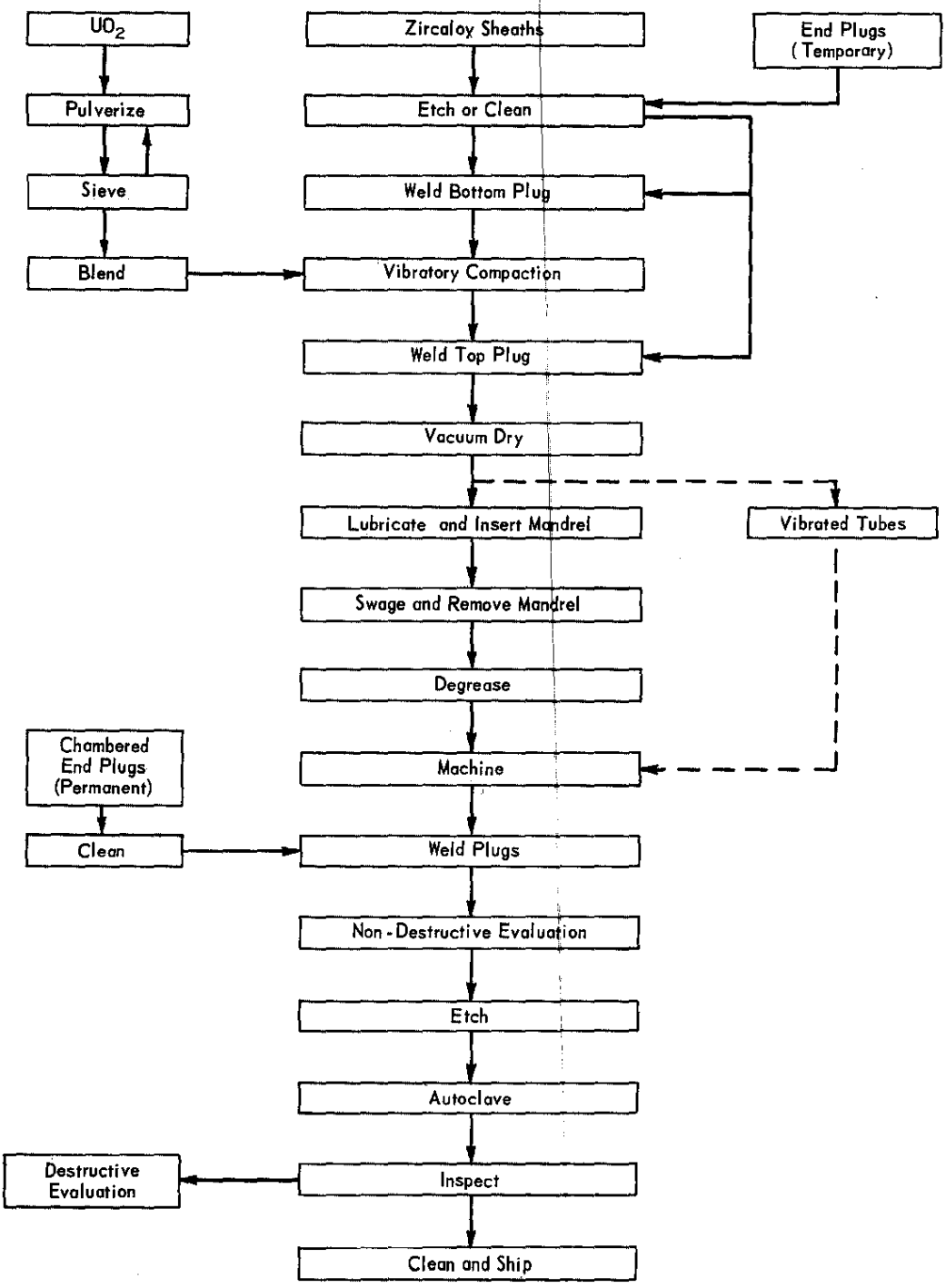
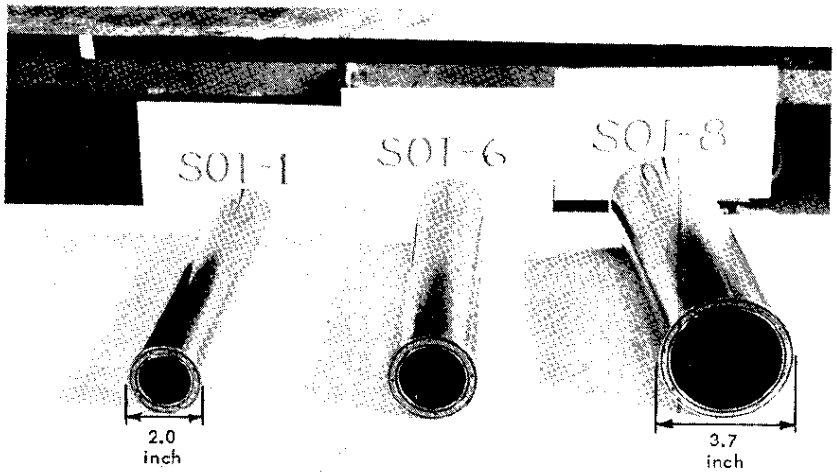


FIG. 1 FABRICATION PROCESS FOR UO_2 TUBES

More than 300 tubes of various sizes were manufactured. Diameters ranged from 1.7 to 3.7 inches, wall thicknesses ranged from 0.29 to 0.54 inches, and sheath thickness ranged from 0.023 to 0.038 inches. The tubes were compacted in lengths of 4 to 10 feet, and were then generally cut into smaller pieces, 14 to 25 inches long, to give added flexibility in testing by increasing the number of specimens that could be irradiated in a single column and to facilitate postirradiation examination. Typical tubes are shown in Figure 2.



NEG. 61099

FIG. 2 TYPICAL ZIRCALOY-CLAD UO_2 TUBES
Designations refer to irradiation tests of "base case" (SOT-1) and "reference size" (SOT-6, -8) tubes

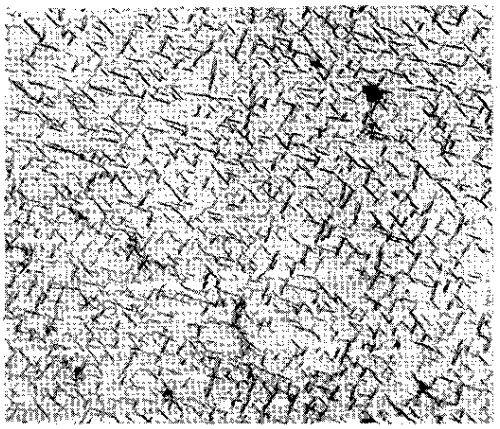
QUALITY OF FUSED UO_2

This section discusses the effect of impurities on the irradiation behavior of fused UO_2, and compares the effectiveness of several treatments used to reduce nitrogen impurity in the oxide.

Impurity Content

Impurities normally found in arc-fused UO_2 consisted of sorbed gases and microscopic inclusions.[4] The inclusions were identified by metallography as:

- A phase containing excess oxygen (U_4O_9, Figure 3a)
- Metallic uranium (Figure 3b)
- Uranium carbide (UC, Figure 3c)
- Uranium nitride (UN_2 or UN_{2-x}, Figure 3d)



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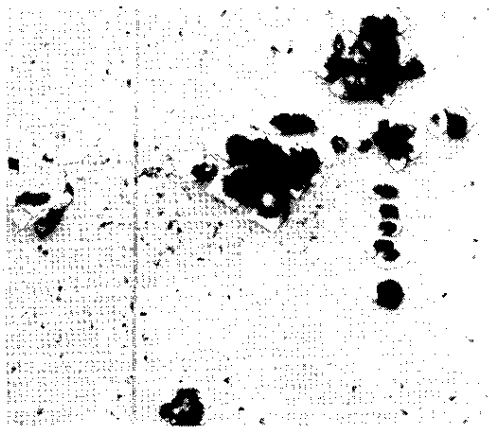
a. Excess Oxygen as U_4O_9



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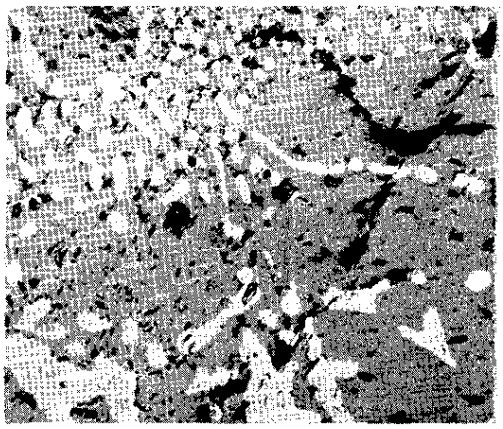
b. Excess Uranium as Metallic Uranium



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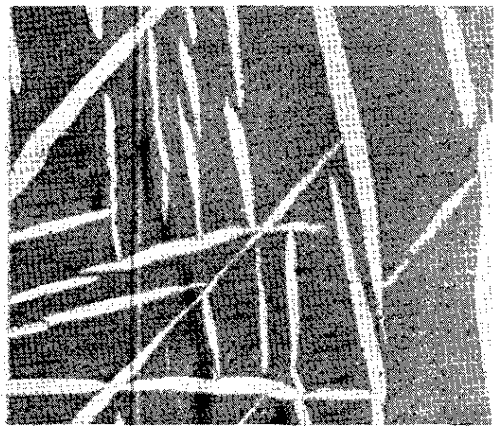
c. Uranium Carbide Inclusion



NEG. 32038 250X

d. Uranium Nitride Inclusions

FIG. 3 INCLUSIONS IN FUSED UO_2 CONTAINING EXCESS OXYGEN OR URANIUM

Considerable variation was found in the oxygen-to-uranium ratio (O/U) of fused UO_2 from different suppliers; early in the program, significant variations occurred from order to order from a given supplier (in some cases, even within a single order). Later, the suppliers were able to produce fused UO_2 with adequate control of the O/U ratio. Inclusions of metallic uranium, uranium carbide, and uranium nitride were generally associated with O/U ratios of 2.00 or below.

Gases were desorbed from fused oxide when it was heated in a vacuum or when it was irradiated. The principal gases released at 1000 and 1300°C in vacuum were hydrogen, nitrogen, carbon monoxide, and carbon dioxide. Studies of gases released from fused UO_2 were begun after unexpected amounts of N_2 and CO_2 were found in swage-compacted rods irradiated in the MTR.[5]

Effect of Impurities on Irradiation Performance

Some UO_2 tubes irradiated early in production reactors at Savannah River failed because hydrogen gas in the tubes reacted with and embrittled the Zircaloy sheaths (page 41).[6,7] Hydrogen was evolved in the tube from the reaction between absorbed moisture and impurities in the UO_2. In later stages of work, this difficulty was eliminated by controlling the moisture content of the cores; procedures for this technique are described on page 28.

Also, release of nitrogen gas from the decomposition of nitride inclusions is believed to have caused failure in an oxide tube by collapse of the inner sheath during irradiation in a Savannah River production reactor.[8] Cladding integrity was maintained, however, and the gas collected from this tube after irradiation was estimated to have generated a pressure of about 1100 psi in the tube during irradiation (page 41).

Control of Impurity Gases

Vacuum outgassing and hydrogen annealing at elevated temperatures removed gaseous impurities in the as-received oxide. Both treatments are effective in reducing higher nitrides by either thermal decomposition or formation of ammonia.[9]

Most of the oxides were outgassed in a large vacuum furnace. Charges ranging from 50 to 80 pounds were loaded to a depth of about 8 inches in crucibles of tungsten, tantalum, columbium, or graphite lined with columbium, and heated under vacuum by induction for 1 to 4 hours. Temperatures in the oxide bed ranged from 1000 to 1600°C. The surface temperature of the oxide bed was controlled at 1000-1200°C by adjusting the furnace power; oxide temperatures at the surface and center of the bed were measured with an optical pyrometer and with W and W-Re thermocouples, respectively.

Less nitrogen was released during irradiation of fused UO_2 that had been annealed in hydrogen before irradiation than from comparable UO_2 that had been vacuum outgassed, as shown in Table I for oxide tubes irradiated in the HWCTR. The results indicated that nitrogen releases were reduced to acceptably low levels whenever the treatment consisted of heating the oxide to 1300°C or higher for several hours. However, outgassing in vacuum (and presumably annealing in hydrogen) for less than 1 hour at 1000-1100°C is much less effective than the higher temperature treatments and can lead to excessive amounts of nitrogen being released during irradiation.

TABLE I

Free Nitrogen Gas in Irradiated UO_2 Tubes

Pretreatment of Fused UO_2[a]		Number of Tubes	Peak $\int kd\theta$, watts/cm		Free Nitrogen Released During Irradiation, cc at STP/kg of UO_2	
			Mean	Range	Mean	Range
Vacuum Outgassed						
Time, hr	Avg Temp, °C					
3/4-1	1000-1100	11	19	11-25	55	0.2-101
4	1300-1400	10	29	21-39	15	5-25
4	1500-1600	18	31	4-55	6	0.2-23
Hydrogen Treated						
4	1300	1	52	-	1.2	-
12	1750	2	38	32-45	0.14	0.13-0.16
Hypostoichiometric ($UO_{1.98}$)						
No thermal treatment		3	39	18-54	0.04	0.03-0.05

(a) Oxygen-to-uranium ratio (O/U) = 2.00 to 2.02 unless otherwise shown.

Hypostoichiometric UO_2 may offer further control on the buildup of nitrogen gas in an oxide element during irradiation. Tubes fabricated with UO_2 that was not outgassed but which had an O/U ratio of about 1.98 contained less free nitrogen after irradiation than tubes fabricated with stoichiometric UO_2 that was outgassed by thermal treatments. The nitrogen content was essentially the same as the as-received UO_2, whether stoichiometric or hypostoichiometric, and ranged up to about 240 ppm.

Metallic inclusions in hypostoichiometric UO_2 may act as a getter to control the buildup of nitrogen gas during irradiation and at shutdown. Combined nitrogen present in the hottest part of the element is probably released during irradiation but may then combine with metallic uranium in the cooler parts of the element. Laboratory tests established that all metallic uranium inclusions in particles of hypostoichiometric UO_2 were transformed into uranium nitride after heating in nitrogen gas for 4 hours between 700 and 1300°C.

Before irradiation, nitrogen in the oxide was measured by two methods: 1) a modified Kjeldahl method, and 2) vacuum extraction of gases from fine particles (-325 mesh) heated to 1300°C. Nitrogen values by the Kjeldahl method (which measures all combined nitrogen) were consistently higher than those determined by vacuum extraction or found as free gas in irradiated tubes (Appendix B). Because Kjeldahl procedure was not available until late in the program, nitrogen release in irradiated tubes could not be correlated. Nitrogen determined by vacuum extraction, however, could be correlated with free nitrogen found in irradiated tubes. Whenever the nitrogen in the unirradiated UO_2 was 50 ppm or less, the free nitrogen after irradiation at thermal ratings up to 50 watts/cm was less than 25 cc/kg UO_2 (equivalent to 20 ppm in UO_2). However, when the nitrogen content of the UO_2 exceeded 50 ppm, the amount of free nitrogen in irradiated tubes made with this oxide was unpredictable; some exceeded 125 ppm (100 cc/kg).

Status

Thermal treatments of stoichiometric or hypostoichiometric UO_2 are recommended for irradiations at thermal ratings above 30 watts/cm, e.g., vacuum outgassing above 1300°C for 4 hours or hydrogen annealing at 1750°C for 12 hours. Without such a treatment, the size of void space required to accommodate impurity gases released from fused UO_2 during irradiation may be excessive.

Additional development is needed before outgassing conditions can be defined that will reduce impurity nitrogen in fused UO_2 to a specific level. Late in the program, approximately 3800 pounds of fused oxide was obtained, for which the vendor was permitted to use either of the following treatments to reduce nitrogen content to less than 50 ppm: a) hydrogen annealing at 1700°C for 4 hours, or b) vacuum outgassing at 1400°C for 4 hours. The vendor experienced difficulty in meeting the specification reproducibly with either method; one fusion lot contained nitrogen in excess of 100 ppm after treatment by both techniques.

FABRICATION TECHNIQUES

Process development for the major fabrication steps are discussed in the following sections.

Vibratory Loading

Vibratory loading was investigated as a method of (1) decreasing the required amount of swaging reduction by increasing the packed density before swaging and (2) producing by vibrational compaction alone, a tube with the highest possible compacted density. Before the introduction of vibratory loading and using only tamping, many swaging passes were required for adequate compaction. Tubes clad with stainless steel were swaged in up to 13 passes,[10] and tubes clad with Zircaloy cladding were swaged in up to 7 passes.[11]

As-loaded densities, obtained with vibration of particle blends suitable for swaging, were sufficiently high for final compaction to 91% density in one or two swaging passes. Densi-

ties achieved by vibration alone ranged up to 87% of theoretical when the powder blend was optimized for vibratory compaction. Results of irradiation tests in the HWCTR indicate, however, that densities of 90% or greater are required at thermal ratings that produce appreciable grain growth (page 52).

The major factors that affected the maximum density achieved by vibration were powder characteristics (e.g., particle size, distribution, shape, friability) and procedural variables (e.g., equipment, frequency, acceleration, time). Other factors such as segregation of particles by size and shape, geometry of the container, and column height also influenced the final density but were not investigated.

Equipment

Electromagnetic vibrators were used to study loading and compacting because they offered greater flexibility and control of the operating parameters than did pneumatic impactors. An MB Electronics Model C10E vibration exciter driven by a Model T151M amplifier[12] was used for initial development. The vibrator had a peak thrust output of 1200 pounds and a frequency range of 5 to 2700 cps; the amplifier was rated for a peak output of 2.9 kw. The vibrator was mounted in a pit beneath a glove box for the containment of UO_2. The cabinet was designed so that the tubes or rods up to 12 feet long could be vibrated. A schematic diagram of the apparatus is shown in Figure 4.

Later in the program, vibratory equipment with greater capacity was installed for compacting larger diameter tubes. This equipment was an MB Electronics Model C125 vibration exciter, driven by a Model T888-3 amplifier. Rated capacity for the vibrator was a peak thrust output of 7800 pounds, and the amplifier had a peak output of 72 kilowatts. A sawtooth wave was obtained reproducibly using a signal generator with an automatic sweep frequency control. In early work this input wave form was shown to produce maximum density in a shorter time than a sine wave.[12] The vibration exciter was mounted in a pit beneath a semienclosed glove box for containing oxide while the tube was being loaded. Adjoining the vibrator was a pit for suspending a tube from the beam, as

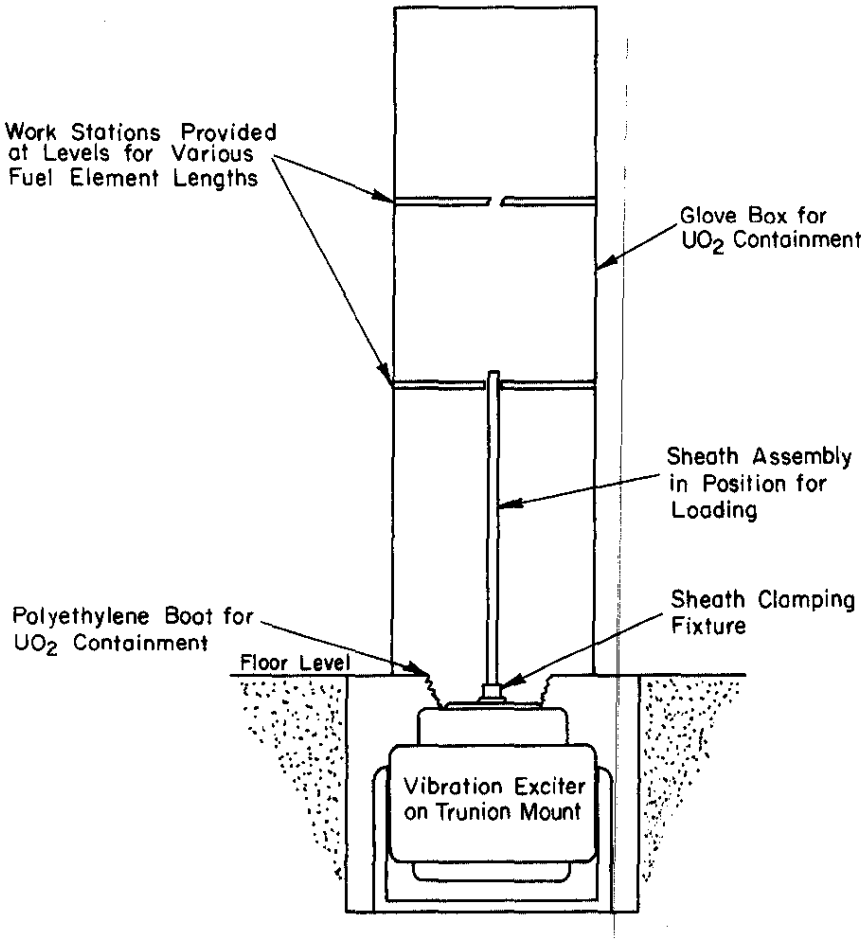


FIG. 4 SMALL FACILITY FOR VIBRATORY COMPACTION

shown in Figure 5; however, the program ended before beam excitation was attempted.

Particle Size Distribution

The maximum density that could be reproducibly obtained in annular tubes by vibratory compaction was 85 to 87% of theoretical maximum (TD) -- obtained with the following blend of particle sizes:

Particle Size Range, U. S. Standard Sieve	Amount in Blend, Weight Percent
-6, +10	52
-40, +70	28
-325	20

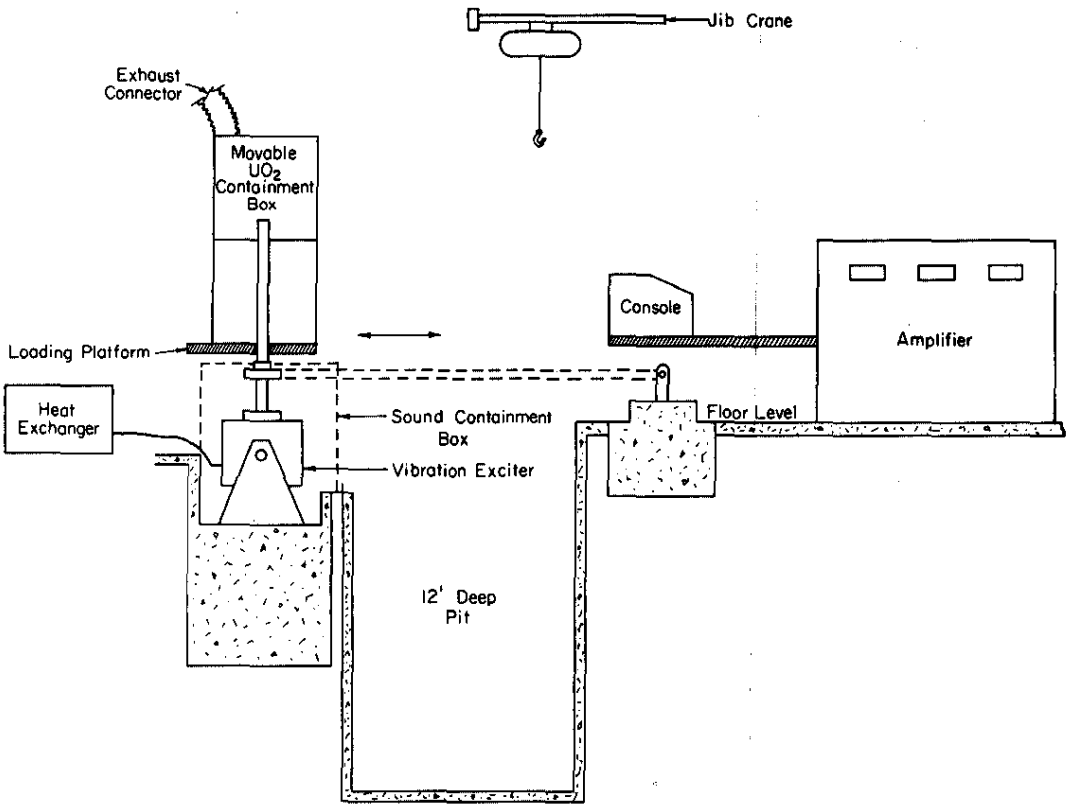


FIG. 5 LARGE FACILITY FOR VIBRATORY COMPACTION

This blend was a variation of a calculated composition that had been derived using the semiempirical treatment of Furnas.[13] The intermediate particle size satisfies the relationship

$$\frac{d_1}{d_2} = \frac{d_2}{d_3}$$

where: d_1 = diameter of coarse fraction
d_2 = diameter of intermediate fraction
d_3 = diameter of fine fraction

Increasing the weight ratio of coarse particles to 60% lowered the final density, in contrast to Hanford results.[14] Decreasing the size of intermediate fractions to -60, +100, and increasing the fine fraction size to -200 (containing, typically, 70 to 80% of -325 mesh) produced the same oxide density (86%), but required higher acceleration levels.

Compaction was greatest when the maximum diameter of the coarse particles was about 1/3 of the annulus thickness. Maximum densities were 2-4% less, however, with similar blends when the diameter of the course fraction was 1/2 the annulus thickness.

A blend of finer particles (-16/+20 ∿52%, -60/+100 ∿28%, -200 ∿20%) was used in tubes that were swaged after loading to reduce the penetration of oxide particles into the sheath during swaging. With this blend, the deepest indentations caused by oxide particles (0.004 inch) were about half the depth of those formed from the coarser blend. Maximum densities with this fine particle blend were about 4% lower (81-83% TD) than attained with the coarser particle blend.

Loading Procedures

Two loading techniques were used that gave essentially equivalent compaction densities for similar blends. During initial development, the full charge weight of UO_2 was poured into an overlong tube and then compacted; later, an incremental amount equivalent to one foot of tube length was charged and lightly vibrated (200 cps, 5 to 10 g) before the next incremental charge. For tubes longer than 4 feet, oxide was poured through a spacer to maintain sheath concentricity. The final step was to vibrate with maximum power (2.9 kw) while manually sweeping the frequency from 300 to 1500 cps. When the frequency was maintained at a resonant value, the oxide became unsettled; the resulting lower density could not be subsequently increased by further sweeping.

The variation in density of the compacted oxide with different accelerations measured on the small vibrator indicated that, in large oxide tubes, densities might be increased with a larger and more powerful vibrator. Densities of blends of similar particle sizes compacted at successively higher accelerations in the same sheath assembly, are shown in Figure 6. The increase of density with accelearation was most marked below 25 g, and demonstrates that high accelerations are needed for highest packing efficiencies.

Investigations of loading variables during compaction showed that annular thickness, tube length, sheath straightness, and type

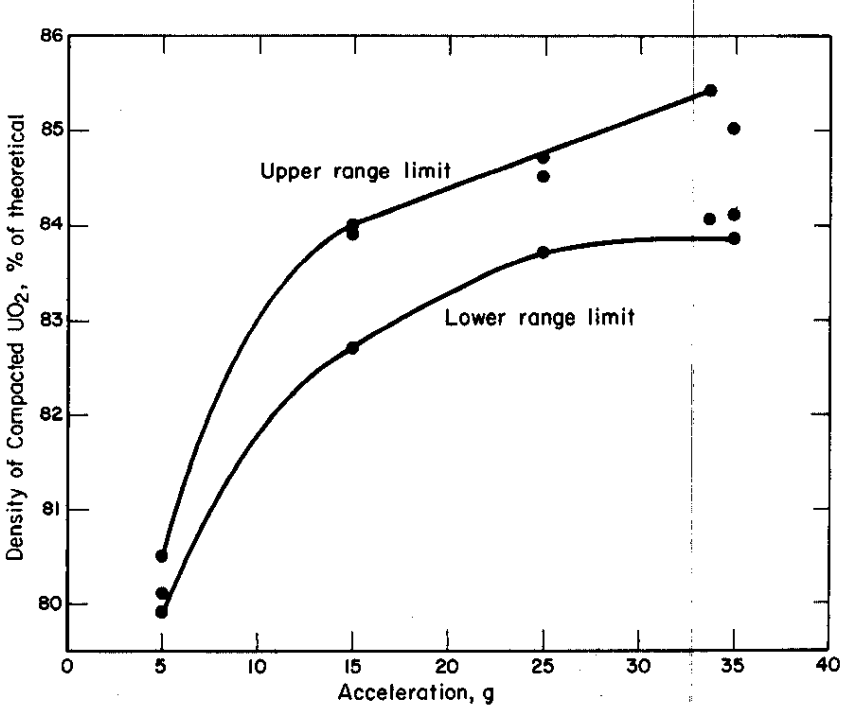


FIG. 6 EFFECT OF ACCELERATION ON COMPACTED DENSITY
(1200 lb thrust vibrator)

of spacing guide have a pronounced effect on concentricity of the inner and outer sheaths. Ten-foot-long tubes with a ≤1/4-inch-thick annulus were difficult to load within a 5% variation, although the same relative variation in concentricity was easily maintained in five-foot lengths of thicker tubes.

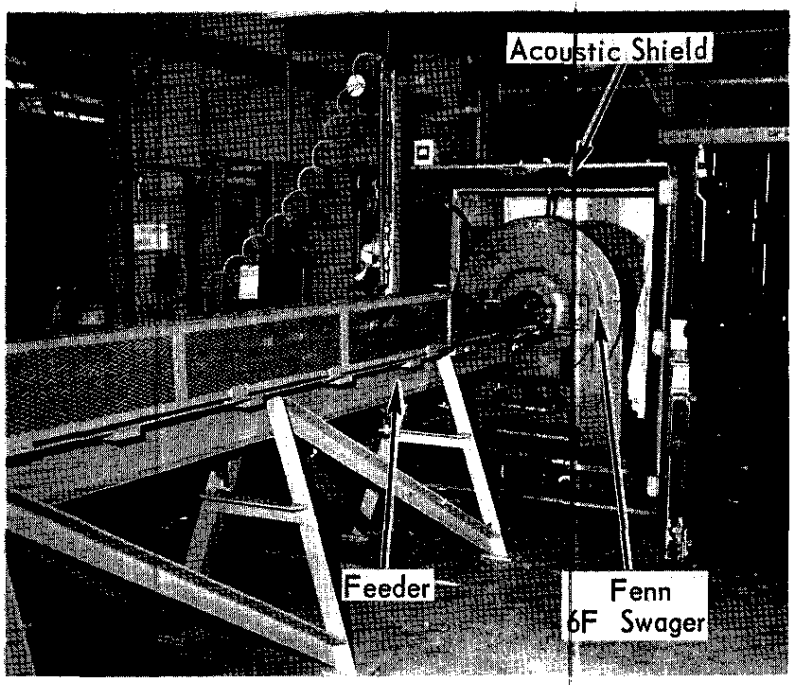
Swage Compaction

Cold swaging provides a relatively easy way of compacting UO_2 tubes to the densities desired for good irradiation performance (>90% TD). Also, cold swaging provides significantly better control of tube diameters than is otherwise available with commercially fabricated tubing, particularly in the larger diameters. In tubes loaded by vibration to 81-83% TD, final densities were 91-92% TD after two swaging passes (85-87% was the highest with vibration alone).

Zircaloy does not have the extended work-hardening capacity of stainless steel and cannot be swaged as severely without cracking. No cracking occurred, however, when annealed, fine-grained Zircaloy was swaged to moderate reductions (less than 10% in sheath cross section) in two or three passes using tubing fabricated by extrusion and drawing or extrusion and tube-reducing.

Equipment and Procedure

All tubes were swaged over a steel mandrel in a Fenn Model 6F, four-die rotary machine. A hydraulic feeder, Figure 7, pushed



NEG. DPSTF-1-9087

FIG. 7 SWAGER AND HYDRAULIC FEEDER

the oxide tubes through the swager while restricting tube rotation to less than that of the swaging dies (four fins would form in the outer sheath if the tube were allowed to rotate at the same speed as the swaging dies). Mandrels for supporting the inner sheath during swaging were made of SAE-4140 steel, hardened to Rc 45-48, ground, and polished to a surface finish of 10-16 microinches rms. The procedure for swaging was as follows:

- Mandrel and bore of the tube were lubricated with "Klingfast"*, and the mandrel was inserted into the tube.
- Other surfaces (tube OD, die faces) were lubricated with either coconut palm oil or motor oil, SAE grade 30.
- The tube was swaged at a feed rate of 24 to 30 inches per minute.
- After swaging, the mandrel was pulled from the tube on a drawbench; an estimated force of about 1 ton was required to pull the largest mandrel from a 10-foot tube.

Swaging Behavior of Tubes

Swaging in 2, 3, and 7 passes increased the packed density of the core and elongated the tube as shown in Figure 8. Densities >90% TD were obtained at reductions in core cross sections of 15 to 25% in oxide tubes; the greatest reductions were required for tubes with lowest initial density. Tubes that were loaded by vibration to 75% TD required a 25% swaging reduction to reach maximum density; those at 82% TD required only a 15% reduction.

The tube elongated during swaging, and the rate increased sharply as the density approached 89-90% TD; beyond 90%, additional swaging reductions were primarily reflected in elongation of the tube. The thickness of the inner cladding decreased at a rate commensurate with the elongation; thinning of the outer sheath was somewhat less because of diametral reductions during swaging.

Dimensional Control

Diametral control was improved by swaging. Diametral variations in tubes compacted by vibration alone was the same as that in the as-fabricated sheath tubing and ranged from ±0.008 inch for two-inch-diameter tubes to ±0.030 inch for 3.8-inch tubes. By comparison, the diametral variation in swaged 2-inch-diameter tubes was about ±0.005 inch (Appendix C).

Diametral variations in swaged tubes increased with tube size and the greatest variation occurred in the inner diameter. Outer

*Trademark of Brooks Oil Company, Cleveland Ohio for an asphalt base lubricant for gear reducers.

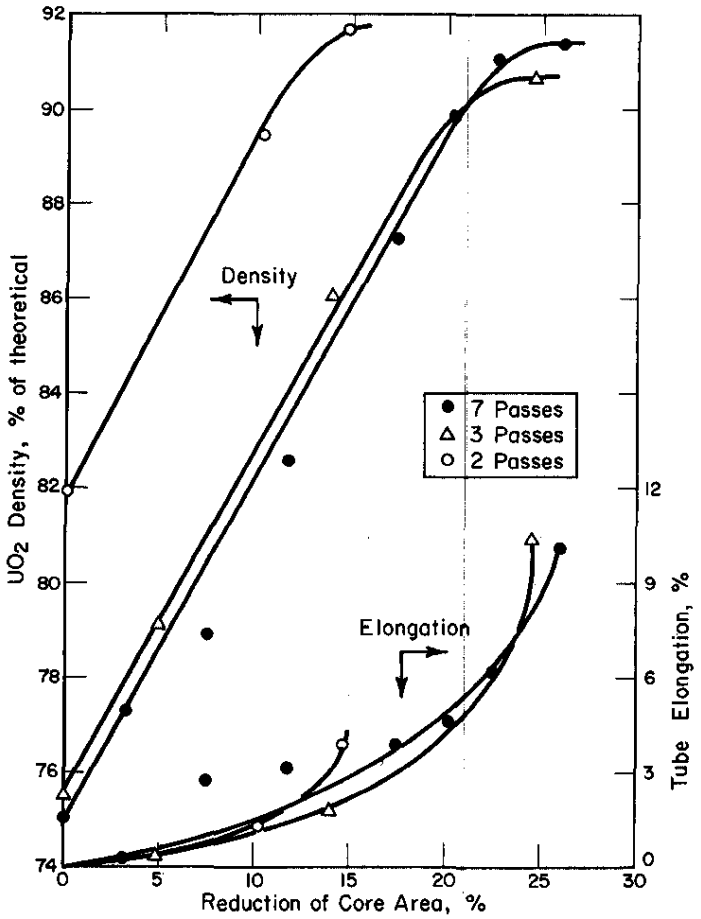


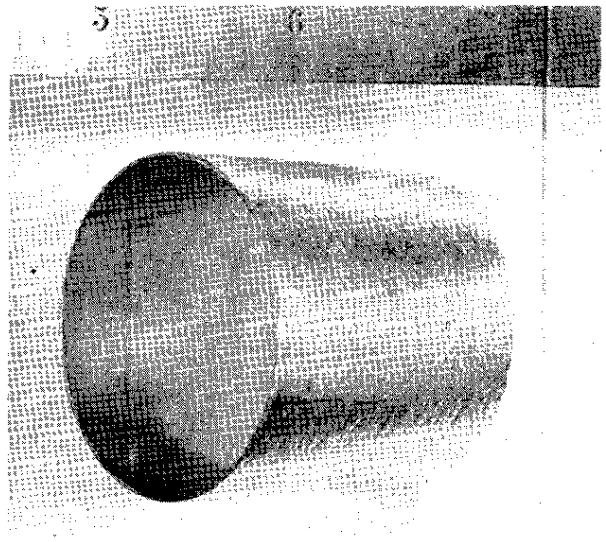
FIG. 8 SWAGING BEHAVIOR OF UO_2 TUBES
(All tubes swaged to about 2.25 inches O.D.)

diameters of 2.54- and 3.65-inch tubes varied ±0.005 and ±0.008 inch; the inner diameters varied ±0.004 and ±0.012 inch, respectively. The greater variation of the inner diameters in the larger tube sizes was probably caused by increased clearance between the swaging mandrel and inner sheath tube and a thicker film of lubricant.

Wall thickness variations present in loaded tubes were not significantly changed by swaging. Measurements with a magnetic reluctance instrument[15] on tubes before and after swaging showed little, if any, change in the concentricity of the sheaths.

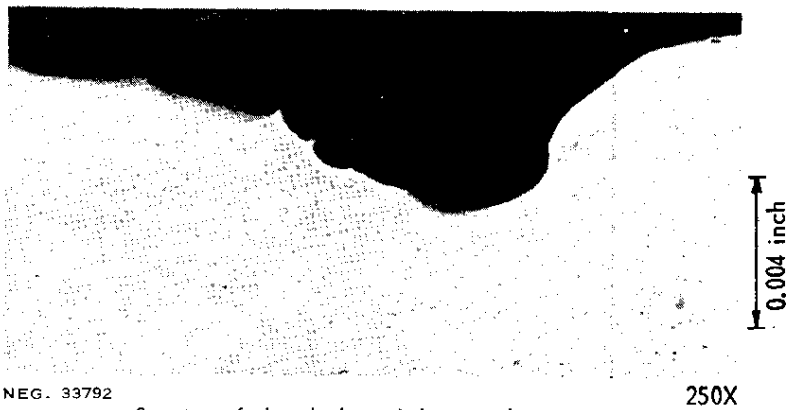
Surface Condition

Although the external surfaces of swaged tubes were usually smooth and burnished, the internal surfaces of the cladding were indented by oxide particles. The typical appearance of indentations are shown in Figure 9. The average and maximum depth



NEG. 53527 1X

Surface appearance



NEG. 33792 250X

Section of sheath through large indentation

FIG. 9 ROUGHENED SURFACE OF ZIRCALOY INNER SHEATH
(Indented by UO_2 particles during swaging)

were less than 0.001 inch and 0.004 inch, respectively, for oxide particles passing a 16-mesh sieve (specified for the reference process). Larger particles (-5 mesh) caused indentations up to 0.009 inch.

Vacuum Drying

Pulverized UO_2 has a strong affinity for moisture; both heating and evacuation are required to effectively remove the moisture.[16] Loosely adsorbed water is removed in vacuum at 25°C, but heating to above 300°C is required to remove the remaining water (Figure 10).[17]

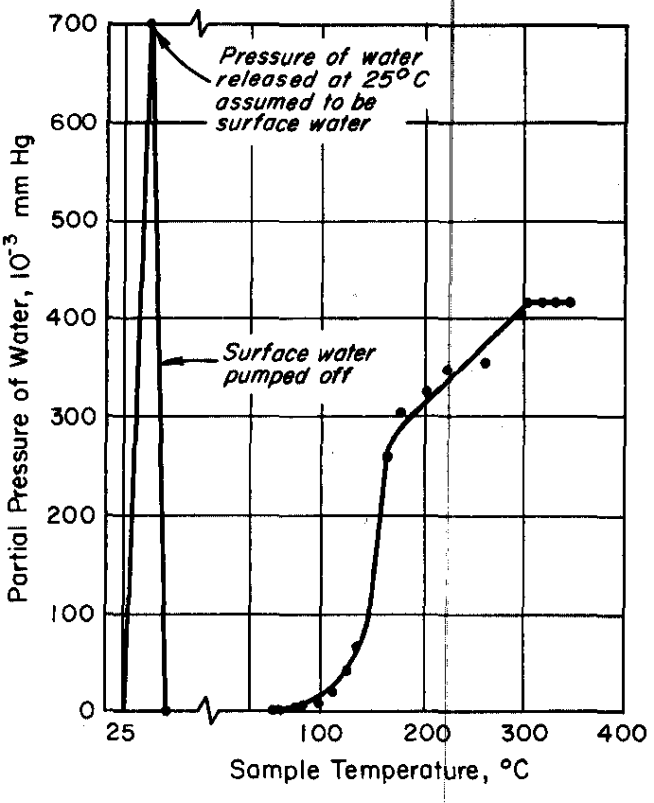


FIG. 10 DESORPTION OF WATER UNDER VACUUM FROM CRUSHED-AND-GROUND FUSED UO_2
(Water released from a five-gram specimen into a volume of 135 cc)

Early in the irradiation program, several in-pile failures of UO_2 tubes were caused by moisture in the oxide core. The fabrication process was subsequently modified to include drying the UO_2.[18] Compacted tubes were dried under vacuum at 250° to

300°C for 20 hours through two vent holes provided in each end plug. After drying, the tubes were backfilled with helium at 1 atmosphere, and the vent holes were sealed.

In the reference process, tubes were dried prior to swaging because of faster permeation rates in the as-loaded core. Pump-down rates for a 54-inch-long vibrated tube and a 22-inch-long swaged tube are compared in Figure 11. The internal pressure in the vibrated tubes was reduced to about 75 mm Hg after 5 hours of evacuation from one end of the tube; about 40 hours of evacuation were required for the swaged tube to reach the same internal pressure. A 20-hour evacuation cycle was selected for the reference process as one that was conservative and convenient. Moisture contents of the compacted UO_2 decreased with increasing drying temperature as shown in Table II. In the reference process, the maximum temperature for drying was restricted to 275°C because the Zircaloy surfaces oxidized at higher temperatures.

TABLE II

Effect of Drying Temperature on Moisture Content in UO_2 Tubes

Drying Temp, °C[a]	No. of Tubes	Moisture Content, ppm of UO_2[b]	
		Average	Range
200	4	32	16 - 56
250	5	14	3 - 54
275	4	6	1 - 7

a Under vacuum for 20 hours.
b Measured with electrolytic moisture analyzer.[17]

Welding

End closures were made with TIG welds in a closed chamber under a slight overpressure of helium. Oxide tubes were transferred into the welding box from an adjoining chamber (Figure 12), which could be evacuated and backfilled with helium. The tube was positioned during welding in a special rotary chuck, and welded with an argon-cooled torch. Trial welds on scrap Zircaloy tubing were performed before each closure was made;

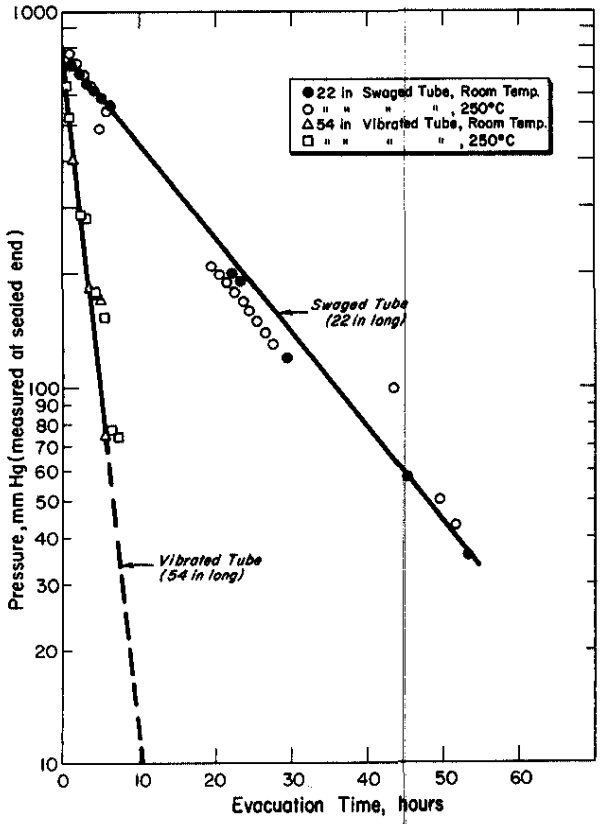


FIG. 11 PUMPDOWN RATES IN COMPACTED UO_2 TUBES
(Evacuation from one end only)

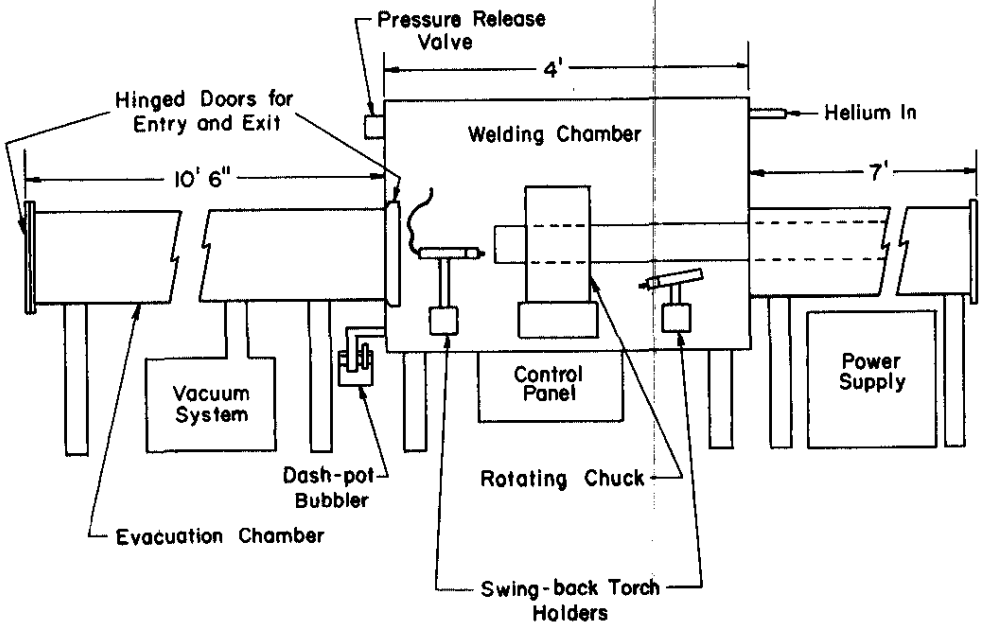


FIG. 12 SCHEMATIC DIAGRAM OF WELDING FACILITY
(Face of welding chamber removed to show equipment)

if the trial weld was bright or strawcolored, the atmosphere was considered acceptable. Welding power was supplied by a Harnischfeger (P and H) 300-ampere machine with an automatic timing cycle; less than 40 amperes of direct current was required for welding closures.

Usually the weld closure was made by fusing the edges of the Zircaloy end plug to an equal thickness of Zircaloy sheath (Figure 13). Weld penetrations were generally equal to the cladding thickness. Gaps of 0.010 inch or greater between end plug and sheath reduced penetration and caused undesirable weld overhang; chill rings and expanding plugs were used later in the program to assure that components would fit well.

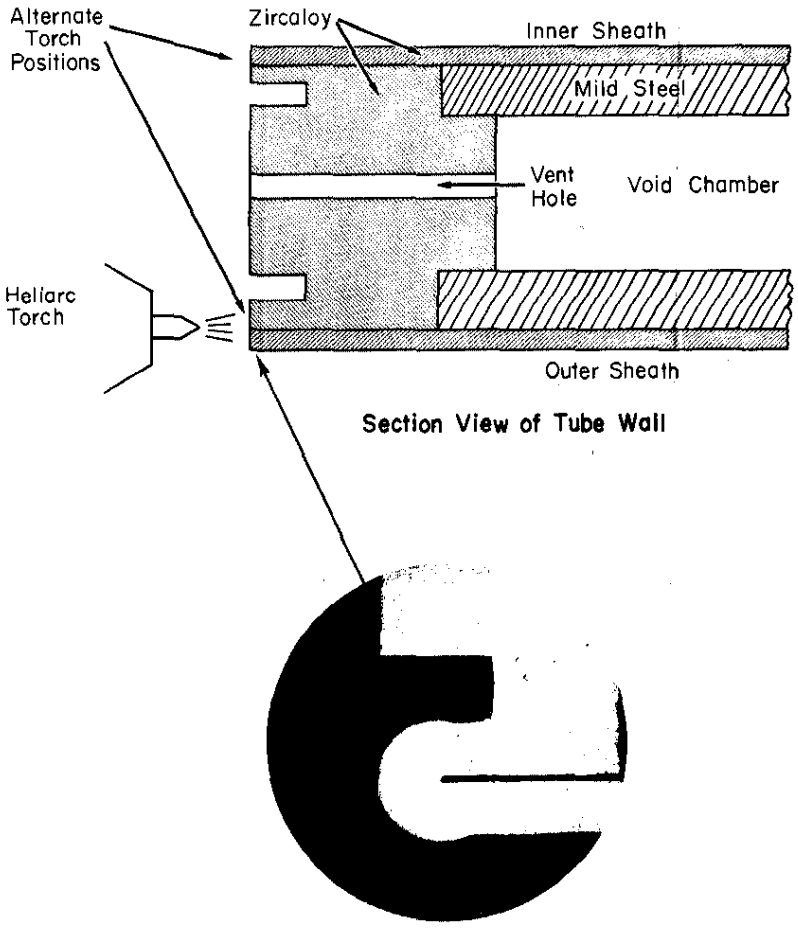


FIG. 13 WELD DESIGN AND TYPICAL WELDMENT FOR UO_2 TUBES
(Weld throat equal to cladding thickness)

Etching and Autoclaving

Zircaloy sheath tubing and the compacted tubes were etched in nitric-hydrofluoric acid solution in a conventional manner except that, when etching long tubes, part of the etching solution was pumped through the bore of the tube. For shorter tubes (5 feet or less) frequent longitudinal movement of the work piece through the solution gave the desired uniformity of etch. Faint gray marks coincident with swaging die marks were sometimes visible on the surface of the tube after autoclaving unless at least 0.0015 inch of metal was removed during etching.

The fuel tubes were autoclaved for 48 hours in 750°F steam (1500 to 1750 psi) to test the corrosion resistance of the Zircaloy sheath.

End Plug Design

End plug design in UO_2 tubes was complicated because void space was needed to accommodate gases released by the oxide core during irradiation. The first design proved unsatisfactory because the carbon steel septum became too hot, yielded during irradiation, and allowed uranium dioxide to flow into the chamber.[19] This weakness was eliminated in the standard design shown in Figure 14; these plugs performed satisfactorily during irradiation.

However, chambers of this general design were difficult to install in large-diameter tubes with thin cores, and an improved design shown in Figure 14 was developed that offers the following advantages:

- Improved fit of chamber and core (little counterboring of the compacted core is required)
- Extensive accommodation of tube eccentricity
- Reduced weld overhang (without the use of chill rings)
- Easier attachment of spacers (short radial spacers can be welded to the end plug before it is welded to the fuel tube)

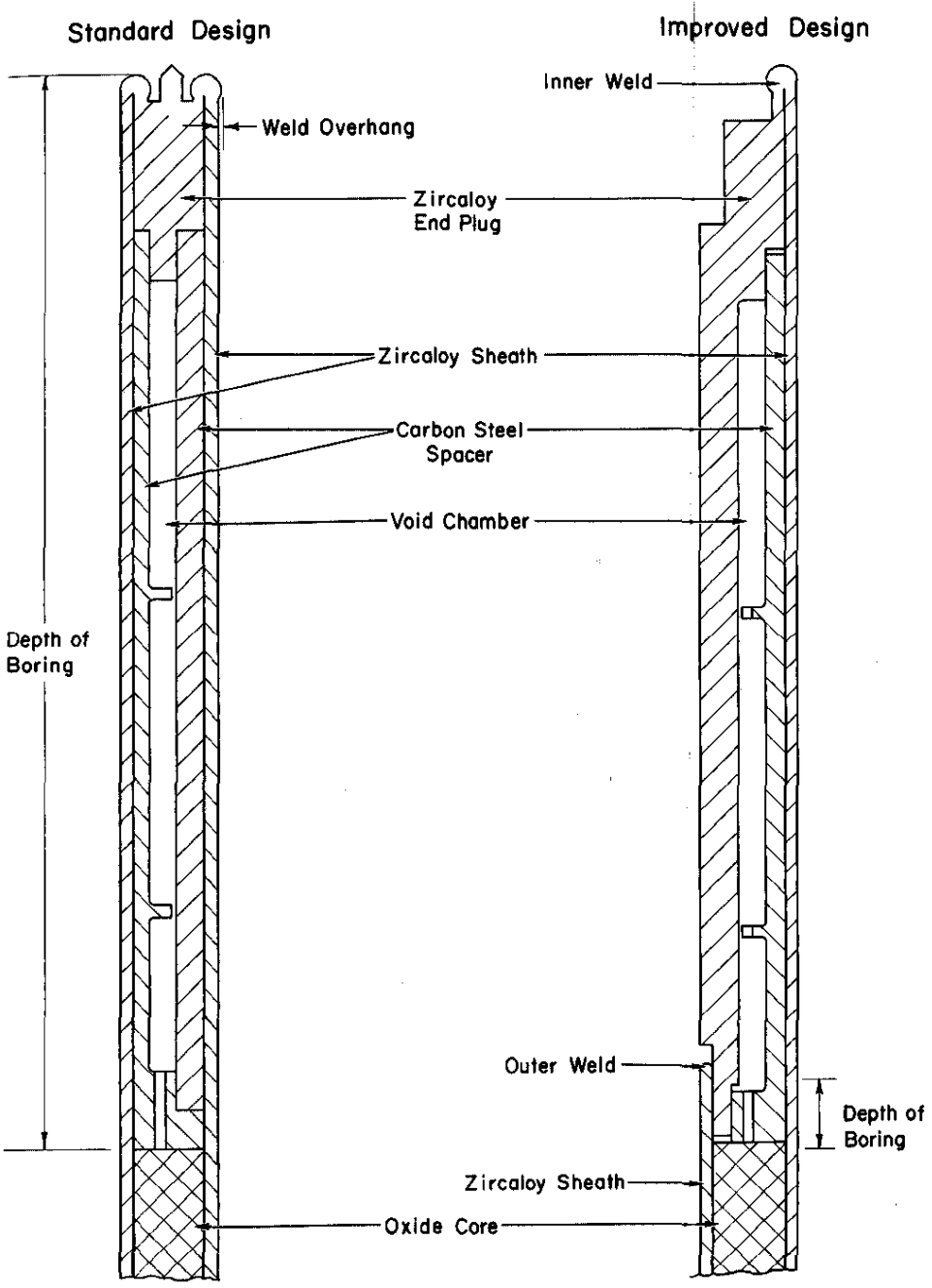


FIG. 14 COMPARISON OF END PLUG DESIGNS FOR UO_2 TUBES

The program ended before oxide tubes with the new end plug were irradiated; however, adequate resistance to collapse was demonstrated in an autoclave test in which two tubes with the improved end plug were successfully tested in 400°F water for 72 hours at 2200 psi followed by 48 hours at 2550 psi.

Inspection and Testing

Compacted tubes were inspected for cladding integrity and tube quality as follows:

Nondestructive Tests

Test	Purpose and Conditions
Radiography of welds	Detect 0.004 inch voids or inclusions; 86-95 kv, 5 ma, 8-16 sec, 6 radiographs per weld
Helium leak test	Detect minute leaks; mass spectrometer-detector, $<1 \times 10^{-9}$ STD cc/sec
Bubble test	Detect gross leaks; immerse in ethylene glycol at 60 torr
"Zyglo" test	Detect leaks and surface defects in the weld; "Zyglo"* XL-22 dye penetrant and ultraviolet light
Eddy-current test	Measure cladding thickness
Ultrasonic test	Detect flaws in sheath tubes
Concentricity	Measure variations in total wall thickness; magnetic reluctance instrument with ferromagnetic mandrel in place
Wall thickness and density	Detect circumferential variations in UO_2 distribution; gamma ray densitometer[20]
Visual inspection	Detect surface blemishes (e.g., scratches, gouges) that could impare the strength and ductility of the sheath or which could lead to accelerated corrosion

*Trademark of the Magnaflux Corporation.

Destructive Tests

Test	Purpose and Conditions
Immersion density	Measure core density in ring sections
Sheath metallography	Measure indentation depths, determine hydride distribution, and examine weld microstructure
Chemical analysis	Measure gas and moisture content of compacted UO_2, hydrogen content of sheaths
Mechanical properties	Measure strength and ductility of sheath and weld zone

Collapse Tests

The stability of oxide tubes to internal pressure generated during irradiation was estimated from collapse tests on unirradiated tubes with diameters from 1.24 to 3.05 inches and sheath thicknesses from 0.023 to 0.038 inch. Collapse pressures were measured at temperatures of interest by introducing successive increments of helium gas into the tube annulus until the inner sheath collapsed; the time at each pressure increment was 3 minutes. Test temperatures were as follows:

- Room temperature
- 100°C (the maximum sheath temperature expected during reactor shutdown after the coolant pressure was removed)
- 350°C (the sheath temperature expected during reactor operation)

Compacted cores provide appreciable support to inner sheaths. Collapse pressures in tubes compacted by vibration to about 85% of theoretical were approximately 4 times higher than for empty tubes as shown in Table III. Other tests on swaged tubes showed that swage-compaction following vibrational loading provides additional resistance to collapse. Tubes of similar size compacted by vibration only to 87% of TD and by vibration-plus-swaging to 92% TD collapsed at stress levels of about 18,700 psi, and 21,000 psi, respectively, when tested at 350°C.

TABLE III

Collapse Pressures of UO_2 Tubes and Empty Sheath Assemblies

Element Type	Inner Sheath Dimensions, inches OD	Thickness	Collapse Pressure, psi RT	100°C	350°C	Calculated Hoop Stress in Inner Sheath at Collapse RT	100°C	350°C
Empty Sheath Assembly								
Base case, SOT-1 (2.06 in. OD x 1.48 in. ID x 14 in. long)	1.54	0.032	330	-	-	6650	-	-
Vibratorily Filled Tubes								
SOT-1, as above								
80.5% density	1.54	0.032	-	-	600	-	-	14,900
86.5% density	1.54	0.031	1430	-	780	28,900	-	18,700
Swaged Tubes: 90-92% TD								
Large tube of reference design (3.66 in. OD x 2.99 in. ID x 13 in. long)	3.05	0.032	520	435 475	400	24,800	23,600 20,800	19,200
Intermediate tube of reference design (2.54 in. OD x 1.83 in. ID x 14 in. long)	1.88	0.023	600	570 575	490	25,600	24,300 24,500	20,900
2.06 in. OD x 1.47 in. ID x 14 in. long (SOT-1)	1.54	0.032	1620	1225	880 830 1000[a]	38,800	29,300	21,000 19,800 23,900
2.13 in. OD x 1.16 in. ID x 14 in. long (SOT-2)	1.236	0.038	-	1725	1225	-	28,000	19,900

a Tube temperature probably less than 350°C when tested.

The collapse pressure of the inner sheaths in oxide tubes of different diameters but swaged to about the same density was affected more by a limiting stress in the sheath than by the diameter, length, or sheath thickness. The limiting stress is believed to be that which initiates inelastic collapse and is closely approximated by the proportional limit of the Zircaloy. At 350°C where stress relief is operative, the hoop stress to cause collapse was about 20,000 psi for all swaged tubes, regardless of diameters or sheath thickness (Figure 15). However, at lower temperatures, the stress to cause collapse was significantly higher than at 350°C, and differed for the different tube sizes, probably because of differences in prior fabrication history of the cladding.

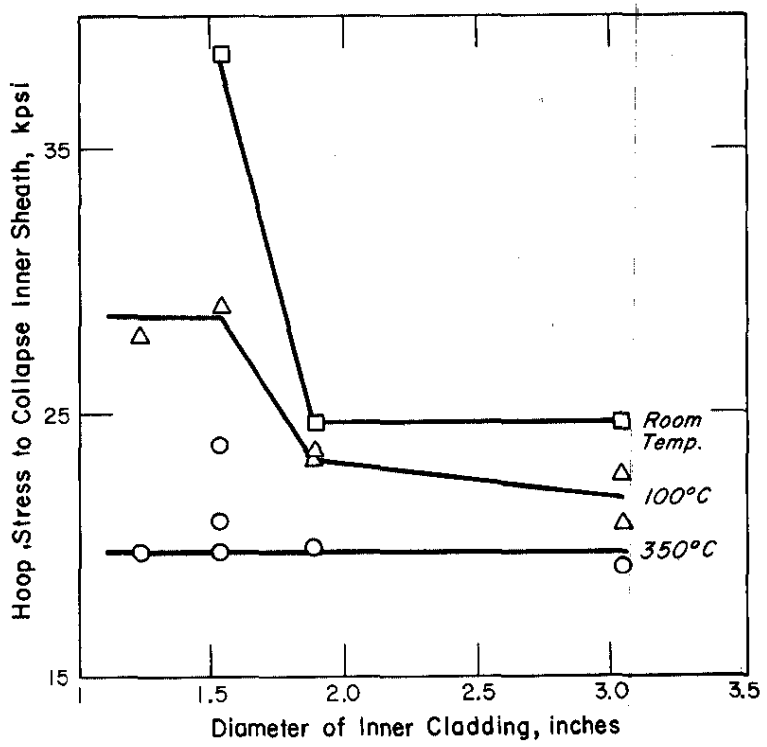


FIG. 15 EFFECT OF TUBE DIAMETER AND TEST TEMPERATURE ON COLLAPSE STRESS OF INNER SHEATH OF ZIRCALOY-CLAD UO_2 TUBES

Thermal Cycle Tests

Dimensional stability was maintained when UO_2 tubes fabricated by the reference process were cycled twenty times through a temperature range greater than that predicted for reactor operation. Both vibrated and swaged tubes were immersed in molten lead at 450°C for 5 to 19 minutes, and then quenched in boiling water; diametral changes were less than 0.001 inch.[21] When the lead bath was raised to 650°C, the outer diameters of the tubes increased about 0.007 inch after twelve cycles. No changes in inside diameter or length were observed; testing was discontinued when the Zircaloy cladding showed excessive oxidation.

Status

A mechanical compaction process for fabricating oxide tubes was demonstrated by producing development quantities of tubes suitable for irradiation. Satisfactory methods for controlling the dimensions and quality of the fuel elements were developed. Possible additional improvements in the process are single-pass swaging, shorter drying cycles, and a swageable permanent end plug.

Radial spacing of concentric oxide tubes was the only major fabrication technique not fully developed. Longitudinal rib spacers attached to the sheath by electron beam welding were being investigated when the program ended.[22,23] Rib spacers can be attached to the inner sheath before the fuel tube is fabricated or on the outer sheath after fabrication.

Swaging experiments conducted on two 5-foot-long fuel tubes fabricated with ribbed inner sheaths indicated that these tubes can be swaged-compacted over grooved mandrels to greater than 90% of theoretical density.[24] The ribs were attached to the inner sheath by electron beam welding through the outer surface. Ribs appeared intact when the mandrel was removed, but some portions of the ribs broke loose while the tube was being sectioned for examination. These failures are believed to have occurred at areas of poor weld penetration. A photomicrograph of a fully welded rib portion after swaging is shown in Figure 16.

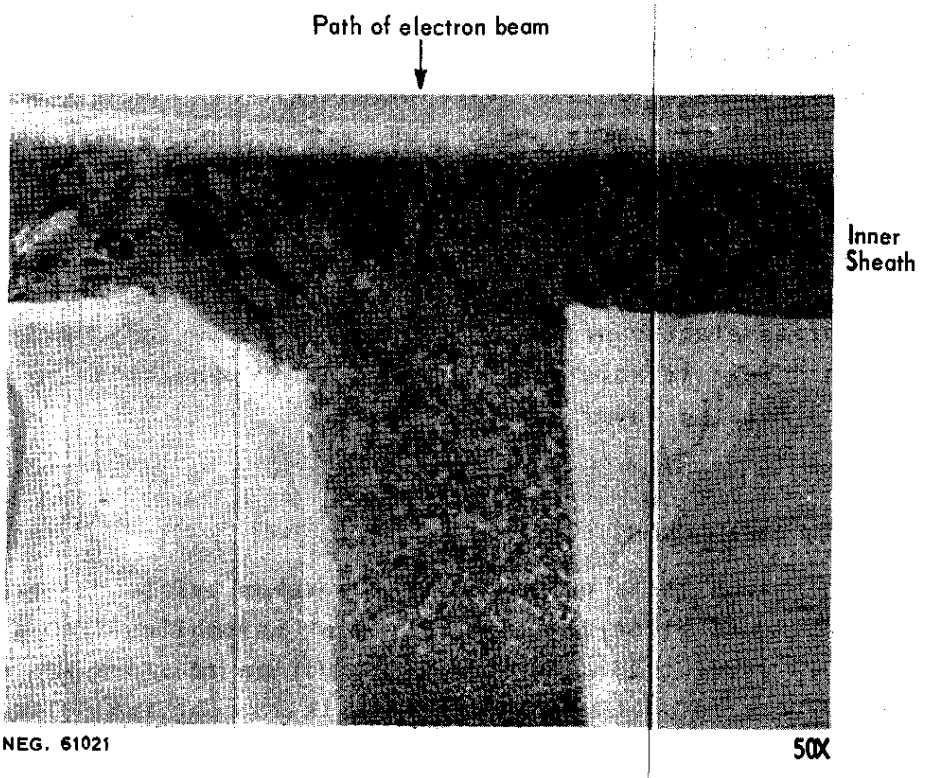


FIG. 16 ZIRCALOY SHEATH WITH FULLY WELDED RIB AFTER SWAGING

Additional work is needed to develop a satisfactory method of attaching longitudinal rib spacers to fuel tubes by electron beam welding. The mechanical traversing mechanism needs additional improvement, and weld conditions must be optimized to produce welded joints that meet the stringent requirements for service on fuel tubes.

IRRADIATION TESTS

Preliminary irradiation tests of ten assemblies containing 54 fuel tubes with compacted UO_2 cores were performed in Savannah River production reactors, which operated at cladding temperatures and coolant pressures substantially below those of interest for an economical full-scale power reactor. Tubes in the first three assemblies were fabricated with stainless steel sheaths as a stand-in for Zircaloy. All other tubes contained Zircaloy-2 sheaths.

Later irradiations of UO_2 fuel tubes were performed in the Heavy Water Components Test Reactor (HWCTR). In these irradiations, coolant pressures, sheath and core temperatures, and heat flux were representative of full-scale power reactors. Nearly one hundred tubes were irradiated in 19 assemblies; 45 were examined after irradiation.

Prior to startup of the HWCTR, two Zircaloy-sheathed uranium oxide tubes were irradiated at power reactor conditions in the Vallecitos Boiling Water Reactor (VBWR).

PRELIMINARY IRRADIATIONS

Irradiation tests on compacted UO_2 tubes before the startup of the HWCTR are given in Table IV. Test assemblies SPRO-2 through -13 were irradiated in SRP production reactors and consisted of vertical columns of five or six tubular fuel elements. Assemblies ZE-195 and ZE-197 were single tubes irradiated in the VBWR. All tubes were irradiated at low to moderate powers and exposures.

TABLE IV

Compacted UO_2 Tubes Irradiated Before Startup of HWCTR

Assembly Number[a]	Number of Tubes per Assembly	Nominal Dimensions, inches			Number of Swaging Passes	Oxide Treatment
		OD	ID	Core Length		
SPRO-2,3	5	2.14	1.46	22.5	12	None
SPRO-4	5	2.07	1.46	22.7	12	None
SPRO-5,6,7	6	2.30	1.46	20.6	7	None
SPRO-8,9	5	2.07	1.46	22.1	3	None
SPRO-12	6	2.06	1.47	18	2[b]	(c)
SPRO-13	5	2.06	1.47	22.1	2[b]	(c)
ZE-195[d]	1	2.06	1.47	36.0	None (Vibration only)	(c)
ZE-197[d]	1	2.06	1.47	36.0	2	(c)

a Tubes in Assemblies SPRO-2-4 fabricated with stainless steel sheaths; all other tubes fabricated with Zircaloy-2 sheaths.

b Three tubes in Assembly SPRO-12 and two tubes in SPRO-13 compacted by vibration only.

c Oxide outgassed in vacuum for 1 hour at 1000°C before loading and dried at 200°C after loading.

d Irradiated in the VBWR; all others irradiated in SRP production reactors.

Savannah River Production Reactor

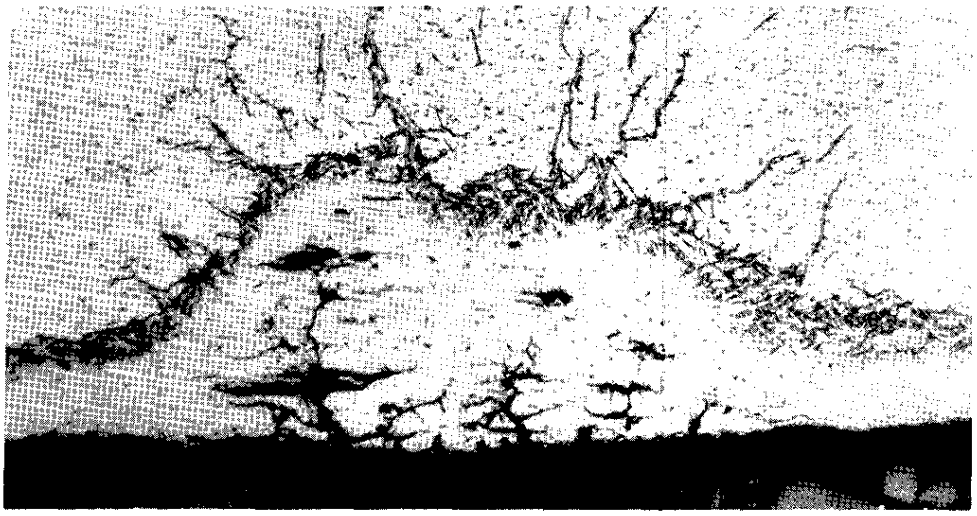
Sheath failure of a brittle nature occurred in six tubes with Zircaloy sheaths, irradiated in assemblies SPRO-5 through -9.[7] The outer sheaths failed during a startup following a shutdown from previous operation. Moisture associated with the powdered UO_2 is believed to have reacted with impurities in the UO_2, probably uranium metal inclusions, to liberate hydrogen which reacted with the relatively cool cladding and formed clumps of zirconium hydride. Embrittlement was due to hydride platelets which formed during migration of hydrogen from the core-sheath interface to the sheath-coolant interface, Figure 17. Subsequently, the fabrication process was modified to eliminate this form of embrittlement by controlling the moisture and hydrogen content of the oxide.[18]

Irradiation of the eleven tubes in Assemblies SPRO-12 and -13 verified that the modified fabrication process effectively eliminated hydrogen embrittlement of the Zircaloy sheath. No attack of zirconium hydride at the core-sheath interface was observed in the irradiated sheaths, and the concentration of hydride platelets was essentially unchanged during irradiation.

However, tube Z-211D in Assembly SPRO-13 failed during irradiation by collapse of the inner sheath. Cladding bulges developed in three of the four coolant passages bounded by the fuel tube and ribs of the adjacent housing tube, Figure 18. The cladding remained intact and no distortion occurred in the outer sheath.[8]

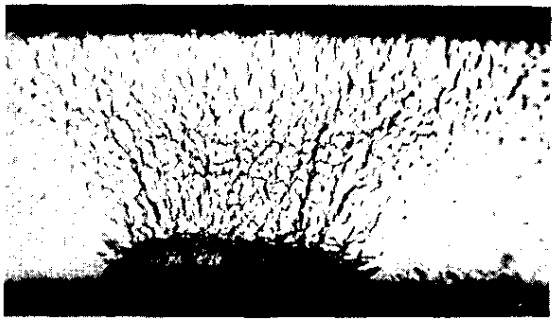
Subsequent examination showed that this tube contained three times as much free gas as any of the other tubes in the assembly; the failure was attributed to excessive gas release. The total gas release in the collapsed tube was approximately 200 cc/kg of UO_2, of which 54% was nitrogen and 31% was xenon; during irradiation this amount of gas generated an estimated internal pressure of about 1100 psi. The remaining four tubes released 45 to 62 cc/kg of gas.[25]

A higher-than-expected operating temperature was believed responsible for the high gas release that caused tube Z-211D to collapse during irradiation. Structural changes in the core of



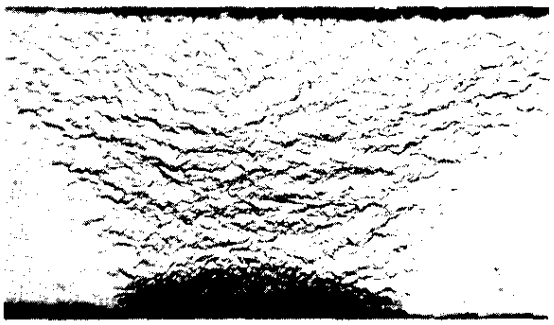
NEG. 33378 350X

Zirconium hydride on inside surface of outer sheath



NEG. 34154 40X

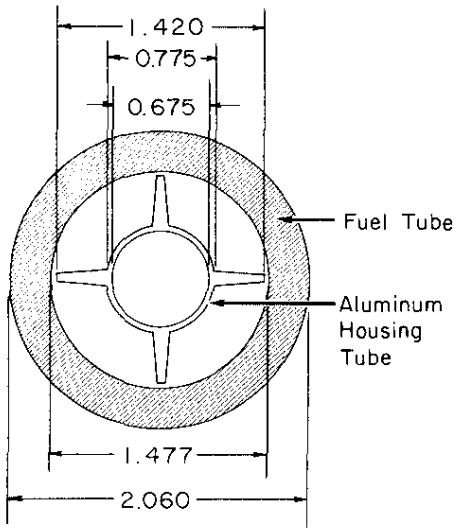
Radially oriented hydride in outer sheath



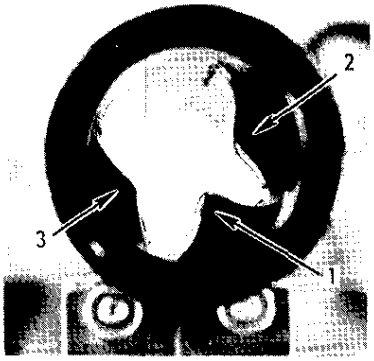
NEG. 34107 40X

Circumferentially oriented hydride in inner sheath

FIG. 17 DISTRIBUTION OF ZIRCONIUM HYDRIDE IN EMBRITTLED SHEATHS

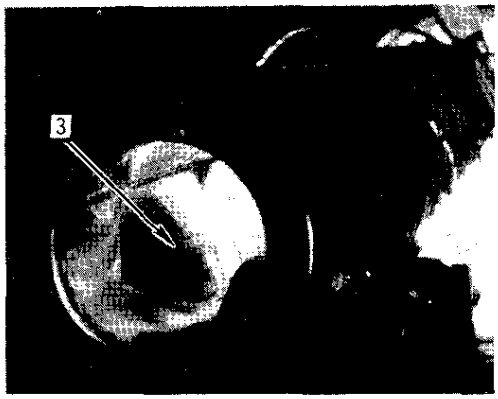


a. Before deformation (nominal)



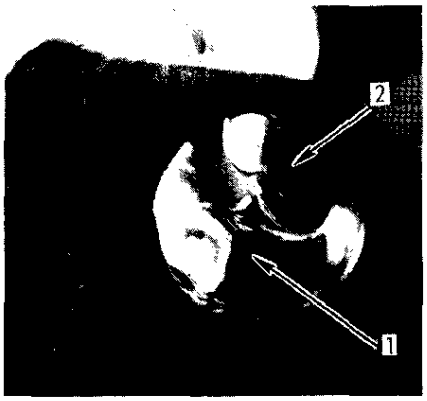
NEG. 43549

b. Silhouette of bulges



NEG. 43554

c. First oblique view



NEG. 43550

d. Second oblique view

FIG. 18 COLLAPSED INNER SHEATH IN IRRADIATED UO_2 TUBE Z211D
(The bulges are numbered in order of size)

this tube verified that it operated at a much higher temperature than the four remaining tubes in the assembly. The reason for the higher temperature was not determined, but the oxide temperatures in this assembly are believed to have been higher than in Assembly SPRO-12 as evidenced by greater release of fission product xenon in the SPRO-13 tubes (approximately 4 cc/kg vs 1 cc/kg for tubes in SPRO-12).

Two failures of swaged UO_2 tubes with stainless steel sheaths occurred during irradiation early in the program, and a third was detected when the tubes were examined after irradiation. In the most severe failure (tube 20A in Assembly SPRO-3), the outer sheath was cracked longitudinally for about 10 inches and defects were found in the welded end closure.[7] Branching transgranular cracks, characteristic of stress-corrosion cracking, were observed in the end plug and in the sheath near the split. Initial failure was attributed to the defective welded closure, but the subsequent fracture of the outer sheath was probably caused by stress-corrosion cracking induced by residual chlorides that remained in the fuel element after fabrication.

Two other failures of stainless steel sheathed tubes were identified but were not investigated further. One was a weld defect in tube 30C, Assembly SPRO-3, and the second was a small hole in the outer sheath of tube 44A, Assembly SPRO-4. The failures were too small to be seen by visual examination but were located by observing gas escaping from the core space when the tubes were pressurized with helium and immersed in water.

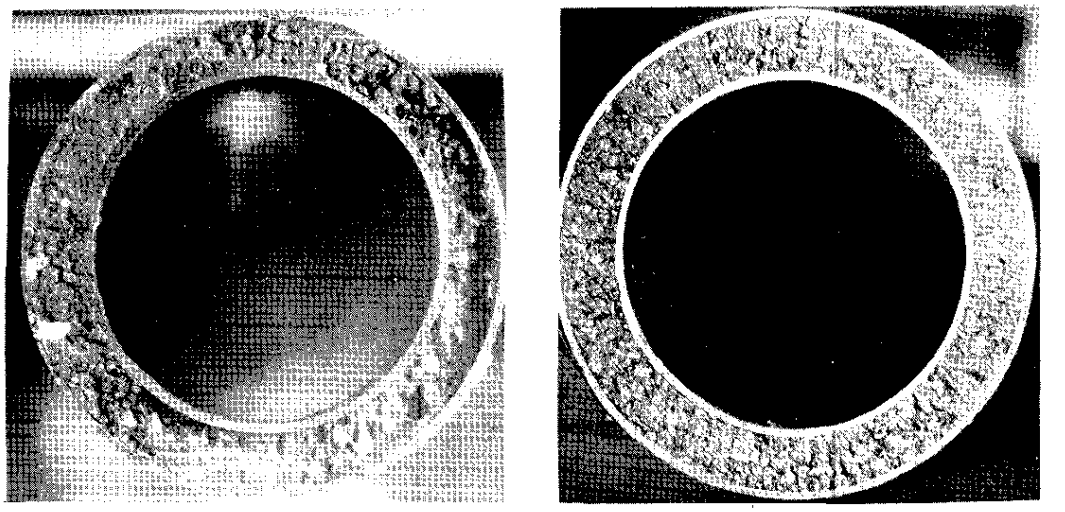
Vallecitos Boiling Water Reactor

Two tubular UO_2 elements were irradiated in the Vallecitos Boiling Water Reactor (VBWR) and discharged without incident after accumulating average exposures of about 3000 MWD/Te_u at a peak thermal rating of 22 watts/cm. The test was conducted to give an early demonstration of compacted UO_2 tubes irradiated under power reactor conditions and was terminated when the HWCTR became available. One tube was compacted by vibration-plus-swaging to 90% TD, the other by vibration only to 84% TD. The fabrication process was the SPRO-12 and -13 and contained steps to eliminate hydrogen embrittlement of the Zircaloy sheaths observed in earlier tests.[21] Tube dimensions are compared with those of earlier UO_2 tubes in Table IV.

Successful operation of the two UO_2 tubes in the VBWR was confirmed by surface inspection and dimensional measurements at Vallecitos and destructive examination at SRP. Changes in surface appearance and dimensions were negligible; the core sintered dur-

ing irradiation but did not undergo grain growth, Figure 19.

The free gas in the vibratorily-packed tube, ZE-195 contained a relatively large amount of nitrogen after irradiation (70 cc/kg UO_2), probably as a result of the limited effectiveness of outgassing coarse particles of fused UO_2 at 1000°C. The swage-compacted tube (ZE-197) released less nitrogen (22 cc/kg) and may have operated at a somewhat lower temperature because of higher thermal conductivity across the swaged core. Release of fission product xenon was about the same in both tubes (∿2 cc/kg).



NEG. 52813 2X

ZE-195 Vibrated

NEG. 52812 2X

ZE-197 Swaged

FIG. 19 CORE STRUCTURE OF UO_2 TUBES IRRADIATED IN THE VBWR
(Both sections show moderate core sintering but no grain growth)

IRRADIATIONS IN THE HWCTR

Description

The UO_2 tubes irradiated in the HWCTR were fabricated after the process was changed to eliminate causes of failures observed in earlier irradiations, i.e., sheath embrittlement and cracking from an internal source of hydrogen, and internal pressure build-up and collapse of the inner sheath from excessive release of gas by the UO_2. The changes consisted of (1) outgassing and drying

the powdered oxide and the compacted core, respectively, and (2) providing void chambers about one-inch long in the ends of each tube. The chambers contained perforated septums that separated the core from the void chamber and allowed passage of gases released from the core during irradiation.

The cores of some tubes were compacted by sonic vibration only to 82 to 87% TD; other tubes were further compacted by swaging over a mandrel to 89 to 91% TD. The tubes were compacted in lengths of 4 to 10 feet. These were generally cut into ∿14-inch sections to permit multiple irradiations in each reactor position, and to facilitate the postirradiation examination, but some 9-foot cores were also irradiated.

The oxide tubes were fabricated to "base-case" sizes with outer diameters of approximately 2.1 inches or to "reference-design" sizes approximating the intermediate and large tubes of a 3-tube design of practical interest.[1] Average dimensions of the tubes in each of the irradiation tests are given in Table V; the comparative sizes of the base-case tubes and the reference-design tubes are shown in Figure 2. The base-case tubes (SOT-1, SOT-2, SOT-5, and SOT-7 tests) were irradiated to explore the general behavior of powder-compacted UO_2 tubes and the effect of variations in fabrication practice and irradiation conditions. Tubes of the reference design (SOT-6, SOT-8, SOT-9 tests) were irradiated to demonstrate the behavior of full-size tubes and to extend technology developed in earlier tests.

The irradiation tests covered a range of operating conditions and fuel element dimensions, and included several material variables. Wall thicknesses and uranium enrichments were chosen to provide successively higher thermal ratings ($\int k d\theta$), from the relatively low values of the SOT-1 test to values in the SOT-7 test that resulted in center melting. As shown in Tables VI through X, the uranium oxides used for these tubes were outgassed by heat treatments at various conditions to reduce the amounts of impurity gases.

TABLE V

Average Dimensions of UO_2 Tubes Irradiated in the HWCTR

Dimension	Base Case								Reference Design	
	SOT-1(a)		OT-1(b)		SOT-2		SOT-5 SOT-7		Intermediate SOT-6 SOT-9	Large SOT-8
	V(c)	V+S(d)	V	V + S	V	V + S	V	V + S		
Outside Diameter, in.	2.060	2.062	2.060	2.062	2.153	2.121	2.152	2.121	2.540	3.651
Inside Diameter, in.	1.477	1.465	1.477	1.465	1.168	1.155	1.078	1.076	1.833	2.985
Sheath Thickness, in.										
Outer	0.029	0.029	0.029	0.029	0.030	0.034	0.030	0.033	0.023	0.036
Inner	0.031	0.031	0.031	0.031	0.038	0.037	0.038	0.037	0.023	0.032
Core Thickness, in.	0.232	0.238	0.232	0.238	0.424	0.411	0.470	0.455	0.308	0.260
Core Length, in.	11.38	11.38	111.4	111.4	10.84	10.84	10.75	10.75	10.75	9.88
Core Volume, $in.^3$	14.8	15.2	143.8	147.5	24.4	23.2	26.0	24.6	22.7	27.2
Void Volume (in end chambers), $in.^3$	1.08	1.08	1.08	1.08	3.40	3.30	3.67	3.54	2.65	2.74
Total Tube Length, in.	13.75	13.75	113.8	113.8	13.75	13.75	13.75	13.75	13.75	13.0

(a) SOT-1, etc. = Designations for the 14-inch segment of oxide tubes that comprise a single irradiation test in the HWCTR.
(b) OT-1 = Designation for the single tubes with 9-foot cores that comprise a single irradiation test in the HWCTR.
(c) V = Compacted by vibration.
(d) V + S = Compacted by vibration plus cold-swaging.

TABLE VI

Characteristics and Irradiation Conditions of UO_2 Tubes in SOT-1 Test

(Assemblies SOT-1-2 and SOT-1-3)

Tube No.[a]	Method of Compaction[b]	Core Density, % of Theoretical	∫kdθ, watts/cm Average in Time & Space	∫kdθ, watts/cm Peak	Exposure, MWD/MTU Average	Exposure, MWD/MTU Max.
SOT-1-2						
ZE-229E	V	85	6	18	5,000	6,600
ZE-225F	V + S	90	11	25	9,500	11,500
ZE-232E	V	84	16	25	13,500	15,400
ZE-222E	V + S	92	19	25	16,500	17,200
ZE-232D	V	84	18	24	16,700	17,200
ZE-225C	V + S	90	15	24	13,400	15,400
ZE-229F	V	85	8	18	7,400	10,400
SOT-1-3						
ZE-232F	V	84	6	14	2,900	3,700
ZE-225A	V + S	90	12	20	5,200	6,300
ZE-228D	V	84	17	23	7,600	8,600
ZE-225E	V + S	90	21	25	9,100	9,400
ZE-228G	V	86	20	25	9,100	9,400
ZE-222G	V + S	91	17	22	7,300	8,300
ZE-228B	V	84	9	17	4,100	5,600

(a) All tubes contained stoichiometric UO_2 enriched to 1.5% ^{235}U. All oxide was vacuum outgassed for 1 hour at 900-1000°C. Sheath material was low-nickel Zircaloy-2.
(b) V = Compacted by vibration only; V + S = Compacted by vibration plus swaging.

TABLE VII

Characteristics and Irradiation Conditions of UO_2 Tubes in SOT-2 Test

(Assemblies SOT-2-2 and SOT-2-3)

Tube No.[a]	Method of Compaction	Core Density, % of Theoretical	∫kdθ, watts/cm Average in Time & Space	∫kdθ, watts/cm Peak	Exposure, MWD/MTU Average	Exposure, MWD/MTU Max.
SOT-2-2						
Z-257C[b]	V + S	89	3	10	300	500
Z-250B	V	83	12	26	900	1300
Z-256A	V + S	90	24	43	1900	2400
Z-251A[c]	V	87	37	49	2900	3300
Z-251C[c,d]	V	82	44	50	3400	3500
Z-255B	V + S	90	40	48	3200	3400
Z-257B[b]	V + S	89	30	41	2400	2900
Z-252C	V	82	16	25	1100	1600
SOT-2-3						
Z-254A	V + S	90	3	7	200	400
Z-251B	V	83	10	20	800	1000
Z-256B	V + S	90	21	34	1500	2000
Z-252B[c]	V	83	35	47	2400	2800
Z-254B	V + S	90	41	48	3100	3200
Z-253C[c,d]	V	85	41	48	3000	3100
Z-257A[b]	V + S	89	31	40	2300	2500
Z-253B	V	85	16	29	1200	1600

(a) All tubes contained stoichiometric UO_2 of natural enrichment that was vacuum outgassed for 4 hours at 1500-1600°C.
(b) Sheath material was Zircaloy-2; all other tubes fabricated with low-nickel Zircaloy-2 sheaths.
(c) Longitudinal wrinkle formed in outer sheath during irradiation.
(d) Outer sheath cracked at apex of wrinkle.

TABLE VIII

Characteristics and Irradiation Conditions of UO_2 Tubes in SOT-5 and SOT-7 Tests

(Assemblies SOT-5-2 and SOT-7-2)

Tube No.	Oxide Treatment or Type(a)	Method of Compaction	Core Density, % of Theoretical	∫kdθ, watts/cm Average in Time & Space	∫kdθ, watts/cm Peak	Exposure, MWD/MTU Average	Exposure, MWD/MTU Max.
SOT-5-2							
Z-262A(b)	VO	V	86	14	35	400	600
Z-260C	HT(1750)	V + S	90	25	52	800	1000
Z-259C	VO	V + S	89	39	55	1300	1500
Z-259D(d)	VO	V + S	90	49	56	1600	1700
Z-261D	Hypostoichiometric	V + S	89	49	56	1600	1700
Z-261B	Hypostoichiometric	V + S	89	39	50	1300	1500
Z-262B(b)	VO	V	85	23	34	700	1000
SOT-7-2							
Z-265C	VO	V + S	90	8	12	100	200
Z-261C	Hypostoichiometric	V + S	90	14	23	200	300
Z-260B	HT(1750)	V + S	89	24	41	400	500
ZE-266C(c)	VO	V + S	89	50	66	800	1000
ZE-266A(c,e)	VO	V + S	91	64	68	1100	1100
Z-264A(f)	HT(1300)	V + S	90	49	54	800	900
Z-265B	VO	V + S	90	31	45	600	700

(a) VO - Stoichiometric UO_2 vacuum outgassed 4 hours at 1500-1600°C.
HT() - Stoichiometric UO_2 annealed in hydrogen for 12 hours at 1750°C or 4 hours at 1300°C as indicated.
Hypostoichiometric - Untreated arc-fused UO_2 with average O/U of 1.98.

(b) Sheath material was low-nickel Zircaloy-2; all other tubes fabricated with Zircaloy-2 sheaths.

(c) Enriched to 0.9% ^{235}U by blending natural UO_2 with 1.5% enriched UO_2; all other tubes fabricated with UO_2 of natural enrichment.

(d) Failed during irradiation when crack occurred in outer sheath through local area of severe hydriding located on outer surface of sheath; cause of hydriding unknown.

(e) Failed during irradiation when hole formed in inner sheath at top of tube over steel end chamber. Molten UO_2 had penetrated into the void chamber in this region during irradiation and may have caused excessive temperatures that resulted in sheath failure.

(f) The computed values of ∫kdθ for this tube are believed to be low because postirradiation examination shows that center melting occurred in this tube. Center melting also occurred in the two tubes in this assembly that contained enriched UO_2; in these tubes, center melting began at ∫kdθ of about 60 watts/cm. The error in the computed ∫kdθ values is attributed to assumptions made to simplify the calculation of power generation in UO_2 tubes containing different enrichments in the same assembly.

TABLE IX

Characteristics and Irradiation Conditions of UO_2 Tubes of Intermediate Reference Size

(Assemblies SOT-6-2 and SOT-9-2)

Tube No.(a)	Core Density, % of Theoretical	$\int kd\theta$, watts/cm Average in Time & Space	$\int kd\theta$, watts/cm Peak	Exposure, MWD/MTU Average	Exposure, MWD/MTU Max.
SOT-6-2(b)					
Z-275A	90	6	19	1700	1300
Z-279B	92	11	27	3000	2600
Z-273A	91	18	28	4300	3800
Z-278C	91	22	28	5100	4900
Z-272C	91	22	27	5000	4800
Z-273C	91	18	24	4400	3800
Z-275C	91	9	15	2800	2000
SOT-9-2(b)					
ZE-283A	92	13	24	1500	1900
ZE-282A	92	24	34	2600	3100
ZE-281B	92	32	38	3500	3900
ZE-280C	91	36	40	4000	4000
ZE-299A	92	33	40	3700	4000
ZE-283B	92	25	34	2800	3200
ZE-283C	91	14	22	1400	2100

(a) All tubes contain stoichiometric UO_2 vacuum outgassed for 4 hours at 1300-1400°C, have Zircaloy-2 sheaths, and were compacted by vibration plus swaging.

(b) SOT-6 tubes contain UO_2 of natural enrichment. SOT-9 tubes are identical in size, but are enriched to 1.2% ^{235}U by mechanically blending natural UO_2 with 1.5% enriched UO_2.

TABLE X

Characteristics and Irradiation Conditions of UO_2 Tubes of Large Reference Size

(Assembly SOT-8-3)

Tube No.(a)	Core Density, % of Theoretical	$\int kd\theta$, watts/cm Average in Time & Space	$\int kd\theta$, watts/cm Peak	Exposure, MWD/MTU Average	Exposure, MWD/MTU Max.
SOT-8-3					
ZE-297A	89	10	21	1400	1800
ZE-289A	92	18	29	2600	3200
ZE-297C	91	25	30	3800	4100
ZE-297B	92	29	33	4300	4300
ZE-290B	91	26	32	3900	4200
ZE-289D	92	19	28	3000	3400
ZE-290A	90	10	16	1600	2200

(a) All tubes -

- contain stoichiometric UO_2 that was enriched to 1.2% ^{235}U by mechanically blending natural UO_2 with 1.5% enriched UO_2 and was vacuum outgassed for 4 hours at 1300-1400°C.
- were fabricated with Zircaloy-2 sheaths.
- were compacted by vibration plus swaging.

Information on the operating conditions of individual test assemblies and elements was obtained from calorimetric (flow, ΔT) data from the HWCTR operating records that were processed by appropriate computer codes to provide values for such parameters as $\int kd\theta$, specific power, heat flux, and exposure. The computer code provided a good approximation to the actual values of the various parameters at any axial location in the fuel column by computing individual values of the parameters for each of 19 equally spaced locations between the top and bottom of the fuel column. Plots of three important parameters, the peak $\int kd\theta$, the time-weighted average $\int kd\theta$ (very nearly equal to exposure-weighted values), and the total exposure were prepared for each assembly;[2] a typical plot for Assembly SOT-2-3 is shown in Figure 20.

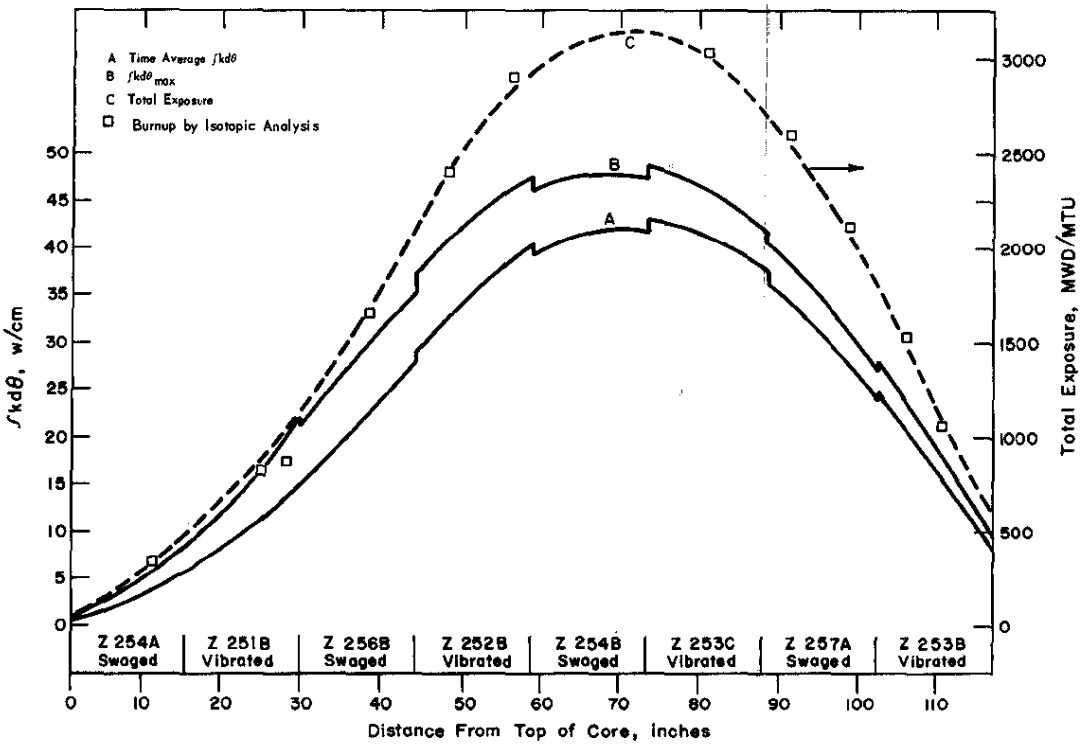


FIG. 20 IRRADIATION CONDITIONS FOR ASSEMBLY SOT-2-3

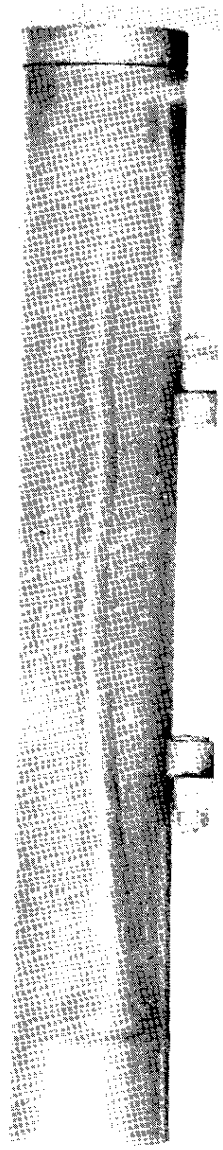
Isotopic analyses of core samples from the irradiated fuel tubes in three assemblies (including Assembly SOT-2-3, Figure 20) confirmed the cumulative exposures as obtained by calculation from the calorimetric data. Agreement between the two sets of data was good. Most of the analytical values were slightly higher than the calculated values; the average difference was only 4%.

Results

Effect of Core Density and Thermal Rating on Outer Sheath Wrinkling

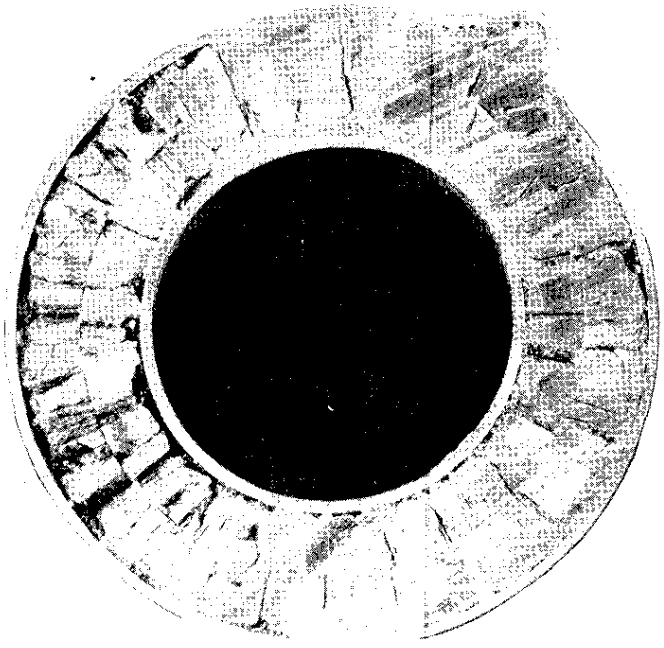
The outer sheaths of four tubes with core densitites of 82 to 87% formed longitudinal wrinkles during irradiation (Figure 21). On two of these tubes (one each in the SOT-2-2 and SOT-2-3 assemblies), deformation of the sheath at the apex of the ridge was severe enough to cause small cracks that released fission gases to the reactor coolant and required discharge of the assemblies. The core density and thermal rating of the tubes with and without wrinkled outer sheaths are compared in Figure 22. The data indicate that tubes compacted to 80 to 85% TD can be operated safely at thermal ratings of about 25 watts/cm, and that an increase in the density to ≥90% TD is required for satisfactory operation at thermal ratings of 40 to 50 watts/cm. The data were not sufficient to evaluate the effect of exposure or tube size; however, higher exposures and actual dimensions of interest in power reactor service are not expected to affect the conclusions from the present tests on UO_2 tubes irradiated to about 3,000 MWD/MTU.

Sheath wrinkling was accompanied by a reduction in core volume that varied inversely with the compacted density of the UO_2; the decrease in core volume ranged from 2% to 9% for tubes that varied from 87% to 82% TD, respectively. The stabilization of these cores at final densities of about 90% is consistent with the finding (Figure 22) that cores above 90% TD are stable with respect to outer sheath wrinkling at the highest thermal ratings tested.

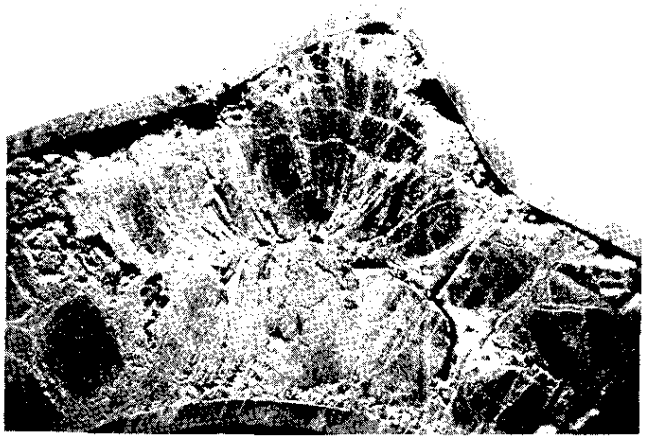


NEG. 52108 0.4X

a. Surface Appearance



NEG. 52145 1.7X



NEG. 52042 3.8X

b. Core Section

FIG. 21 TUBE FAILURE BY WRINKLING OF OUTER SHEATH

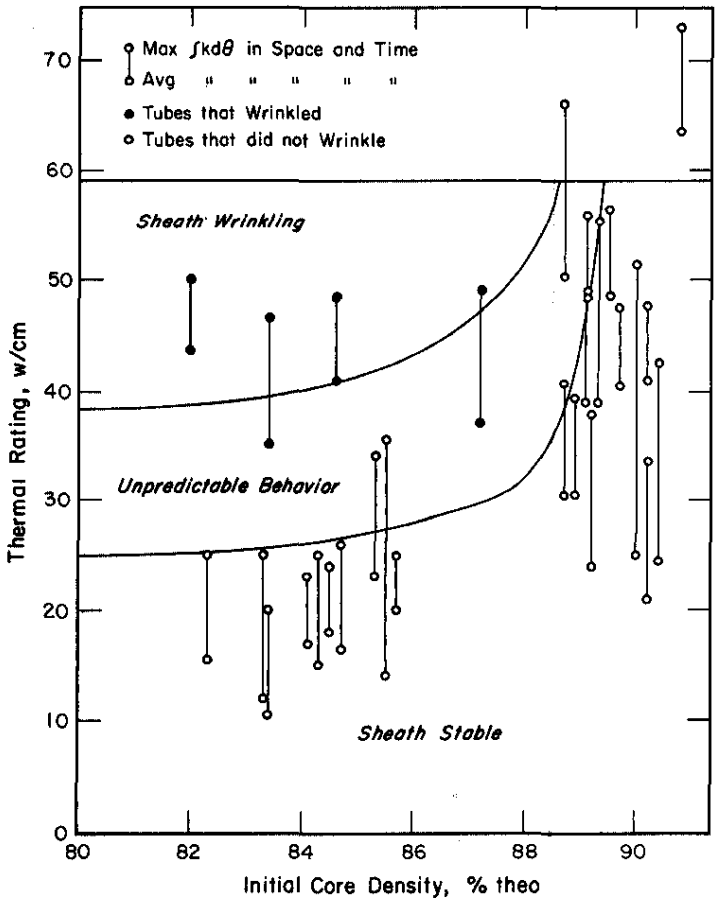


FIG. 22 EFFECT OF THERMAL RATING AND CORE DENSITY ON OUTER SHEATH WRINKLING OF UO_2 TUBES

The core structure of wrinkled tubes indicates that the UO_2 became sufficiently plastic to allow the wrinkles to form by collapse induced by the external coolant pressure (1200 psi). At the wrinkle, both the deformation of the columnar grains and the distribution of UO_2 that had not undergone grain growth indicate that sheath and core deformed as a unit with little relative movement at the core-sheath interface (Figure 23). The wrinkle is believed to have initiated early in the irradiation and to have proceeded at a decreasing rate determined by the creep strength of the UO_2 core; as wrinkling proceeded, the core densified and became less plastic. Sheath failures occurred at the apex of the wrinkle in two severely wrinkled tubes when sheath temperatures were low and stresses were high -- during shutdown or startup after a shutdown.

NEG. 53092 5X

Note grain deformation along thermal center indicating lateral movement of UO_2 into the region of the wrinkle and then vertical movement toward the wrinkle. Tube Z-252B.

FIG. 23 CORE STRUCTURE AT WRINKLE IN OUTER SHEATH OF UO_2 TUBE

Effect of Oxide Treatment and Thermal Rating on Gas Release

The gas in the void spaces of the fuel tubes after irradiation was principally xenon, nitrogen, and helium. Xenon is a fission product; nitrogen is an impurity of the UO_2 core,[4] and helium was admitted to the evacuated fuel tube prior to final sealing -- traces of argon from TIG welding were also admitted during fabrication. The gas contents of irradiated tubes are summarized in Tables XI-XV.

TABLE XI

Free Gas in Irradiated UO_2 Tubes from SOT-1 Test

(Assemblies SOT-1-2 and SOT-1-3)

Tube No.	Gas Release (Major Constituents Only), cc/kg UO_2								Volume of He + Ar, cc at STP	Initial Free Volume, cc	Xenon Release, % of That Produced by Fission
	Total	Xe	Kr	N_2	O_2	He	Ar	CO_2			
Assembly SOT-1-2											
ZE-229E[a]											
ZE-225F[a]											
ZE-232E[a]											
ZE-222E	68	19.1	3.2	21	0.26	19.1	3.2	0.05	54	39	4.8
ZE-232D	224	84.0	13.3	97	0.18	21.7	7.1	0.24	64	55	21
ZE-225C	73	25.3	2.1	29	0.05	15.7	0.5	0.05	39	40	7.8
ZE-229F	23	3.7	0.3	0.2	0.03	18.4	0.3	0.01	41	54	2.1
Assembly SOT-1-3											
ZE-232F	48	0.90	0.14	26	0.17	20.1	1.0	0.05	47	55	1.3
ZE-225A	97	3.14	0.52	74	0.10	15.7	3.0	0.04	43	41	2.5
ZE-228D	94	5.70	0.90	61	0.33	24.7	1.4	0.04	58	56	3.1
ZE-225E	135	13.8	2.1	101	0.08	13.6	4.5	0.05	44	41	6.4
ZE-228G	136	22.2	3.8	83	0.43	26.1	0.2	0.05	59	53	10
ZE-222G	76	7.10	1.1	49	0.05	15.4	3.5	0.03	45	40	4.0
ZE-228B	93	1.82	0.31	66	0.21	23.8	0.50	0.02	54	56	1.8

(a) Not examined.

TABLE XII

Free Gas in Irradiated UO_2 Tubes from SOT-2 Test

(Assemblies SOT-2-2 and SOT-2-3)

Tube No.	Gas Release (Major Constituents Only), cc/kg UO_2 at STP						Volume of He + Ar, cc at STP	Initial Free Volume, cc	Xenon Release, % of That Produced by Fission
	Total	Xe	N_2	D_2[a]	He	Ar			
Assembly SOT-2-2									
257C	23.5	0.05	0.7	ND	23	0.1	86	96	0.6
250B	31.0	1.1	0.3	ND	29	0.6	107	122	0.5
256A	48.8	4.8	16	ND	26	0.1	97	90	10
251A	69.2	31	5.6	ND	28	1.3	111	107	45
251C	Gas lost via sheath cracks								-
255B	69.2	39	0.2	ND	26	0.1	94	92	50
257B	40.2	14	5.0	ND	16	0.2	56	96	24
252C	31.5	ND	1.0	ND	30	0.03	109	126	-
Assembly SOT-2-3									
254A	30.4	0.06	6.0	ND	24	0.3	86	92	1.1
251B	30.3	0.3	1.5	ND	27	1.3	104	122	1.6
256B	29.6	0.9	4.5	ND	24	0.1	88	91	2.5
252B	76.4	29	11	ND	32	0.2	117	118	48
254B	302	27	23	218[b]	30	0.2	112	91	35
253C	Gas lost via sheath cracks								-
257A	50.0	13	5.7	ND	28	0.3	102	96	23
253B	26.6	0.3	5.9	ND	16	2.5	71	117	1.0

(a) "ND" signifies none detected.
(b) Presence of deuterium indicates that coolant leaked into the element during irradiation. Pressurization of the element with helium during the postirradiation examination failed to locate the leak site.

TABLE XIII

Free Gas in Irradiated UO_2 Tubes in SOT-5 and SOT-7 Tests

(Assemblies SOT-5-2 and SOT-7-2)

Tube No.	Gas Release (Major Constituents Only), cc/kg UO_2					Volume of He + Ar, cc at STP	Initial Free Volume, cc	Xenon Release, % of That Produced by Fission
	Total	Xe	N_2	He	Ar			
Assembly SOT-5-2								
Z-262A	27.3	0.19	3.0	22.6	1.25	96	122	1.7
Z-260C	24.9	2.06	0.16	21.8	0.49	89	99	10
Z-259C	39.1	10.6	0.35	22.8	3.68	78	101	34
Z-259D	Gas lost through hole in sheath					-	101	-
Z-261D	36.9	10.6	0.05	22.5	2.07	96	101	27
Z-261B	27.1	3.69	0.03	22.0	0.97	88	102	12
Z-262B	28.2	0.37	3.5	23.0	1.18	97	123	2.1
Assembly SOT-7-2								
Z-265C	(a)	-	-	-	-		97	-
Z-261C	20.9	0.05	0.05	20.4	0.39	82	100	0.9
Z-260B	21.7	0.47	0.13	20.5	0.45	83	102	4.7
ZE-266C	(b)	-	-	-	-	-	102	-
ZE-266A	(c)	-	-	-	-	-	96	-
Z-264A	24.1	6.45	1.23	13.8	1.59	62	99	32
Z-265B	32.1	1.20	12.6	17.0	1.07	72	97	8.4

(a) Over half the collected sample consisted of H_2, HD, and D_2; of the remainder, 10% was fission gas and 25% was helium and argon. Anomalous behavior of the pressure of the gas sample prevented a determination of the amount of gas collected. The pressure rose unusually slowly during collection and continued to rise in the sample bulb when the bulb was removed from the sampling system. This behavior is believed to result from D_2O and H_2O vapor collected with the sample.

(b) The gas sample contained about 61% nitrogen, 22% oxygen, 16% helium and argon, 0.2% carbon dioxide, and 0.1% fission gas. The sample was collected normally, but is believed to have picked up air between the time of gas collection and analysis (the sample pressure increased from 47 to 75 mm Hg). A reliable correction for inleakage of air is not possible, however, for the nitrogen and oxygen contents were much larger than could be accounted for by the observed pressure rise.

(c) Failed during irradiation; no gas sample available.

TABLE XIV

Free Gas in Irradiated UO_2 Tubes of Intermediate Reference Size

(Assemblies SOT-6-2 and SOT-9-2)

Tube No.	Gas Release (Major Constituents Only), cc/kg UO_2								Volume of He + Ar, cc at STP	Initial Free Volume, cc	Xenon Release, % of That Produced by Fission
	Total	Xe	Kr	N_2	O_2	He	Ar	CO_2			
Assembly SOT-6-2											
Z-275A[a]											
Z-279B[a]											
Z-273A	37	4.2	0.4	12	0.06	19.3	1.0	0.0	76	76	4.0
Z-278C	48	11.6	0.9	14	0.05	21.1	0.8	0.02	79	77	9.5
Z-272C	35	11.6	0.9	5	0.04	16.4	1.0	0.1	64	77	9.5
Z-273C	40	5.0	0.4	14	0.1	19.4	1.4	0.02	72	76	4.8
Z-275C[a]											
Assembly SOT-9-2											
ZE-283A[a]											
ZE-282A	51	7.6	0.7	20	0.12	21.3	0.6	0.01	79	73	12
ZE-281B	68	19.7	3.2	22	0.07	21.3	0.5	0.05	82	74	24
ZE-280C	90	40.0	3.4	25	0.45	20.6	0.1	0.06	76	76	42
ZE-299A[a]											
ZE-283B[a]											
ZE-283C[a]											

(a) Not examined.

TABLE XV

Free Gas in Irradiated UO_2 Tubes of Large Reference Size

(Assembly SOT-8-3)

Tube No.	Gas Release (Major Constituents Only), cc/kg UO_2								Volume of He + Ar, cc at STP	Initial Free Volume, cc	Xenon Release, % of That Produced by Fission
	Total	Xe	Kr	N_2	O_2	He	Ar	CO_2			
Assembly SOT-8-3											
ZE-297A[a]											
ZE-289A[a]											
ZE-297C[a]											
ZE-297B	40	12.0	1.0	8	0.04	17.4	1.1	0.02	83	83	11.6
ZE-290B	40	9.6	0.8	13	0.02	15.8	0.4	0.02	72	84	10.2
ZE-289D	34	3.4	0.3	14	0.22	15.7	0.9	0.01	75	75	4.8
ZE-290A[a]											

(a) Not examined.

The oxide temperature, as measured by the thermal rating, sharply affected the xenon release, as shown in Figure 24. The sharp increase in xenon release above 25 watts/cm was associated with columnar grain growth in the UO_2 cores. The effects of core density or exposure on the observed fractional release of xenon were minor.

Nitrogen gas released from the fused UO_2 during irradiation ranged from 0.03 to 101 cc gas (STP) per kg of UO_2. The influence of preirradiation treatment of the UO_2 on nitrogen release in-pile is shown in Table I. High temperature treatments of fused urania in either vacuum or in hydrogen were effective in reducing both the nitride content and the amount of nitrogen released during irradiation. Substoichiometric UO_2, with no preirradiation treatment, released extremely small amounts of nitrogen during irradiation. The table includes elements irradiated under a wide range of conditions, but these variations appeared less important than the effect of pretreatment.

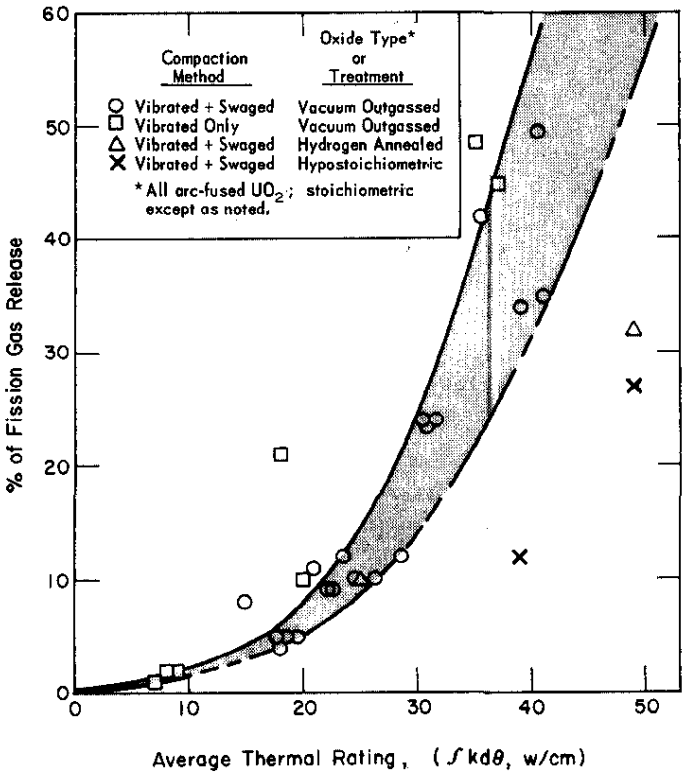


FIG. 24 FISSION GAS RELEASE vs THERMAL RATING IN UO_2 TUBES

Dimensional Changes

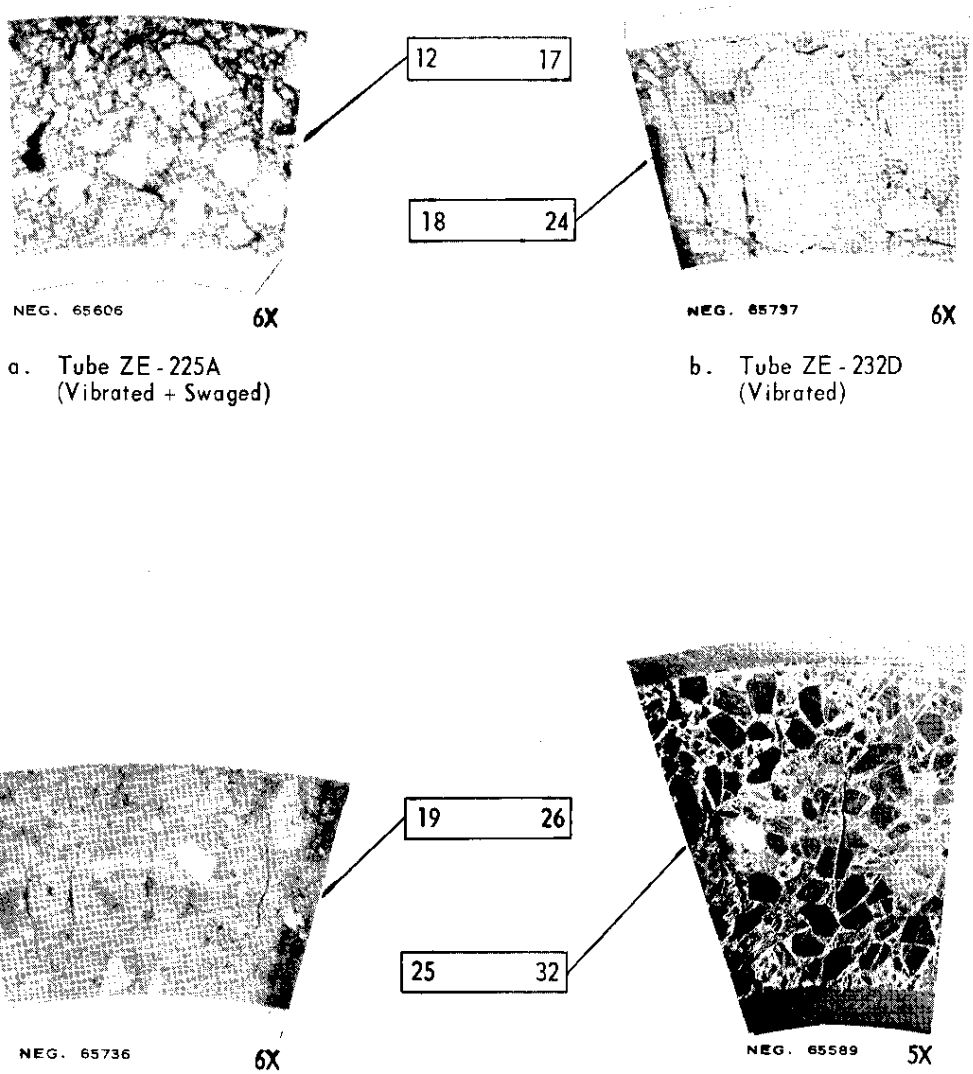
The average changes in dimensions of the irradiated tubes were acceptably small except where the outer sheaths wrinkled, or where melting was extensive in the central part of the core. Essentially no change in dimensions occurred during irradiation of tubes of the reference design that operated at the highest power levels in the SOT-6, SOT-8, and SOT-9 tests. The average changes in tubes that did not undergo wrinkling or central melting ranged from +6 to -3 mils for the outer diameters and +3 to -3 mils for the wall thicknesses -- ±3 mils is about the limit of the reliability of the data. For tubes that developed wrinkles in the outer sheath, the diameters at the crest of the associated ridge increased 19 to 105 mils; away from the ridge in the same region, the outer diameters decreased 14 to 54 mils.

In contrast to small decreases in volume generally observed for tubes irradiated at lower thermal ratings, volumes increased for tubes that operated with molten centers. Tube ZE-266A, irradiated at an average peak thermal rating of 66 watts/cm, increased 4% in volume; the outer sheath of this element was bulged over its lower half and exhibited a maximum residual strain of 3% (76 mils) without rupture. Tube ZE-266C, operated at an average peak of 58 watts/cm and a maximum peak of 66 watts/cm, increased 1.2% in volume. Both increases are believed to result from the volume increase of UO_2 on melting.

Average changes in length calculated from pre- and post-irradiation measurements varied from -30 mils to +55 mils; however, this is believed to represent scatter in the data rather than actual changes in length.

Core Microscopy

The structure of the powder cores was modified during irradiation with the greatest changes occurring in tubes that operated at the highest thermal ratings ($\int k d\theta$). The gross appearance of cores irradiated at different ratings is shown in the mounted-and-polished sections of Figure 25.



a. Tube ZE-225A
(Vibrated + Swaged)

b. Tube ZE-232D
(Vibrated)

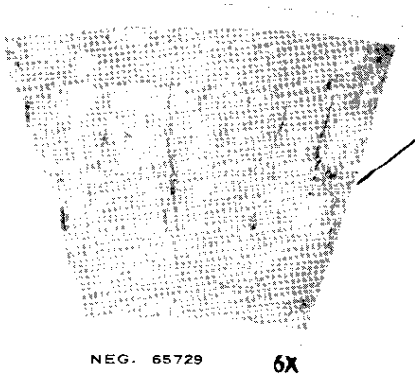
c. Tube ZE-222E
(Vibrated + Swaged)

d. Tube Z-256B
(Vibrated + Swaged)

FIG. 25 EFFECT OF THERMAL RATING ON CORE STRUCTURE OF

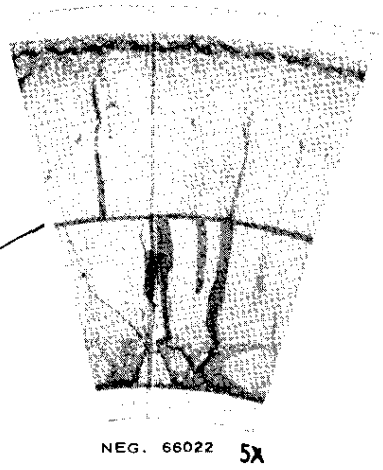
Thermal Rating,
watts/cm

Avg	Max
36	39
49	55
66	68



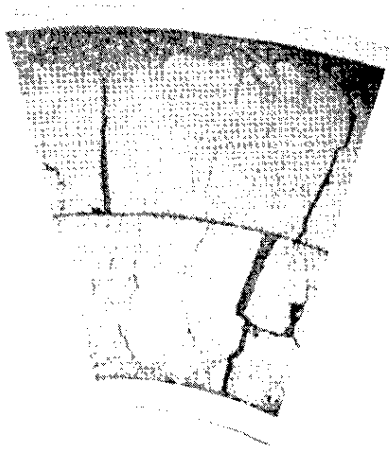
NEG. 65729 6X

e. Tube ZE-280C
(Vibrated + Swaged)



NEG. 66022 5X

f. Tube Z-259D
(Vibrated + Swaged)



NEG. 62243 5X

g. Tube ZE-266A
(Vibrated + Swaged)

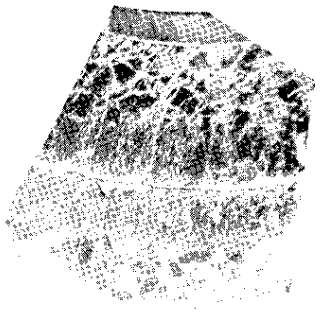
GROUND-AND-POLISHED SECTIONS OF IRRADIATED UO_2 TUBES

The extent of interparticle sintering increased as the thermal rating increased to about 20 watts/cm. Before irradiation, cores compacted by vibration alone were easily emptied from the tube, but after irradiation, even at low ratings (5 watts/cm), enough interparticle bonding had occurred to hold the core in place. Below 10 watts/cm, the interparticle bonding was weak as evidenced by loss of UO_2 particles during sectioning; this loss did not occur at higher thermal ratings. At 15-20 watts/cm, the core was sintered enough to crack in the cut section (Figure 25); loss of oxide particles during cutting occurred only in the cooler regions near the sheath.

Grain growth began at ∿25 watts/cm for vibratory-compacted cores and at ∿30 watts/cm for swage-compacted cores (Figures 25b and 25d). At 35-40 watts/cm, nearly half of the core had undergone grain growth (Figure 25e). Above 50 watts/cm, columnar grain growth was so extensive that the original particle size distribution was no longer evident (Figure 25f).

Columnar grain growth was extensive in a swage-compacted element irradiated at a peak $\int kd\theta$ of 39 watts/cm (Figure 26). Approximately midway between the edge and center of the core, lenticular voids formed and started to move up the thermal gradient leaving small columnar grains in their wakes. Most of these small grains are unstable, and at higher temperatures the boundaries are believed to collapse upon the void.

At the thermal center, a large void formed and divided the core into two nearly equal parts. Adjacent to the central void were many voids; some were nearly spherical and were randomly distributed within grains and on boundaries. These voids may be anchored at these sites or may be migrating very slowly because of the small thermal gradient in this region. The voids can grow by diffusion of gases from the UO_2 matrix, but probably most of these gases diffuse along grain boundaries to the central void. The region of core containing spherical voids is separated from the region containing lenticular voids by a region of large columnar grains (Figure 26). These grains are probably the combined results of previous void migration followed by grain growth at an earlier time in the irradiation.

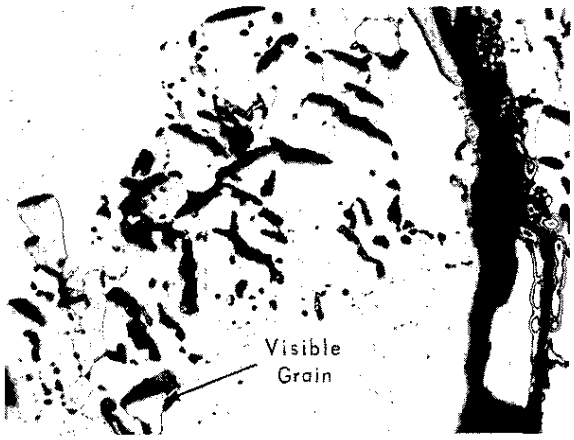


NEG. 65572

a. Section of tube Z-257A, which operated at $\int kd\theta = 35$ avg, 39 max watts/cm. (4X)

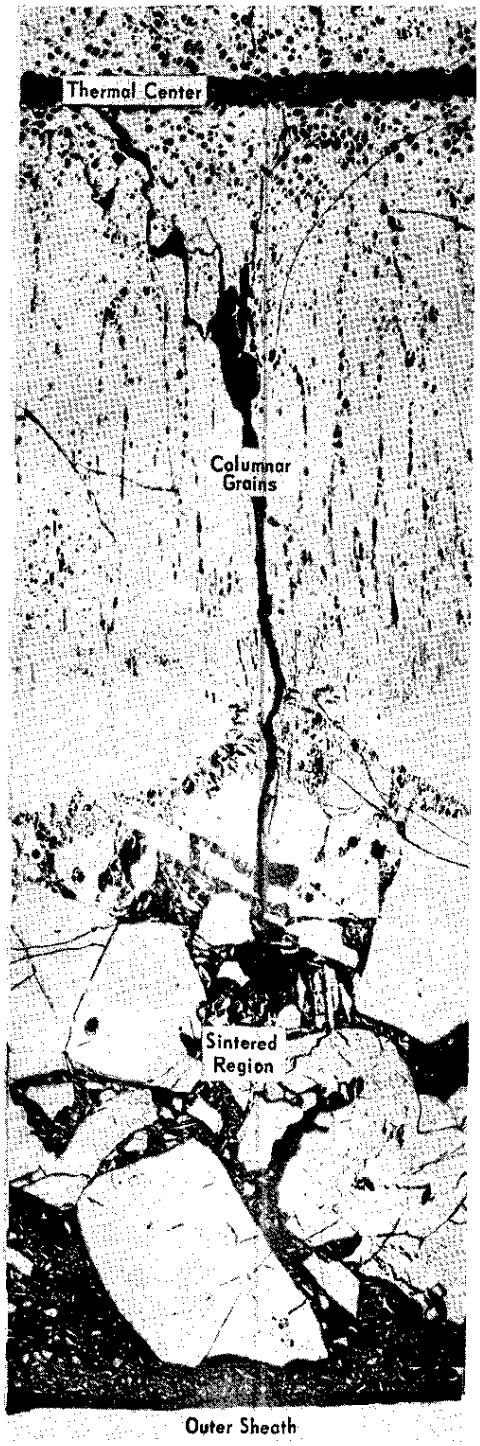
NEG. 65576

c. Randomly distributed voids apparently stabilized near thermal center. (210X)



NEG. 65574

d. Lenticular voids that migrated short distances to form small columnar grains. (210X)



NEG. 65685

35X

b.

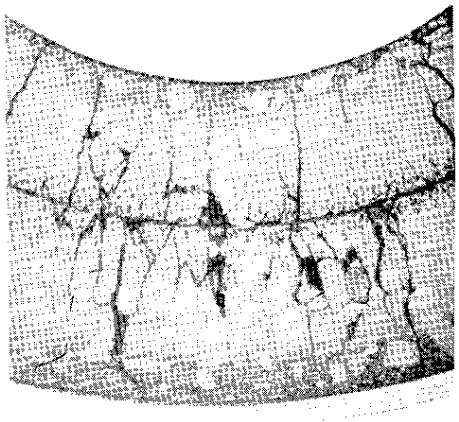
FIG. 26 COLUMNAR GRAINS IN CORE OF IRRADIATED UO_2 TUBE

Central melting began at 55 to 60 watts/cm. Tube ZE-266A, irradiated at a peak thermal rating of 68 watts/cm, exhibited an annular shrinkage cavity (~1 inch long) near the top of the tube, and a solid cross section over the remainder of the core. The porous zone that extends about a third of the way from the central void to the surface of the core provides additional evidence of melting in this element (Figure 27). Central porous zones containing both large isolated voids and small voids distributed on a substructure network have been identified by Hanford[26] as regions that were molten during irradiation.

Sheath Metallography

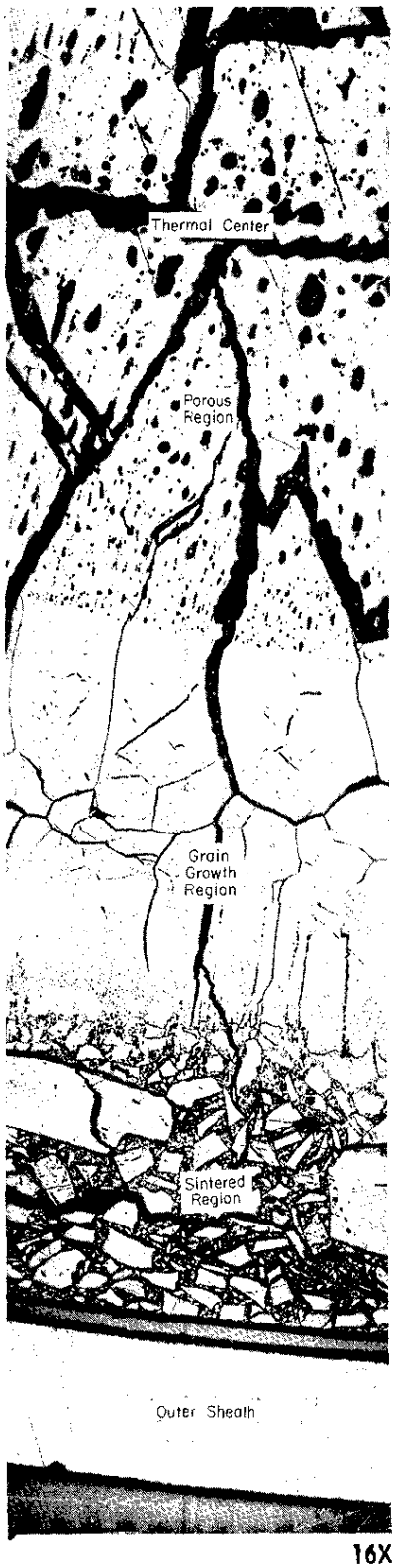
Sheath specimens were examined metallographically with particular attention to the incidence and distribution of zirconium hydride because of the known embrittlement that can result when hydride platelets are present in large amounts or in unfavorable distributions, for example, in strongly radial orientations. The hydride platelets tended to concentrate at the coolant side of the Zircaloy-2 sheathing, and a thin layer of surface oxide was observed on the core side of most sheath specimens. In the outer sheaths, the hydride platelets were oriented circumferentially; in the inner sheaths, the hydride platelets were generally oriented randomly with only a slight preference for radial orientation. The heaviest concentrations of hydride were in the Zircaloy-2 sheaths of the intermediate-size tubes, SOT-6 tests, irradiated for 160 days (Figure 28). The regions of concentrated hydride were confined to within about 0.007 inch of the surface; neither hydride nor oxide formation appears likely to impair the serviceability of the cladding.

Sheaths fabricated from low-nickel Zircaloy-2 exhibited very little zirconium hydride after irradiation (Figure 29). Test pieces from SOT-1-2 (irradiated for 416 days) exhibited trace-to-light concentrations of fine hydride platelets similar to the hydride concentrations in like tubes after irradiation of much shorter duration in an SRP production reactor.



NEG. 66007 4X

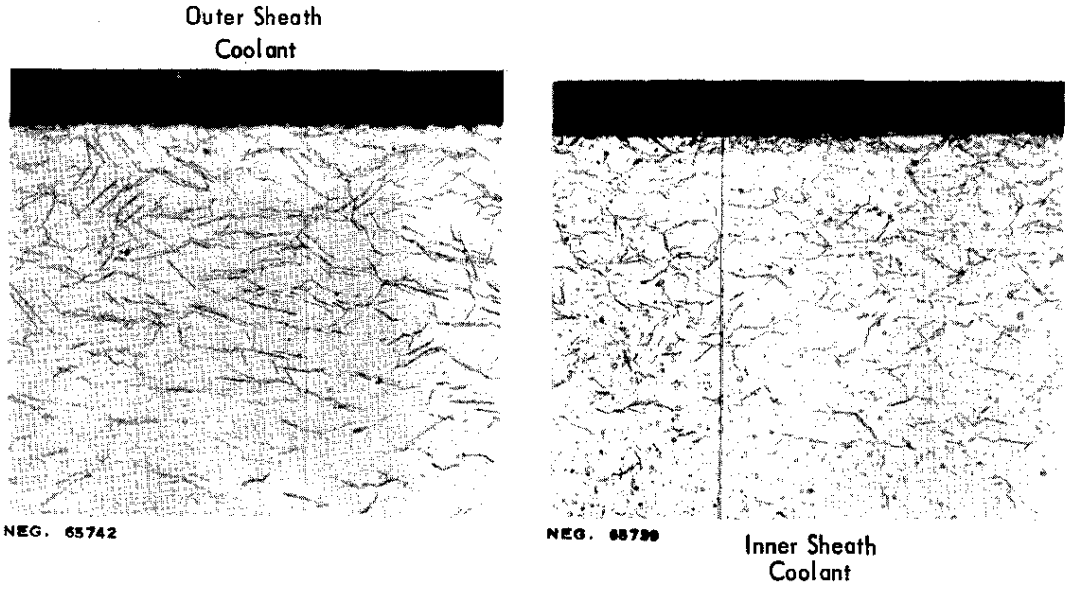
a. Section of tube ZE-266A, which operated at $\int kd\theta = 63$ avg, 67 max, watts/cm.



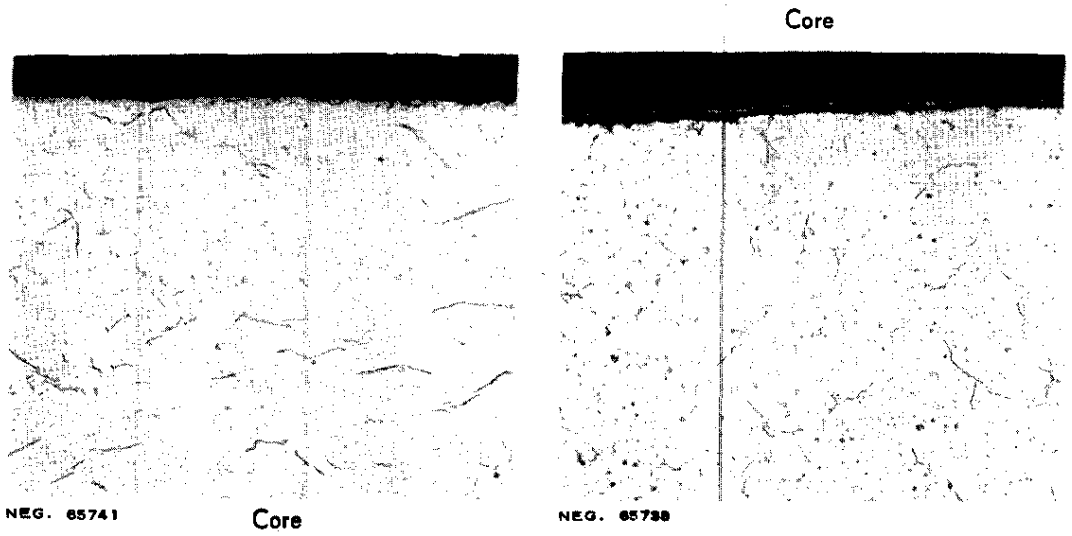
16X

b. Large porous zone is believed to define a region of central melting

FIG. 27 STRUCTURE OF UO_2 CORE AFTER IRRADIATION AT HIGH THERMAL RATINGS



Etched sections, showing hydride platelets nearest coolant. Note strong tendency for preferred circumferential orientations in the outer sheath tubing. (250X)

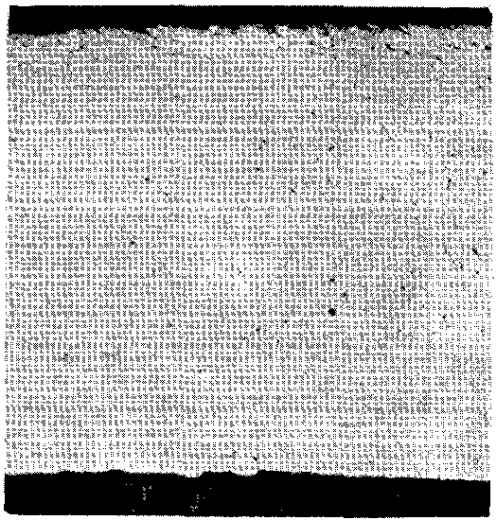


Etched sections, showing hydride platelets nearest core. Note that platelets in outer sheath are circumferential; those of inner sheath are largely radial. Layer of ZrO_2 is also visible on inner sheath specimen (250X)

FIG. 28 DISTRIBUTION OF ZIRCONIUM HYDRIDE IN ZIRCALOY-2 SHEATHS (2.5 inch-diameter reference design)

Outer Sheath

Coolant

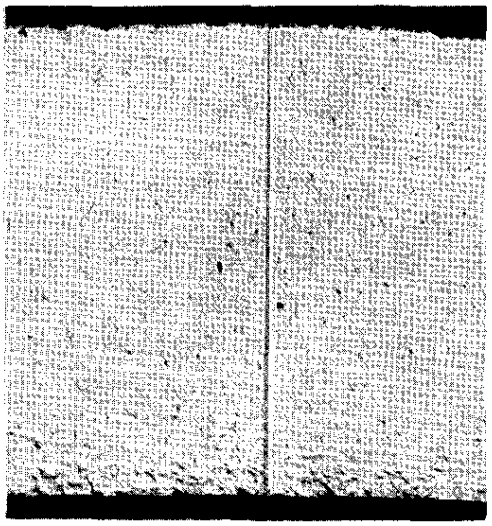


NEG. 65735

Core

Inner Sheath

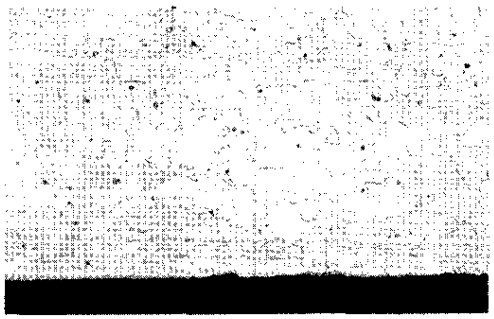
Core



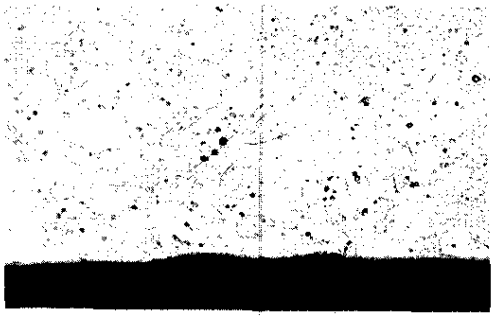
NEG. 65734

Coolant

As-polished sections (75X)



NEG. 65731



NEG. 65732

Etched sections showing light concentrations of fine platelets nearest the coolant. (250X)

FIG. 29 DISTRIBUTION OF ZIRCONIUM HYDRIDE IN LOW-NICKEL ZIRCALOY-2 SHEATHS
(Transverse sections of tube ZE-22E, SOT-1-2, irradiated 416 days)

Sheath Failures

In the HWCTR irradiation program, eleven of the 97 UO_2 elements failed during testing; in nine of these failures the integrity of the Zircaloy sheath was lost. Significant amounts of gaseous fission products were released in seven of the sheath ruptures. In two cases, no release was noted; however, the tubes contained deuterium gas when destructively examined after irradiation. In no case was there any extensive physical damage or appreciable release of UO_2. Information on the eleven failures, including four failures caused by wrinkling of the outer sheath discussed earlier, is given in Table XVI.

TABLE XVI

Failures Of UO_2 Tubes Irradiated In The HWCTR

Type of Failure	No. of Tubes	Known or Probable Cause
Wrinkling of outer sheath	4	Plastic flow of UO_2 compacted to less than 90% theoretical density and operated at high temperature with high coolant pressure.
Collapse of inner sheath	1	Faulty weld closure.
Penetration of outer sheath	2	Fretting wear from mechanical vibration.
Penetration of inner sheath	1	Sheath perforated by reaction with carbon steel end chamber (molten UO_2 intruded into chamber).
Rupture of outer sheath	1	Not determined. Cracked found in outer sheath through small hydride area on outer surface.
Sheath rupture	2	Not determined. Deuterium found in tube after irradiation was only evidence of failure.

Status

A total of 138 UO_2 tubes with Zircaloy sheathing were irradiated over a broad range of exposures and thermal ratings. The maximum exposure was 17,000 MWD/MTU, and the maximum thermal rating was 68 watts/cm. Performance limitations that were identified in these tests are discussed below.

Outer Sheath Wrinkling

As described on page 51, four UO_2 tubes failed during irradiation in the HWCTR by the formation of longitudinal wrinkles on the outer sheath. These failures occurred in tubes that had been compacted by vibration alone to rather low packed densities (82-87% TD) and then irradiated at moderate-to-high thermal ratings (37-49 watts/cm, peak). Volume reductions during irradiation indicated that, in each tube, the UO_2 core densified to about 90% TD. The failure of the vibratory-compacted elements was attributed to plastic flow of the core during the densification process, which led to reduction in sheath support and consequent wrinkling of the outer sheath by the pressure of the coolant.

Cores swaged to 90% TD or greater will provide sufficient support to prevent collapse of the outer sheath of tubes operated up to at least 40 watts/cm. This conclusion is based on:

(a) Satisfactory performance of 7 swaged tubes operated at thermal ratings of 40-55 watts/cm (exposures to 4000 MWD/MTU).

(b) Negligible change in core density during irradiation of swaged tubes in (a) above, whereas cores of vibratory-compacted tubes that exhibited sheath collapse densified to about the density obtained by swage-compaction (90% TD).

(c) Negligible change in density of other swage-compacted and vibratory-compacted tubes irradiated to exposures of 14,000-17,000 MWD/MTU at thermal ratings between 17 and 25 watts/cm.

Inner Sheath Collapse

Early in the program, one UO_2 tube failed by collapse of the inner sheath during irradiation in an SRP production reactor at low coolant pressure. After irradiation, the failed element contained an unusually large amount of fission and sorbed gas (page 41), and the columnar grain structure of the core showed that the

element had operated at an unusually high central temperature. The reason for the higher-than-expected operating temperature is not known; however, it is believed that a higher-than-normal release of sorbed gas from the core caused a pressure buildup that displaced the inner sheath and impaired the ability of the element to dissipate heat. The resulting increase in central temperature led to the release of additional gas which eventually caused the inner sheath to collapse.

The only other instance of inner sheath collapse resulted from a leaky weld, and occurred when the coolant pressure was removed after nuclear shutdown (Table XVI).

Urania fuel tubes fabricated by present techniques can probably operate in a power reactor at a rating of 30 watts/cm and reach burnups of 20,000 MWD/MTU without collapse of the inner sheath. This prediction is supported as follows:

(a) Sorbed gases in urania can be effectively controlled by outgassing during fabrication. The only impurity gas desorbed from the UO_2 in the current irradiations was nitrogen; in UO_2 that was vacuum outgassed at 1300°C or above, the release of nitrogen was less than 25 cc/kg UO_2. Hydrogen annealing was even more effective and should restrict release of impurity gases to less than 5 cc/kg UO_2; in three tubes containing hydrogen-annealed UO_2, the maximum release was 1 cc/kg UO_2. Thus the total amount of nonfission-product gas in irradiated tubes should not exceed 30-50 cc/kg UO_2.

(b) A urania tube compacted to about 90% TD and operated at 30 watts/cm will release about 25% of the fission gas formed during irradiation (Figure 24).

(c) Under the above conditions, the internal pressure during operation will be approximately 1300 psi at a burnup of 20,000 MWD/MTU, based on the following assumptions:

No void chambers at the ends of the element.

No axial flux gradient.

A central temperature of 1600°C and a surface temperature of 400°C throughout the operating life of the element.

(d) The internal pressure in the fuel element must exceed the reactor operating pressure (assumed here to be 1500 psi) before a failure mechanism that depends upon displacement of the inner sheath to impair heat transfer can become effective.

For a full-length tube operating in a full-scale power reactor, the internal pressure will be less than calculated above because the axial flux gradient will reduce the average temperature in the element, and because void chambers can be used to provide additional free volume at the ends of the tube. However, for such tubes operating at a moderate $\int k d\theta$, the maximum allowable exposure will be limited by buildup of internal pressure, even if each tube contains void chambers of reasonable size.

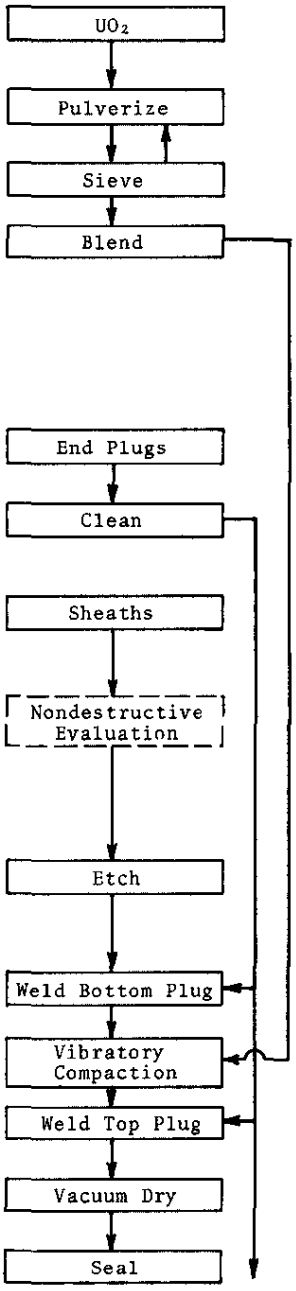
Results of calculations indicate that the inner sheaths of fuel tubes of interest will not collapse during reactor shutdown when the coolant pressure is removed. The internal pressures were calculated for various times after shutdown and compared with collapse pressures determined in out-of-pile tests (Table III). For any reasonable mode of reactor shutdown, the calculated pressure in the element at the time of depressurization was always less than the collapse pressure.

Hydride Embrittlement of Zircaloy Sheathing

Sheath failures due to hydride embrittlement caused by an internal source of hydrogen were eliminated by incorporation of a drying step in the fabrication process for the urania tubes. Before the assembled tubes are swaged, they are dried at 250-275°C for approximately 20 hours. The residual moisture content of the core is less than 15 ppm. One hundred and ten tubes made by a process that included the drying step have been irradiated with no repetitions of the earlier failures. Over 50 of these tubes were sectioned after irradiation; in all tubes, the Zircaloy sheaths were free of the type of hydriding that characterized the earlier failures.

APPENDIX A

DETAILS OF FABRICATION PROCESS

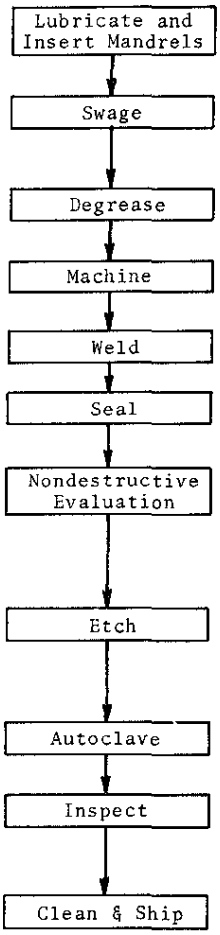


	Remarks
UO_2	Procure fused and crushed UO_2 (-2 mesh) from vendor; outgas in vacuum or hydrogen.
Pulverize	Grind UO_2 to desired size fractions in Braun type (disk) pulverizer approximately three passes per machine setting; yield - 96% usable material.
Sieve	Sieve desired fractions and separate steel particles magnetically.
Blend	Blend as follows:

U. S. Sieve Size	Weight Fraction %
-16 + 20	52
-60 + 100	28
-200	20

End Plugs	Machine temporary and permanent end plugs from Zircaloy stock.
Clean	Degrease plugs in caustic solution; rinse and agitate in hot flowing water, repeat in deionized water. Rinse in alcohol and drain. Dry in forced hot air system and store in polyethylene bags.
Sheaths	Procure Zircaloy-2 sheath tubing to specifications given in Appendix D.
Nondestructive Evaluation	Check sheath dimensions and visually inspect. Examine sheath walls with ultrasonic inspection test and reject sheaths with defects greater than 120 square mils (60 mils long by 2 mils deep). Determine wall thickness variation by eddy current test. Inspect surfaces with "Zyglo-XL22" dye penetrant for defects and reject sheaths with persistent indications. Cut sheaths to required length. (Note: All of these items can be done by the tube vendor.)
Etch	Degrease pieces in caustic solution and rinse. Remove .1 to .5 mils of Zircaloy in $HF-HNO_3$ acid bath; immerse in $Al(NO_3)_3-HNO_3$ stop bath; rinse in hot flowing water and deionized water, successively. Wipe with alcohol and store in polyethylene bags.
Weld Bottom Plug	TIG weld in dry box. (Use spacers to maintain sheath concentricity.)
Vibratory Compaction	Load oxide to 81-83% of theoretical density (for swaged tubes).
Weld Top Plug	
Vacuum Dry	Vacuum dry tubes at 275°C for 20 hours (through vent holes in temporary plugs), cool to about 40°C in vacuum, and backfill with helium gas.
Seal	Insert Zircaloy wire into vent holes and fuse to end plug.

DETAILS OF FABRICATION PROCESS, (cont'd)



Paint hot "Klingfast" (asphaltic gear lubricant) on mandrels and push mandrel into warm tube (80°C).

Lubricate exterior surfaces with palm oil and swage to 13% reduction in core area with two passes through swager. Measure tube-wall thickness.[15] Pull mandrel from tube with drawbench and stripping die.

Remove lubricant by immersing tube in liquid trichlorethylene.

Saw off two-inch ring sections for density samples; machine tubes to required length; counterbore UO_2 and insert chambered end plugs.

Weld permanent plugs to sheath in same equipment used to weld temporary plugs.

Insert Zircaloy wire and seal vent hole in each plug.

Inspect welds for leaks with helium leak detector; inspect tubes for gross leaks by bubble testing in ethylene glycol (pressure over liquid reduced to 60 torr); measure tube dimensions; radiograph weld beads for inclusions and voids; measure core density and wall-thickness variations with gamma densitometer; inspect surfaces with "Zyglo-XL22" dye penetrant.

Degrease tubes in trichlorethylene vapor. Wash in caustic solution; rinse in hot flowing water. Remove 1-1/2 mils from surfaces of swaged tubes and .1-.2 mils from vibrated tubes with $HF-HNO_3$ acid bath. Repeat procedure for rinsing given for initial etching.

Autoclave in steam for 48 hr at 400°C and 1500 psi pressure.

Visually inspect tubes for surface blemishes or signs of poor corrosion resistance. Measure tube dimensions; measure outer cladding thickness with eddy-current equipment.

Wipe tube surfaces with alcohol, and package in polyethylene for delivery to the reactor area.

APPENDIX B

NITROGEN CONTENT IN UO_2 FUEL TUBES[a]
(PRE- AND POST-IRRADIATION ANALYSES)

Assembly and Tube Number	∫kdθ watts/cm Average in time and space	Nitrogen Content, ppm				
		Preirradiation				Post-irradiation
		Kjeldahl		Vacuum Extraction (1300°C)		
		Avg.[b]	Range	Avg.[b]	Range	
SOT-1[c]						
ZE-222	17, 19	-	-	75[4]	66-93	61, 26
ZE-229	8	-	-	101[4]	76-170	<1
ZE-232	6, 18	-	-	98[4]	42-150	31, 121
SOT-2						
Z-250	12	-	-	55[2]	32-78	<1
Z-252	16, 35	-	-	35[2]	34-37	1, 14
Z-254	3, 41	-	-	178[5]	150-190	8, 29
Z-255	40	-	-	220[3]	210-230	<1
SOT-5 & SOT-7						
Z-260[d]	24, 25	29[6]	25-32	5[5]	1-8	<1, <1
Z-261[e]	14, 39, 49	-	-	-	-	<1, <1, <1
Z-262	14, 23	72[2]	61-83	36[6]	15-56	4, 4
Z-264[f]	49	101[5]	86-123	22[5]	4-53	2
Z-265	31	20[9]	13-28	<1[3]	-<1	16
SOT-6						
(3 crucible charges after outgassing)	-	95[10]	11-199	50[9]	<1-174	-

Both ϕ's and τ's should be reduced the same amount

NITROGEN CONTENT IN UO_2 FUEL TUBES[a]
(PRE- AND POST-IRRADIATION ANALYSES)
(cont'd)

Assembly and Tube Number	$\int kd\theta$ watts/cm Average in time and space	Nitrogen Content, ppm: Preirradiation: Kjeldahl: Avg.[b]	Kjeldahl: Range	Vacuum Extraction (1300°C): Avg.[b]	Vacuum Extraction (1300°C): Range	Post-irradiation
SOT-6, (cont'd)						
Z-272	22	-	-	-	-	6
Z-273	18 18	-	-	-	-	15 18
Z-278	22	-	-	-	-	18
SOT-8						
(4 tubes, as-fabricated)	-	70[4]	65-75	40[4]	37-42	-
ZE-289	19	85[1]	-	65[2]	37-93	18
ZE-290	26	80[1]	-	57[2]	42-72	16
ZE-297	29	-	-	-	-	10
SOT-9						
(2 tubes, as-fabricated)	-	-	-	71[6]	65-80	-
ZE-280	36	-	-	-	-	31
ZE-281	32	-	-	-	-	28
ZE-282	30	-	-	-	-	25

a. All tubes contain UO_2 vacuum outgassed 4 hours at 1300°C or greater unless otherwise noted.

b. Number of samples shown in parenthesis.

c. UO_2 outgassed 1 hour at about 1000°C.

d. UO_2 annealed in hydrogen 12 hours at 1750°C.

e. Hypostoichiometric UO_2 ($UO_{1.98}$); no thermal treatment.

f. UO_2 annealed in hydrogen 4 hours at 1300°C.

APPENDIX C

DIMENSIONAL SUMMARY OF UO_2 TUBES

OT-1 (Ten-foot-long)

Dimension, inches	Average	Range	Confidence Limits*	Number of Measurements
Vibratorily-Filled**				
Outer diameter	2.059	0.020	±0.008	180
Inner diameter	1.478	0.016	±0.005	60
Wall thickness	0.295	0.030	±0.016	206
Swaged				
Outer diameter	2.065	0.012	±0.005	180
Inner diameter	1.464	0.010	±0.004	60
Wall thickness	0.300	0.027	±0.016	186

SOT-6 (Five-foot-long)

Swaged				
Outer diameter	2.540	0.016	±0.005	410
Inner diameter	1.834	0.023	±0.010	252
Wall thickness	0.351	0.030	±0.010	312

SOT-8 (Five-foot-long)

Swaged				
Outer diameter	3.650	0.019	±0.008	238
Inner diameter	2.986	0.026	±0.012	270
Wall thickness	0.331	0.042	±0.016	320

* 2σ limits

** Dimensions of vibratorily-filled tubes are identical to dimensions of as-received tubing.

APPENDIX D

SPECIFICATIONS FOR ZIRCALOY-2 SHEATH TUBING

Dimensions

The diameter and wall thickness of the tubing shall be as specified in the purchase order. Lengths of 8 ft.-0 ± 1/8 in. are to be furnished. The maximum deviation from straightness shall be 0.080 inch in the eight-foot length.

Chemical Composition

The chemical composition of the finished tubing shall conform to the following limits:

Tin Percent	Iron Percent	Chromium Percent	Nickel Percent
1.20 - 1.70	0.07 - 0.20	0.05 - 0.15	0.03 - 0.08

The sum of the iron, chromium, and nickel contents determined from the average of all analyses made for a single ingot shall be within the range of 0.18-0.38%.

Impurity Content

The impurity content of the finished tubing shall not exceed the following limits:

Element	PPM	Element	PPM
Aluminum	75.	Magnesium	20.
Boron	0.5	Manganese	50.
Cadmium	0.5	Molybdenum	50.
Carbon	500.	Nitrogen	50.
Cobalt	20.	Oxygen	1200.
Copper	50.	Silicon	100.
Hafnium	200.	Titanium	50.
Hydrogen	25.	Tungsten	100.
Lead	130.	Vanadium	50.
		Uranium	5.

Should be same reduction as p. 79

Hardness

The average hardness of tubing after annealing shall not exceed a diamond pyramid hardness number (DPH) of 180.

Grain Size

The maximum average grain size shall not exceed 0.03 mm as measured by the applicable method described in ASTM Standard E 112-58T.

Finish

Tubing shall be pickled after the final forming operation and then fully vacuum annealed.

Marking for Identification

Each length of tubing shall be marked with the ingot melt number. Metal die stamping is prohibited.

Surface Quality

All tubing shall have a uniform outside surface that is free from cracks, blisters, seams, laps, pits, etc., and inclusions as revealed by visual examination.

Waivers

This material is for development purposes and deviations from some of the limits noted above may be permissible upon request. However, material that does not meet these specifications should not be shipped without prior approval by the purchaser.

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