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HW-33726(Rev.)

Metallurgy and Ceramics

AEC RESEARCH AND DEVELOPMENT REPORT

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H.C. # 5 no; MN 65

AN INVESTIGATION OF THE PROPERTIES OF ROLLED URANIUM RELATED TO THE QUENCH AFTER BETA HEAT TREATING

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By

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For The Atomic Energy Commission

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Fuel Technology Sub-Section

Chief, Declassification Branch

December 13, 1954

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AN INVESTIGATION OF THE PROPERTIES OF ROLLED URANIUM
RELATED TO THE QUENCH AFTER BETA HEAT TREATING

INTRODUCTION

The beta heat treating of uranium rod prior to the machining and canning of uranium fuel elements has some important advantages over triple-dip techniques which employ beta heat treatment during canning. Among these advantages are the possibility of inspecting beta heat-treated uranium prior to canning, the reduction in the slug-to-can tolerances for canning resulting in increased reactivity, and the overall lower cost of fuel element fabrication by lead-dip canning. In recent months, however, fuel elements manufactured from beta heat-treated rod and lead-dip canned have shown an in-pile rupture frequency that exceeds that of fuel elements triple-dip canned. One of the more frequent types of ruptures, cap failures, may be caused by the concave end-dishing of fuel elements in the piles. The amount of concavity reported for four slugs ruptured in the piles by cap failures ranged in depth from 0.04-inch to 0.08-inch at the base ends⁽¹⁾. This amount of concavity is in excess of that possible from stress relief alone, therefore orientation effects were thought to influence this behavior. Since for eight-inch slugs this type of rupture was first reported for PT-25-M material⁽²⁾, an investigation was made to determine whether or not the end dishing of slugs during pile operation could be traced to the beta heat treating in rod form.

In addition to cap failures, warping threatens to become an important problem with irradiation to higher exposure goals. Although the amount of warp has not been sufficient to seriously rupture slugs in the piles, it

- (1) HW-33084, "Interim Report on the Examination of Failures from Production Test - 313-105-25M", D. L. Zimmerman and W. V. Cummings, October 30, 1954, Secret.
- (2) A HAPO code for a certain pile test of slugs machined from FMPC rods alpha rolled and salt bath beta heat treated using a room-temperature water quench. The slugs machined from these rods were subsequently lead-dip canned at the HAPO.
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has sometimes been sufficient to restrict the ejection of irradiated slugs from pile tubes. Because such warp has been observed on several slugs within a tube, an explanation was sought that would relate the cause for warping to an effect common to a large percentage of slugs. Again the beta heat treating of rods was investigated for such an effect.

OBJECTIVE

The objectives of this work were to investigate the effects of the rate of quenching during beta heat treatment on the metallurgical characteristics of pre-irradiated uranium slugs and to determine if, by modifying the quench following the beta heat treatment of uranium rod, a sounder, more uniform fuel element could be produced which would have a lower pile-rupture rate.

SUMMARY AND CONCLUSIONS

Dimensional data obtained for beta heat-treated slugs quenched in different media indicate that a more effective dimensional recovery (essentially stress relief) during the beta heat-treating cycle can be obtained by using a slower quench than the room-temperature water bath presently employed at FMPC for HAPO rods.

Further, orientation data collected on beta heat-treated slugs both by sonic and x-ray methods suggest that the high (200) orientation present in beta heat-treated slugs recently shipped to HAPO can be reduced by selecting a less drastic quench. By modifying the present quench during beta heat treating, the zone of columnar grains common to PT-25-M⁽²⁾ material can be reduced without affecting the grain size of uranium.

Quenching uranium rod lengths horizontally on iron-hook supports was demonstrated to give an assymetric macrostructure at the hook locations due to uneven cooling across the transverse section. This practice might contribute to the warping of fuel elements in the pile. A support designed

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to make minimum possible contact with the rod and/or a quenching media with a less severe boiling stage may reduce this effect.

Cooling-curve data obtained with thermocouple slugs reveal a large variance in the cooling rate for uranium with the temperature of the water quench. Quenching from 730 C into a water bath between 20 C and 60 C allows the steepest part of the cooling curve to overlap the beta to alpha transformation. Water baths hotter than 60 C give a much slower cooling rate for uranium and should yield a uranium fuel element with fewer lattice defects and less cold work after beta heat treating.

DISCUSSION

The Nature of Quenching

A quench during a heat treating operation is intended to effect some change in the structure of a material in addition to appropriately removing heat from the material. In uranium, the end sought for is a random arrangement of fine alpha transformed-grains since such a grain structure is known to give good dimensional stability during both thermal cycling and irradiation. When an object at a high temperature is quenched in most media, liquid at room temperature, three independent stages of cooling exist⁽³⁾:

"A" stage - Immediately upon immersion of the hot object in the liquid a vapor blanket forms at the surface of the object separating it from the body of the liquid. Since heat is removed at the rate of the heat of vaporization of the liquid, cooling during this stage is slow. This stage lasts until the temperature of the object's surface is low enough to be wetted by the liquid.

(3) Grossmann, M. A., "Elements of Hardenability", American Society for Metals, 1952.

"B" stage - This stage commences with the liquid making momentary contact with the surface of the object. Bubbling occurs because the liquid vaporizes instantly; however, the surface is wetted by the liquid. Heat is removed most rapidly during this stage, since heat transfer occurs by both conduction and convection.

"C" stage - During this stage, the surface of the object has cooled below the boiling point of the liquid, and the transfer of heat is largely by convection currents within the liquid. If the material has a low thermal conductivity, it may enter the "C" stage of quenching with the center of the object still at a high temperature.

There are four major variables common to the media which effect the efficiency of the quench: (1) surface tension, (2) agitation, (3) temperature, and (4) boiling point. Increasing the surface tension of a liquid has the effect of depressing vapor formation during the "A" stage of quenching. For water, this is usually done by adding a salt which thereby increases the quenching rate of the water. Agitating the quench bath washes the liquid against the object's surface, thus breaking down the vapor blanket early in the "A" stage of quenching. Raising the temperature of the bath decreases the thermal gradient between the hot object's surface and the liquid, thus decreasing the rate of heat transfer. And, raising the boiling of the liquid diminishes the "A" state of quenching.

Cooling Rates for Water Quenches

The quench presently used at FMPC during the beta heat treatment of uranium rod is a mildly agitated, room-temperature water bath. An easy method to slow down this quench would be to increase the water bath temperature. To determine the cooling rates for uranium quenched in water baths at different temperatures; two four-inch thermocouple slugs were machined from a section of uranium rod, and were fitted with a thermocouple as illustrated in Figure 1. Thermocouple wells, 3/16-inches

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EDGE-WELL THERMOCOUPLE SLUG

CENTER-WELL THERMOCOUPLE SLUG

FIGURE 1

Components of the Thermocouple Slugs Used to Measure Cooling Curves for Uranium

in diameter and 1 1/2-inches deep, were drilled axially; 3/16-inches from the edge of one slug, and at the center of the other slug. A 20-gage, chromel-alumel thermocouple enclosed in glass fiber was beaded by welding and placed in a 3/16-inch stainless steel tube 2 1/2 feet long. This tube made a close fit in the thermocouple well and protected the thermocouple wires from direct contact with the molten salt in the heat-treating bath.

Figure 2 illustrates the recording apparatus used. The thermocouple wires from the slug were attached to a Brush amplifier which drove a Brush strip recorder with a chart speed of 1/10-inch per second and a pen speed of 60 cycles per second. To standardize the chart, a cold junction of ice and water in equilibrium at 0 C was used. Since the thermocouple bead could not be tamped or welded into the well of the slug, powdered tin was placed in the bottom of the well to act as a braze and to insure good heat conduction between the uranium and the thermocouple at high temperatures. Further, by using this type of a braze, the thermocouple could be readily exchanged between the two thermocouple-slugs.

To obtain time-temperature plots for uranium, an assembled thermocouple-slug was submerged in a molten chloride-eutectic-salt bath equipped with automatic temperature-control apparatus. A five-minute chart run was made from the instant of submersion, and a one-minute chart run was made after 11 minutes of submersion, at which time the slug and the bath were in thermal equilibrium. After heating for 13 1/2 minutes in the salt bath, the slug was removed and instantly quenched in a water bath with moderate agitation. The transfer time from the bath to the quench tank was about three seconds, and a five-minute chart run was made from the instant the slug hit the water. By using this procedure, the chart temperature calibration could be checked by three points of thermal equilibrium: (1) the alpha-beta thermal arrest for heating, (2) the salt bath temperature, and (3) the ice-water, cold-junction mixture. With these precautions, an accuracy of within 5 C degrees was obtained.

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Photograph of the Apparatus Used to Obtain Cooling Curves for Uranium Quenched in Water at Different Temperatures

Figure 3 is a series of cooling curves measured near the edge of a slug when quenched from 730 C into water baths at different temperatures. These curves plot the temperature of uranium against time in the quench tank, and indicate a logarithmic spacing among the cooling curves for different water-bath temperatures. The three stages of quenching are illustrated on the graph. For water between 20 and 60 C, the "B" stage of quenching probably overlaps the beta to alpha transformation, because super-cooling during a quench lowers the transformation temperature. It is further probable that part of the beta to alpha transformation during a quench in 80 C water takes place during the "A" stage of quenching. Since a decrease in volume occurs in uranium during the beta to alpha transformation, strain can occur if the uranium is quenched too rapidly through the transformation point. Further, any strain produced by rapid cooling below the recrystallization range can leave the uranium in a highly stressed condition. For these reasons, an increased water temperature during quenching may be desirable.

Figure 4 illustrates the difference in quenching rates between the center and the edge of a slug for two water-bath temperatures. The plots indicate that although water-bath temperatures between 20 and 50 C affect little change in the cooling rate at the edge of a slug, they affect a greater change in the cooling rate at the center of the slug.

Effects of Uneven Quenching

Water quenching during the heat treating of hardenable steel is often avoided because steam formation at the surface of the piece is irregular, resulting in uneven hardness after heat treating. When this situation occurs, either the bath is agitated more vigorously, an additive is made to the water, or another medium is selected. Agitating the bath will shorten the "A" stage of quenching by breaking down the steam pocket more rapidly; an additive, such as salt, will depress the formation of steam; and another medium, such as oil, will have a less vigorous vapor formation.

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FIGURE 3
COOLING RATES FOR URANIUM IN DIFFERENT TEMPERATURES WATER

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FIGURE 4

COOLING RATES AT THE CENTER AND EDGE OF SLUGS QUENCHED IN WATER BATHS AT TWO TEMPERATURES

Another way that the effects of uneven quenching occur is for a part of the hot object being quenched to come in contact with a heat conductor or an insulator. It was assumed, therefore, that quenching uranium rods horizontally on an iron-hook support might yield uranium with assymetric properties.

To confirm this assumption, three 9 1/2-inch rod sections, supported on an especially designed holder similar to hook supports previously used at FMPC, were beta heat-treated in molten chloride-eutectic-salt bath for 13 1/2 minutes at 730 C. The hooks on the holder, which were cut from 1/2-inch iron plate, made contact with the rod sections around half the rod circumference at about 2 1/2-inches in from each end. Quenching was done without agitation in a bath at room temperature. Figure 5 consists of a macrophotograph of a transverse wafer cut at the point of contact between the rod section and the hook, and a macrophotograph of a transverse wafer cut away from the hook. Apparently, the hot iron hook delays that quenching action of the water, and allows more grain growth to occur at the areas of hook contact. This effect was observed on all six of the wafers cut at the hook locations.

An uneven grain size across the section of a slug may cause uneven growth during irradiation and thus be a contributory cause for slugs warping in the pile.

Dimensional Change During Beta Heat Treatment

When a metal is heat treated and quenched, dimensional changes occur which depend upon the temperature of the heat treating bath, the prior texture and residual stress of the metal, and the rate of heating and cooling to mention a few.

To determine the effect of the quenching rate from the beta phase on the dimensional behavior of uranium slugs during heat treatment, 8-inch, as-rolled slugs from comparable rods were given the same beta heat

PHOTOGRAPH UNCLASSIFIED

MACROETCHED TRANSVERSE SECTION OF BETA HEAT-TREATED URANIUM ROD SHOWING THE INFLUENCE OF THE HOOK SUPPORT ON THE GRAIN SIZE (SEE ARROWS), AND ON THE DISPLACEMENT OF THE LARGER-GRAINED CORE. PHOTOGRAPH IS ORIENTED WITH THE HOOK AT THE BOTTOM AND THE HOOK SUPPORT AT THE RIGHT.

Standard Macroetch, Bright Field (2.1 X)

PHOTOGRAPH UNCLASSIFIED

MACROETCHED TRANSVERSE SECTION OF BETA HEAT-TREATED URANIUM ROD SHOWING A SYMMETRICAL MACROSTRUCTURE.

Standard Macroetch, Bright Field (2.1 X)

FIGURE 5

treatment and quenched in different media. Heat treating was done in a molten chloride-eutectic-salt bath at 730 C for a period of 13 1/2 minutes. The quenching baths used are listed in Table I. Length, warp, and dish (end concavity) measurements were made before and after beta heat treating, and the change in these properties are also listed in Table I. There was essentially no warp or dish on the slugs prior to beta heat treating.

Referring to Table I, the amount of length reduction that occurs during beta heat treating varies with the rate of quenching. As-rolled slugs usually shrink during beta heat treating because the orientation prior to heat treatment restricts the thermal expansion upon heating, whereas the near randomness after heat treatment allows normal contraction upon cooling. However, rapid quenching will allow less contraction than slow quenching because dimensional recovery is restricted by the increasing stiffness of uranium at temperatures below the transformation point. A slug may recover more of its contraction by stress relief during thermal cycling⁽⁴⁾.

Referring again to Table I, the amount of dishing that occurs at the end of a slug also varies with the rate of quenching; however, this variance is not large among the quenches used. With rapid cooling, a steep temperature gradient exists from the surface to the center of a slug which causes the edges and ends to become rigid before the central core. During cooling, thermal contraction in the core of the slug puts the ends in axial tension causing them to dish slightly before they cool to room temperature. If the ends become rigid too soon, voids may exist in the core⁽⁵⁾. Dishing becomes less pronounced with the slower quenches because the thermal gradient becomes less steep, and more uniform recovery occurs across the section and along the length of the slug.

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- (4) HW-33622, "A Preliminary Investigation of the Effect of Quenching Rates on the Deformation, Orientation, Macrostructure, and Grain Size of HAPO Slugs", November 3, 1954.
- (5) HW-28305, "Report on Thermal Cycling of Hanford Uranium Slugs Across the Alpha-Beta Transformation Temperature", June 1, 1953.

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TABLE I
THE CHANGE IN THE LENGTH, WARP, AND DISH OF URANIUM SLUGS
DURING BETA HEAT TREATMENT WITH DIFFERENT QUENCHES

Type of Quench	Change in Length (mils)			Change in Dish (mils)			Change in Warp (mils)			No. of Slugs
	High	Low	Avg.	High	Low	Avg.	High	Low	Avg.	
20 C Water ⁽⁴⁾	-3	2	-2	9.0	4.0	7.0	15	5	11	5
55 C Water	-3	2	0	11.0	2.1	5.5	14	5	10	5
33% Glycerine Solution (vol)	-6	-3	-4.4	6.9	3.5	5.2	12	5	10	5
5% KCl Solution (wt)	-6	-3	-4.3	8.2	3.0	6.6	16	10	14	5
100 C Quenching Oil	-19	-12	-15.2	4.2	1.2	2.9	23	9	16	5
100 C Water	-20	-12	-15.6	7.0	0	2.8	26	8	16	5
90 Sec. Air Delay Followed by 20 C Water	-32	-24	-28.4	7.5	1.0	3.1	26	5	15	5

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Dish is the maximum concavity across the slug end.

Warp is double throw.

A 90 sec. air delay provides beta to alpha transformation in air. This step is followed by a room-temperature water quench.

(4) HW-33622, "A Preliminary Investigation of the Effect of Quenching Rates on the Deformation, Orientation, Macrostructure, and Grain Size of HAPO Slugs", November 3, 1954.

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There is no clear correlation of warp with the quenching rate. A combination of causes probably occur simultaneously and give scattered data for each quench. Among these causes are assymetric residual stress or preferred orientation in the as-rolled material causing warp to occur upon heating, and assymetric stress or recovery due to uneven quenching causing warp to occur upon cooling.

The Effect of Quenching Rates on Hardness

In many metals which are effectively strain hardened, the change in hardness values with the quenching rate is a partial measure of the strain and hence the lattice distortion induced by the quench. Uranium, of the purity presently specified for production, strain hardens over a narrow range; therefore, such indications are hard to observe. However, measurements made on one slug from each of the quench groups listed in Table I resulted in a relationship between the quenching rate and the resulting hardness of uranium.

Transverse wafers were cut near the center of the selected slugs. The surface of each wafer was then lightly ground to reduce any cold-worked layer that might have resulted from the cutting operation, and to give a smooth, uniform surface for hardness measurements.

Readings were then taken around the periphery of the flat wafer face, between 1/8 and 3/8-inches from the edge, with a Rockwell Hardness Tester using the "G" scale (1/16-inch ball indenter with 150 lb. load). The hardness values obtained are presented in Table II. These data indicate a decrease in the hardness of beta heat-treated uranium with a decrease in the quenching rate.

After these initial hardness readings were taken, the wafers were annealed for three hours at 400 C in an argon atmosphere, cooled in the furnace, and measured again for hardness near the initial indentions.

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These data, presented in Table II, represent a very slight, and perhaps insignificant, decrease in hardness with the anneal and again indicate a decrease in the hardness of beta heat treated uranium with a decrease in the quenching rate.

TABLE II
THE VARIATION IN THE HARDNESS OF URANIUM
WITH THE QUENCH DURING BETA HEAT TREATING

<u>Type of Quench</u>	<u>Hardness before Anneal*</u>			<u>Hardness after Anneal</u>		
	<u>Range for</u> <u>8 Readings</u>	<u>Average</u>	<u>σ</u>	<u>Range for</u> <u>8 Readings</u>	<u>Average</u>	<u>σ</u>
55 C water	71 - 80	75.9	1.3	72 - 77	74.3	0.33
33% glycerine	72 - 76	74.0	0.36	72 - 76	73.8	0
5% KCl	70 - 75	73.1	0.4	71 - 75	73.6	0.93
100 C oil	70 - 77	73.5	0	68 - 75	70.9	0.92
90 sec delay	65 - 75	71.0	1.4	65 - 77	71.0	0.53
100 C water	66 - 73	69.9	0.5	66 - 75	69.2	1.6

σ = standard deviation calculated by $\sigma = \sqrt{\frac{\sum (x_i - \bar{x})^2}{v}}$

*Hardness readings were taken with a Rockwell Hardness Tester using the "G" scale (1/16" ball indenter with 150 lb. load)

x_i = value

$\bar{x}$ = average value

n = number of values

v = n for n < 7

v = n-1 for n $\geq$ 7

The Effect of Quenching Rates on Orientation

A large percentage of beta heat-treated rods recently shipped to Hanford from FMPC have shown by x-ray methods a high (200) pole orientation along the axis of the slugs. These slugs are from rods quenched in room-temperature water during beta heat treating. Further x-ray data taken on IQS-9 material⁽⁶⁾ before and after heat treatment support these findings and Table III list the significant x-ray rho values⁽⁷⁾ for three slugs fabricated from this material. These data were taken from rotated shots at the center and near the edge of transverse wafers cut from the middle of the slugs.

The data in Table III reveal an increase in the (200) orientation with beta heat treating in five readings out of six with the edges having larger rho values than the centers. This may result from the thermal gradient in quenching being steeper at the edge than at the center of the rod; however, since rolling orientations are more pronounced at the edge than at the center of the rod, a memory effect may also be represented.

To observe if a variation in the cooling rates during quenching could effect the (200) orientation, the 30 slugs in Table I, (excluding these slugs quenched in 20 C water) given different quenches, were measured for orientation by sonic techniques⁽⁸⁾ before and after beta heat treatment. By this method of measuring orientation, the whole slug is placed vertically on a driver which vibrates the slug at sonic frequencies. A pick-up and a counter circuit receive the pulse at the other end of the slug and translate

(6) An ingot-quality study designation referring to material from 7-inch hot-topped ingots cast at both MCW and FMPC, and rolled and beta heat treated at FMPC.

(7) A rho value is ratio of the intensity of (hkl) poles in the material measured to the intensity of the sample poles in a standard powder sample HW-26762, "A Rapid Method for Determining Preferred Orientation in Rolled Uranium Rods", January 7, 1953.

(8) A test developed by E. C. Wood and A. A. Ferguson at HAPO.

TABLE III

ORIENTATION RHO VALUES FOR IQS-9 MATERIAL BEFORE AND AFTER
BETA HEAT TREATMENT WITH A COLD WATER QUENCH AT FMPC

(HKL)	Position of X-ray Shot on Wafer Cut from Center of Slug	Slug 657		Slug 682		Slug 5319	
		Before	After	Before	After	Before	After
200	Center	1.17	2.40	1.56	2.13	1.99	2.63
	Edge	4.34	5.27	4.25	5.32	4.24	2.67
020	Center	1.61	0.83	2.09	1.33	1.83	1.08
	Edge	1.81	0.92	2.03	1.42	1.79	1.08
110	Center	1.56	2.37	1.81	2.00	2.11	1.38
	Edge	2.37	1.97	2.67	1.85	2.35	1.61
040	Center	1.96	0.91	2.11	1.17	1.82	0.91
	Edge	2.24	1.27	2.12	1.36	2.42	1.27
041	Center	2.09	0.72	2.03	1.15	1.80	0.82
	Edge	1.65	0.89	1.00	0.80	1.40	0.82
002	Center	0.10	0.28	0.21	0.47	0.12	0.77
	Edge	0.13	0.53	0.10	0.60	0.12	0.97

rho value

0.76 - 1.5

1.6 - 2.0

2.1 - 2.9

3.0 - 4.5

4.6 -

Description of Orientation

Random

Trace

Slight

Definite

Pronounced

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the resonant frequencies of the longitudinal, flexural and torsional modes of vibration. The fundamental longitudinal mode of sonic frequency was used to compare the orientation influences of uranium. Although all of the slugs prior to beta heat treatment grouped together on the frequency scale, the slugs after beta heat treatment formed separate groups having different frequencies. These groups spread along the frequency scale according to the quench, as illustrated in Figure 6. The amount of orientation varies inversely with the resonant frequency,⁽⁹⁾ therefore, the data represent an increase in orientation with the severity of quench.

The Effect of the Quench on the Macrostructure and Grain Size of Uranium

There are normally two distinctly different zones of grain orientation in the transverse macrostructure of PT-25-M material, as illustrated by Figure 7. The structure on the outer circumference consists of columnar grains probably resulting from the preferred growth of nucleated alpha grains during the beta to alpha transformation, and the central zone consists of equiaxed grains resulting from random grain growth during the quench.

The transverse macrostructures of slugs given the different quenches listed in Table I (excluding the 20 C quench) are described in Table IV and are illustrated in Figures 8 through 13. Grain determined from the microstructure both at the center and 1/8-inch from the edge of transverse wafers are also listed in Table IV. These grain size data indicate that away from the immediate slug surface, grain growth during cooling can be arrested equally by a variety of quenching media having a range of cooling rates for the three stages of quenching.

(9) Private communication with E. C. Wood and A. A. Ferguson.

TABLE IV
GRAIN SIZES AND DESCRIPTION OF MACROSTRUCTURE FOR
BETA HEAT-TREATED SLUGS GIVEN DIFFERENT QUENCHES

<u>Figure</u>	<u>Type of Quench</u>	<u>Grain Size (mm)*</u>		<u>Description of Macrostructure in Transverse Sections</u>
		<u>Center</u>	<u>Edge</u>	
9	55 C Water	0.150-0.169	0.150-0.169	Two zones structure - Strong columnar grained zone and random-grained zone**
10	33% Glycerine Solution	0.150	0.134	" " "
11	5% KCl Solution	0.134-0.150	0.150	" " "
12	100 C Oil	0.118-0.134	0.134	Very little columnar-grained zone - very even-grained appearance.
13	100 C Water	0.118	0.134	Random-grained
14	90 Seconds Air Delay	0.150-0.169	0.169	Random-grained

* Determined on the microstructure by comparison with Battelle grain-size charts for uranium.

** Showing no particular arrangement.

Sonic Resonant Frequency of Vibration - Longitudinal Mode

FIGURE 6

Sonic Resonant Frequency of Vibration in the Longitudinal Mode
for Slugs from Alpha-Phase-Rolled Rod Given Different Quenches

Photograph Unclassified

FIGURE 7

Macroetched Transverse Section of a Slug Fabricated from PT-25-M Rod, Beta Heat-Treated for in a K-Li-Carbonated Salt Bath for 13.5 Minutes at 720-740 C and Quenched in a Room-Temperature Water Bath

Standard Macroetch, Bright Field (3X)

Photograph Unclassified

FIGURE 8

Macroetched Transverse Section of a Beta Heat-Treated Uranium Slug Quenched in a Water Bath at 55 C Showing a Two-Zoned Structure Prevalent in PT-25-M Uranium Slugs

Standard Macroetch, Bright Field (2.1X)

Photograph Unclassified

FIGURE 9

Macroetched Transverse Section of a Beta Heat-Treated Uranium Slug Quenched in a 33% Glycerine Solution (Vol) at Room Temperature Showing a Two-Zoned Structure Prevalent in PT-25-M Uranium Slugs

Standard Macroetch, Bright Field (2.1X)

Photograph Unclassified

FIGURE 10

Macroetched Transverse Section of a Beta Heat-Treated Uranium Slug Quenched in a 5% KCl Solution (Wt) at room Temperature Showing a Two-Zoned Structure Prevalent in PT-25-M Uranium Slugs

Standard Macroetch, Bright Field (2.1X)

Photograph Unclassified

FIGURE 11

Macroetched Transverse Section of a Beta Heat-Treated Slug Quenched in an Oil Bath at 100 C Showing a Weak Zone of Columnar Grains and an Even-Grained Appearance

Standard Macroetch, Bright Field (2.1X)

Photograph Unclassified

FIGURE 12

Macroetched Transverse Section of a Beta Heat-Treated Slug Transformed from the Beta to the Alpha Phase in Air (90 Sec) Followed by a Quench in a Water Bath at Room Temperature. The Grain Structure Shows No Particular Arrangement.

Standard Macroetch, Bright Field (2.1X)

Photograph Unclassified

FIGURE 13

Macroetched Transverse Section of a Beta Heat-Treated Uranium Slug Quenched in a Water Bath at 100 C Showing a Complete Absence of Columnar-Type Grains.

Standard Macroetch, Bright Field (2.1X)

The Effect of the Quench on the Characteristics of Uranium During Thermal Cycling

The object of this test was to determine to what extent thermal cycling in the alpha range would affect the dimensions, macrostructure, and orientation of beta heat-treated slugs quenched in water baths at different temperatures.

The apparatus used for this test employs simultaneous induction heating and water cooling to simulate in-pile temperature fluctuations within a uranium slug⁽¹⁰⁾. The test slug is placed in a 40-mm glass tube which is in turn oriented coaxially within an induction coil. The power source is a 200 KW, 3000 cps Tocco motor-generator, and the water flow within the glass tube is about 30 gpm. During the heating cycle the slug core reaches a maximum temperature of about 640 C while the slug surface is cooled by the water to about 50 C. During the cooling cycle, the power is turned off, and the slug core is cooled by the water to about 30 C⁽¹¹⁾.

The material selected for this test was two slugs from each the 55 C water and 100 C water quench groups listed in Table I. These slugs were given 20 thermal cycles; each cycle consisting of 10 minutes with the power on, and 2 minutes with the power off, allowing the core temperature to fluctuate between 610 C and 30 C. During the test, one slug from the 55 C water quench group was vibrated in the glass tube by the flowing water. This vibration broke the glass tube, and the slug melted from the induction current. The dimensional changes that occurred on the remaining slugs during thermal cycling are presented in Table V.

(10) HW-31766, "Preliminary Report of Uranium Core Failures Produced by Simulated Pile Thermal Cycling", May 20, 1954, SECRET.

(11) Thermocouple readings have indicated that it takes about 30 seconds for the slug to reach equilibrium temperature both heating and cooling.

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TABLE V
DIMENSIONAL CHANGES IN DIFFERENTLY QUENCHED BETA
HEAT-TREATED SLUGS DURING THERMAL CYCLING

<u>Rod No.</u>	<u>5958-1</u>	<u>5955-1</u>	<u>5958-1</u>	<u>6815-1</u>
Type of quench	55 C water	55 C water	100 C water	100 C water
Length change during beta heat treating (mils)	(burned)	-2	-12	-18
Length change during thermal cycling (mils)		-75	-67	-58
Total length change (mils)		-77	-79	-76
Warp during beta* heat treating (mils)		5	26	20
Total warp after thermal cycling (mils)		10	20	20
Dish during beta heat treating (mils)		8.0	8.5	1.5
Total dish after thermal cycling (mils)		7.2	7.3	4.5

* Dish is the maximum concavity across slug end. Warp is double throw.

Referring to Table V, the number of samples used for this survey-type test were too few to indicate any trends in dishing or warping behavior; however, the length changes observed on the slugs indicate a trend that should be further investigated. According to these data, the slug that contracted the least during beta heat treating, that given the 55 C water quench, relaxed the most during thermal cycling; and those slugs that contracted the most during beta heat treating, those given the 100 C water quench, relaxed the least during thermal cycling. This trend indicates higher axial residual compressive stresses in the 55 C water-quenched slug than in the 100 C water-quenched slugs. These data, therefore, suggest that the more dimensional recovery realized in the quench during beta heat treating, the less dimensional relaxation will occur during thermal cycling in the piles.

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The surfaces of the slugs, as they appeared after thermal cycling, are illustrated in Figures 14, 15, and 16. These photographs resolve the cylindrical surface of the slug into a plane surface for clearer observation. For each photograph, the slugs were rotated 1-1/2 times, therefore, identical surface markings must be used for fiducial points.

Figure 14 represents the surface of the 55 C water-quenched slug, number 5955-1, as it appeared after 20 thermal cycles. Striations on or just below the surface opened into small cracks which are oriented axially with the slug. Had the slug been given a larger number of thermal cycles, a split-type rupture originating from these cracks might have occurred.

Figures 15 and 16 represent the surfaces of the 100 C water-quenched slugs, numbers 5958-1 and 6815-1, respectively, as they appeared after thermal cycling. Numerous dome-like eruptions occurred on the surface, the majority of which are on the downstream end of the slug. One cause for this type failure is thought to be small casting voids near the periphery of the slug which were not welded during rolling⁽¹²⁾. However, a comparison of these surfaces with that of the 55 C water-quenched slug suggests that a larger grain size at the surface could also be a cause. Finally, the presence of corrosion products within these eruptions reveals that after the eruptions had occurred on the surface, corrosion of the uranium by the circulating water exaggerated their size.

After the slugs were photographed, three transverse wafers cut from each of the three slugs were examined by x-ray techniques for orientations. The wafers were cut one at the center and one at one inch in from either end of each slug, and rotated shots were taken normal to the transverse face at the center and near the edge of the wafer. Table VI lists the rho values for the (200) poles, before and after thermal cycling, according to the quench during beta heat treating. The orientations of the other poles in the wafer were essentially random and had little relation to the quench.

(12) HW-31766, "Preliminary Report of Uranium Core Failures Produced by Simulated Pile Thermal Cycling", May 20, 1954, SECRET.

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FIGURE 14

Layout Photograph of a Slug's Surface After Thermal Cycling in the Alpha Range for Twenty Cycles. Slug Was Quenched During Beta Heat Treating in a Water Bath at 55 C. Magnification is (0.7X)

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FIGURE 15

Layout Photograph of a Slug's Surface After Thermal Cycling in the Alpha Range for Twenty Cycles. Slug Was Quenched During Beta Heat Treating in a Water Bath at 100 C. Magnification is (0.7X)

FIGURE 16

Macroetched Transverse Section of a Beta Heat-Treated Slug
Quenched in an Oil Bath at 100 C Showing a Weak Zone of Columnar
Grains and an Even-Grained Appearance

TABLE VI
RHO VALUES FOR THE (200) POLES IN
BETA HEAT TREATED AND DIFFERENTLY QUENCHED URANIUM,
BEFORE AND AFTER THERMAL CYCLING

Description of Slug	Wafer from No. End		Wafer from Center		Wafer from other End	
	Center Shot	Edge Shot	Center Shot	Edge Shot	Center Shot	Edge Shot
Before Thermal Cycling						
55 C water						
Quench(avg)	--	--	3.1	1.8	3.2	2.4
100 C water						
Quench(avg)	--	--	0.5	1.6	2.5	1.3
After Thermal Cycling						
55 C water						
Quench	4.8	1.8	3.3	2.9	3.5	2.0
100 C water						
Quench	2.3	1.8	0.5	1.9	2.3	1.4
100 C water						
Quench	1.2	1.9	1.6	1.0	1.4	1.1

<u>Rho Value</u>	<u>Description of Orientation</u>
0.76 - 1.5	Random
1.6 - 2.0	Trace
2.1 - 2.9	Slight
3.0 - 4.5	Definite
4.6 -	Pronounced

The rho values listed in Table VI reveal two effects of interest: (1) the rho values for the 55 C water quenched slugs range from a trace to a pronounced (200) orientation, whereas the rho values for the 100 C water quenched slugs range from random to slight (200) orientation; and (2) the thermal cycling given the slugs does not effect the magnitude of (200) orientation. Most of the edge shots revealed a lower (200) orientation than the center shots; however, the inverse was observed on the IQS-9 material represented in Table III.

The increase of orientation with the quench can be observed in sonic data, where again the more severe quench gave the higher value for orientation. A supposition based on cooling-curve data and orientation data taken before and after thermal cycling would correlate such a mechanism to a polar alignment, according to anisotropic properties, influenced by steep thermal gradients⁽¹³⁾. This follows because the thermal gradients within a slug become steeper when the quench during beta heat treating is made more severe. Further, since such a mechanism is observed to exist for uranium, uneven quenching during beta heat treating could not only result in assymetric macrostructures, but could also result in large orientation variances across the section and along the length of uranium rods; and thus, in this way, be a cause for in-pile slug warping.

CONCLUSIONS AND RECOMMENDATIONS

The data collected during this survey investigation demonstrates that the quenching step during the beta heat treating of rolled uranium has a marked effect on the metallurgical characteristics of uranium that influence pile stability of fuel elements. By quenching rolled uranium in water at temperatures below 60 C with agitation, the beta-alpha transformation is suppressed so that it probably occurs during the most rapid

(13) Investigations by O. K. Shupe, J. C. Bokros, and A. L. Bement at HAPO reveal that the increase in (200) orientation in heat treating is probably a function of the heat treatment rather than of rolling history.

stage of quenching, thus greatly arresting stress relief by dimensional recovery. The utilization of a slower quench; a water bath at a higher temperature, another medium, as isothermal quench in an elevated-temperature bath, or a delay quench in air; should reduce the lattice-strain that occurs after rapid quenching and thus yield a sounder fuel element with a more uniform structure.

The extent of in-pile dishing that occurred at the ends of PT-25-M⁽²⁾ fuel elements can be explained if a variance in the preferred orientation of (200) poles existed across the transverse section of the slugs⁽¹⁴⁾. The orientation of (200) poles in beta heat treated uranium appears to be dependent upon the rate of quenching during beta heat treating. The intensity of this orientation has been shown by both x-ray and sonic techniques to increase with more rapid quenching.

Finally, the zonal texture that occurs in PT-25-M material in which a pronounced zone of columnar grains exists at the periphery of uranium slugs can be minimized or eliminated by reducing the quenching rate during beta heat-treating. The grain size of beta heat-treated uranium given a slower quench remains constant over wide limits; these grain sizes being acceptable under present HAPO specifications.

The data presented in this report represent roughly 60 per cent of the material investigated by the author for these tests. As yet, some of the indications presented in this report have not been subjected to sufficient research; therefore, conclusions that appear firm should be substantiated by further investigation. Typical of the problems requiring further investigation are the extent to which the rate of quenching effects the orientation

(14) HW-33622 "A Preliminary Investigation of the Effect of Quenching Rates on the Deformation, Orientation, Macrostructure, and Grain Size of HAPO Slugs", November 3, 1954.

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of beta heat-treated uranium, the difference between rod heat-treating and slug heat-treating on the in-pile dimensional behavior of uranium fuel elements, and the quenching effects on the metallurgical characteristics of uranium fuel elements fabricated by alpha extrusion, gamma extrusion, and powder compaction.

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ABSTRACT

Data are presented which show that quenching noticeably affects the dimensional change, hardness, orientation, and macrostructure of rolled, beta heat-treated uranium. The warping of slugs during in-pile operation might be traced to uneven quenching techniques. Finally, cooling curves for quenching uranium slugs in water show a marked difference in the quenching rate with the water temperature. (auth)

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