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From: B. R. Hayward

Subject: NOTES FROM TALK ON DIMENSIONAL CHANGES IN URANIUM GIVEN ON SEPTEMBER 19, 1951

ABSTRACT

It was the purpose of this talk to present up-to-date information with regards to dimensional stability of metallic uranium. A brief discussion concerning the history, present status, proposed theories, and recent results are given. The factors affecting, and methods of minimizing dimension changes are outlined.

INTRODUCTION

Early work by British personnel on non-cubic materials showed that dimensional changes from thermal cycling occur in elements exhibiting anisotropic properties. The following table indicates their results:

TABLE I

Element	Crystal Structure	Distortion from Thermal Cycling
Sn	Body Centered Hexagonal	Yes
Zn	Hexagonal Close Packed	Yes
Cd	Body Centered Tetragonal	Yes
Pb	Face Centered Cubic	No

Uranium is highly anisotropic. Alpha uranium has an orthorhombic crystal structure, β uranium has a tetragonal structure, and γ uranium is body centered cubic. The phase transformations occur as follows: $\alpha < 660°$ C, $\beta > 660° < 770°$ C, $\gamma > 770° < 1130°$ C. The coefficients of expansion in the a, b, and c axis are: $a = 23 \times 10^{-6}$ cm/cm °C, $b = -3.5 \times 10^{-6}$ cm/cm °C, $c = 17 \times 10^{-6}$ cm/cm °C at room temperature. The volume coefficient of expansion of α uranium at 20° C = 39×10^{-6} cm/cm °C and at 625° C = 55×10^{-6} cm/cm °C.

Thermal cycling, as used here, refers to the oscillation from one temperature to another under operating conditions, i.e., shutdown, reloading, and start-up constitutes one thermal cycle.

Distortion in the following discussion refers to longitudinal growth unless otherwise noted. All general statements refer to thermal cycling within the α region unless otherwise noted.

EXAMPLES OF DIMENSIONAL STABILITY PROBLEMS

Hanford

Briefly, the 4 inch slugs at Hanford have shown a number of types of dimensional changes: (1) surface blistering due to uranium-aluminum reaction, and surface roughening due to growth of large uranium grains on the slug surface; (2) thermal cycling of α-rolled slugs causes longitudinal growth; (3) radiation damage has indicated slug shortening; and (4) some slugs have warped enough to become stuck in the fuel tubes.

Chalk River

These rods are about 10 feet long suspended vertically from the top. The principal distortion noted was the decrease in length with a corresponding increase in diameter of the rods. As much as a 5 inch decrease in length occurred. Some rods fattened sufficiently to become stuck in the fuel channel.

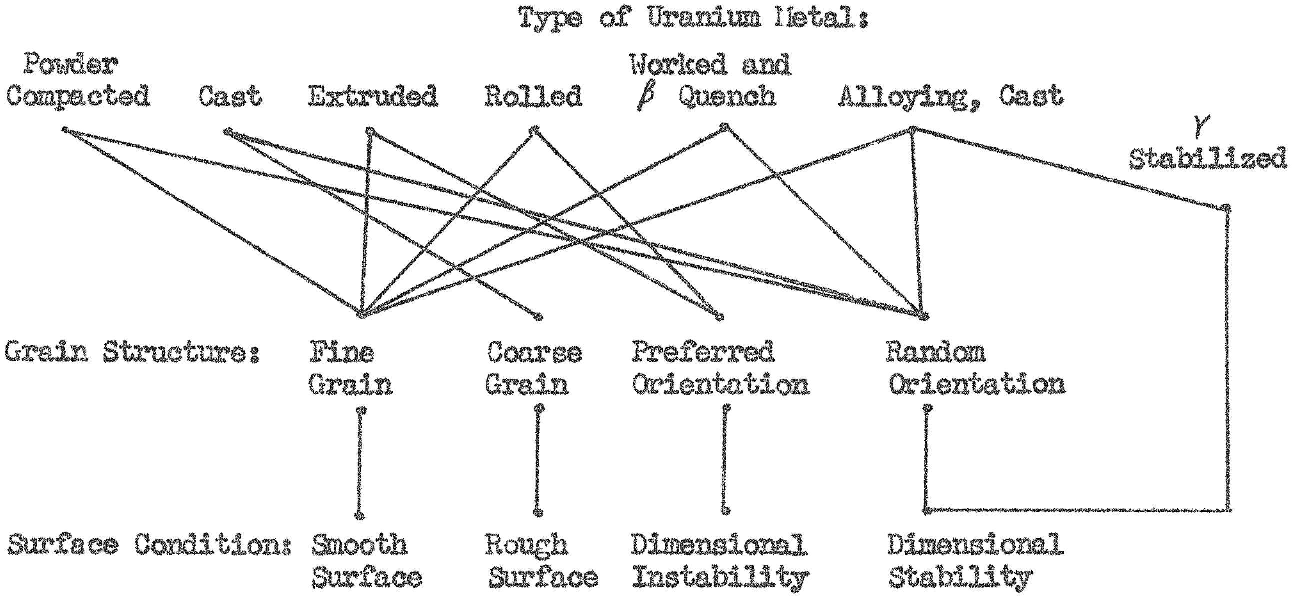
MTA

Thermal cycling is a great problem for some designs in this program. As many as 100,000 cycles are estimated.

PuP

This reactor under development expects to use higher slug temperatures than are now in operating existence. It plans to have one 18 foot slug with large radial and axial thermal gradients. Under consideration is the effect of having two phases present within the slug. Warp is one of the most important problems in this reactor.

GENERAL METAL FEATURES ASSOCIATED WITH DIMENSIONAL STABILITY



FACTORS AFFECTING DISTORTION IN URANIUM

1. Preferred Orientation
2. Grain Size
3. Temperature Level
4. Temperature Difference
5. Temperature Gradient
6. Rate of Thermal Cycling
7. Radiation
8. Assembly of Fuel Element

NOTE: Although the core material is not mentioned as a factor, it is directly related to preferred orientation and grain size.

DISCUSSION

Preferred Orientation and Grain Size

The most desirable grain structure for maximum dimensional stability is a fine grained and randomly oriented condition. Random orientation of the grains is obtained by three techniques: (1) by hot pressing uranium powder to high density, a fine grained, randomly oriented slug is obtained; (2) a cast structure has randomly oriented grains; and (3) a complete transofrmation into the β phase removes all prior thermal and fabrication history and randomizes the grains. This technique is better than No. 1 and No. 2 as it provides for much more versatile methods of fuel element fabrication and assembly, i.e., an 18 foot bonded and jacketed slug for PuP. Grain refinement is accomplished through (1) mechanical working with recrystallization, (2) rapid quenching from the β phase, (3) by alloying additions, and (4) powder compacting. Alloying gives the most uniform and stable condition of grain refinement. Fine-grained material provides a smooth surface and coarse-grained material produces a rough surface as a result of thermal cycling.

It is desirable to rapidly cool from 725° C to produce fine α grains. A β anneal with pure uranium produces very large α grains. [illegible]ters of fine grains produce the same effect as coarse grains. Quenching is not always necessary with alloyed uranium as some alloying additions provide a stable fine-grained structure. There are a number of alloys which refine the grains, but applications of interest need a stable structure with low total neutron capture cross section. The alloys appearing most promising are one atom per cent chromium, up to 10 atom per cent zirconium, one atom per cent molybdenum, and one atom per cent columbium. Of these, the chromium and zirconium are at present undergoing extensive tests.

The largest grain size grown from the α phase in pure uranium is 0.075 mm. The one atom per cent Cr alloy has a grain size of about 0.040 mm.

Another technique for grain refinement is powder compacting. In this technique, through careful control of uranium-hydride preparation and subsequent reduction to powdered metal, a fine-grained material is obtained. This powder is hot pressed and jacketed in one operation at 600° C to a slug of high density, 18.7 g/cc, with a similar structure (fine grain, random orientation) to the β-quenched material. This technique is presently limited to short sections (4-12 inches long).

Since the γ phase is a cubic structure, an alloy which retains this phase at room temperature would be highly desirable. The only alloy which appears to do this is a high percentage Mo-U alloy.

Distortion can be minimized to about one-half of one per cent or less per 100 cycles through the above methods.

Temperature Level

The term temperature level refers to relatively equal temperature differences at varying temperatures, i.e., 500°-550° C versus 300°-350° C. Generally, as the temperature level is raised, the distortion increases. It is important to note that distortion also occurs with a large number of cycles of small temperature difference when the grains have highly preferred orientation but little change with random orientation.

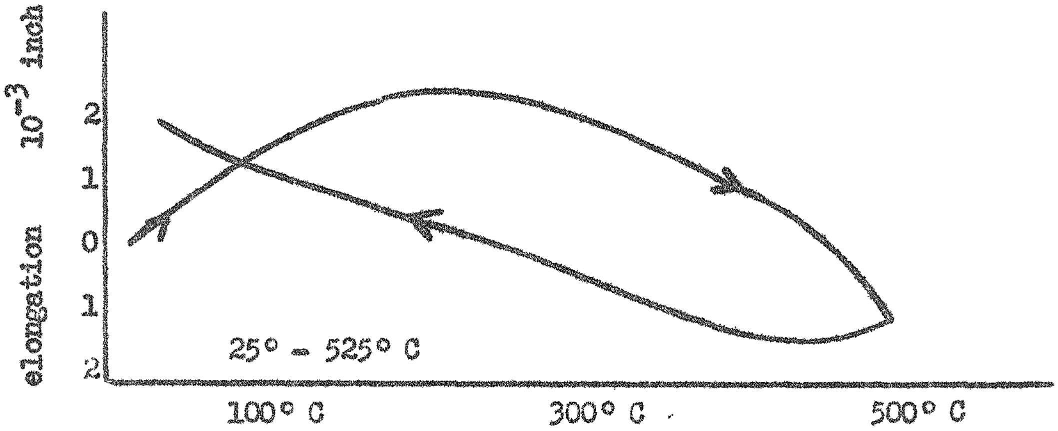
The effects of thermal cycling through the β phase have been briefly investigated. Some cycling tests have been made between 90° C and 1090° C. The distortion was large with pure uranium and decreased sharply with uranium-molybdenum alloys, the major changes being flaring at the ends and an increase in overall length. No heat treatment was found which would produce dimensionally stable pure uranium under cycling conditions of low α to γ temperatures. The U-Mo alloy of fairly low Mo concentration shows promise. However, other problems become evident under these conditions; i.e., (1) voids between the case and jacket with accompanying lower thermal conductivity, and (2) reactions between the case, jacket, and barrier or bond material. The role of the recrystallization temperature, about 400° C, in thermal cycling is in doubt. Table I shows actual dilatometer curves involving dimensional change. Note that a large percentage of the growth occurs during the cooling phase of the thermal cycle.

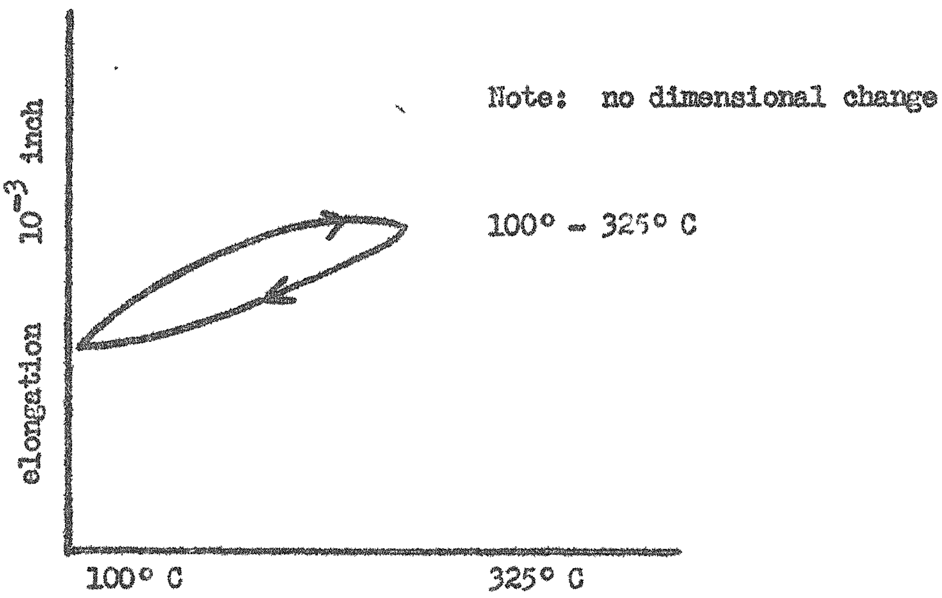
Temperature Difference

Distortion is directly proportional to the temperature difference, i.e., 0°-400° C versus 0°-600° C. Battelle has shown that a large number of small fluctuations ± 4° also produces growth. Very little information is available including temperatures over 600° C. With a constant temperature difference and an increase in temperature level, the distortion increases.

TABLE I

300° C Rolled, α Annealed
Dilatometric Curves





from BMI-P-35

Temperature Gradient

This effect is not completely understood. It is very important when more than one phase is present, i.e., α and β uranium in both axial and radial gradients. The physical properties of the α and β phase (mainly strength) differ enough to make the extrusion of high α material by low β material a possibility. This has not been proved by experimental evidence. Argonne National Laboratory expressed the opinion that operation in the bi-phase may be feasible provided thermal cycling is eliminated. This would necessitate continuous processing. Warp is probably the largest problem associated with gradients. This may be reduced by putting an external load on the fuel element (Chalk River data indicates this). Plans are being made to study the effects of large gradients at high temperatures; i.e., in the proposed PuP reactor approximately 75 per cent of the temperature drop between the core center and the coolant takes place within the core, i.e., 650° C at core center to 425° C at core edge (core radius = 0.625 inch). Also see proposed theories.

Rate of Thermal Cycling

The general indication is that distortion is directly proportional to the rate of thermal cycling. Although numerous tests have been made using different heating and cooling techniques, it is difficult to interpret the results and extrapolate to pile operation conditions. ANL data show that the growth rate becomes constant as the holding time at higher temperatures is increased. See results.

Radiation

Recent work at Knolls Atomic Power Laboratory has shown that the equivalence of thermal cycling to radiation has been estimated; one thermal cycle = one MWD/T of pile exposure. For instance, in PuP this means about 600 thermal cycles = the radiation effects. ANL has recently shown in Report No. 4604 the reduction of radiation damage through uranium-zirconium alloys. New data observed on a recent trip revealed the following. Samples of U-235 ((1) α annealed, (2) β treated, (3) four at per cent Zr) were subjected to a pile exposure of approximately 0.6 per cent burn up. The specimens distorted very similarly to thermal cycling specimens having the same pretreatment without radiation. The large grain specimens showed surface distortion, the α-annealed specimen showed the greatest longitudinal growth, while the β-treated and alloyed specimens showed some distortion but were relatively stable. Future tests will be conducted on specimens varying in U-235 concentration and alloying additions including chromium.

It has been found that the grain boundary flow in pure uranium occurs at about 300° C. This, together with the fact that a large percentage of the distortion occurs during cooling, raises the question as to whether distortion due to radiation may be annealed out by operating above a minimum temperature of 300° C.

There is considerable data from irradiated fuel elements which is highly confusing; i.e., some Hanford fuel slugs show a decrease in diameter together with a decrease in length while others show an increase in diameter with a decrease in length. Rods in the NRX reactor irradiated in the outer regions of the pile show less fattening than rods receiving equivalent exposure in the higher flux area.

Assembly of Fuel Element

Recent indications at Hanford show that the methods of assembly of fuel elements can to some degree affect the distortion, with reference to both materials and techniques. It is believed that small variations in jacket thickness can cause weak spots. Also, there is at present some controversy as to when the slugs should be β treated, before or during the canning operation. Non-uniform β treatment, quenching, or mechanical working during assembly, may cause warp on the fuel element prior to pile exposure.

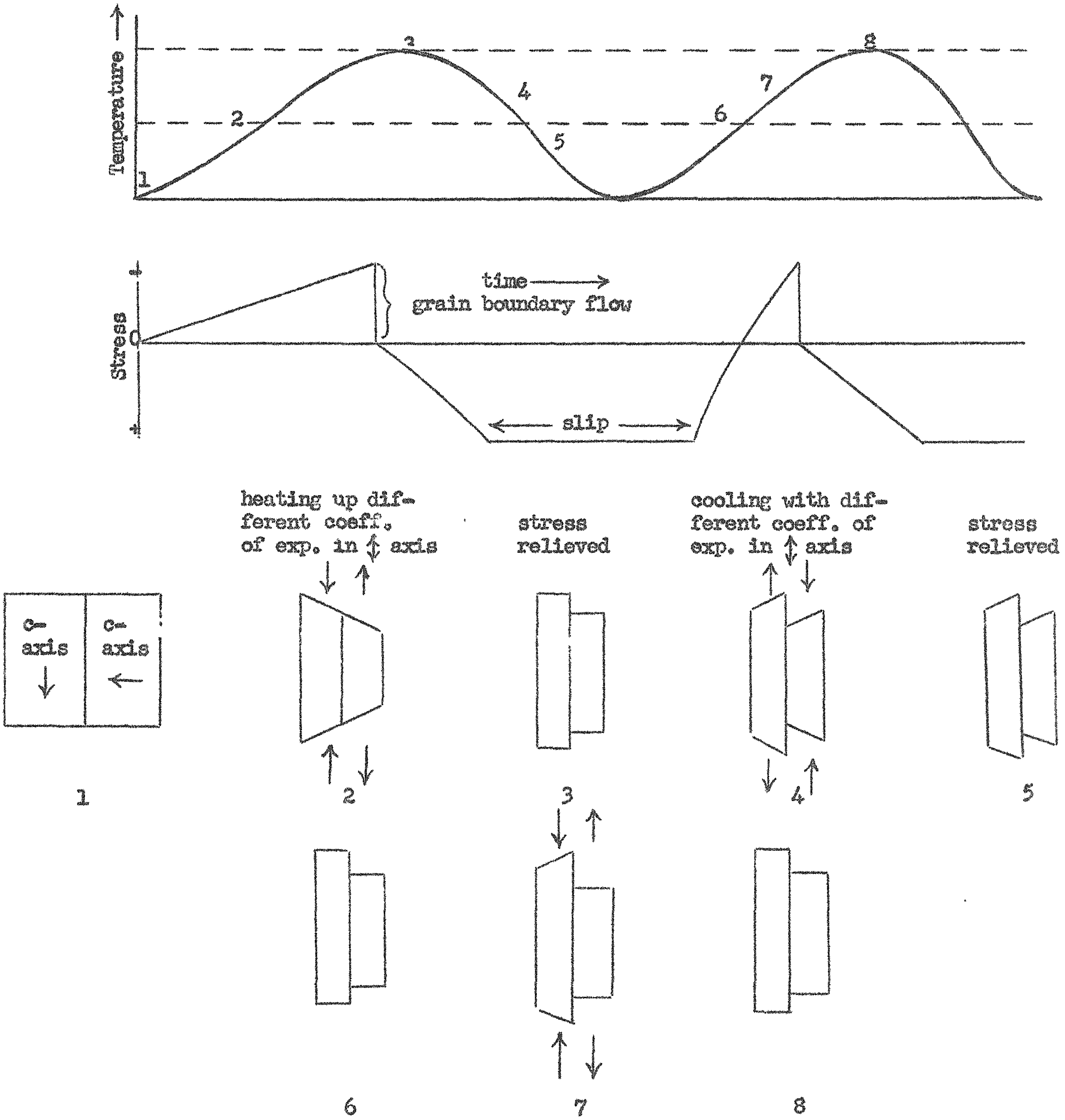
Miscellaneous Data

ANL has shown that a 30 mil stainless steel jacket has sufficient strength to restrain the growth of an α -annealed slug after 5000 thermal cycles between 100°-500° C.

Continued on next page

TWO THEORIES FOR DIMENSIONAL CHANGES

I. The following diagramatic sketch indicates a theory proposed by J. Burke of KAPL. It has a few practical drawbacks; i.e., grain boundary flow doesn't occur in as short a time as indicated.



II. This theory is proposed by J. Woodrow (at Chalk River?). The dimensional changes are explained in terms of the magnitude and variations of the yield points of uranium in tension and compression and the thermal gradients. By this theory the difference between Hanford and Chalk River distortion is related to the non-uniform stresses in the Hanford system caused by end effects.

Under irradiation the center of the rod is hotter than the ends or surface and is held in compression by these cooler areas. The hot material will yield when its yield point is less than half of the yield point in tension of the cooler areas. As the rod cools the center becomes shorter and puts the formerly cooler areas in compression. Then, provided the yield point in compression is less than the yield point in tension, the outer regions are compressed resulting in an overall shortening of the rod. Under thermal cycling different thermal gradients occur. These may result in the yield point in compression at the center never being less than half the yield point in tension at the outer areas. Elongation occurs under these conditions.

If this theory is correct, how does it explain the decrease in distortion through   treatment. Where does grain orientation fit into this theory?

TYPICAL RESULTS FROM THERMAL CYCLING EXPERIMENTS

Specimen Description	Cycling Temperatures	Per Cent Elongation + (Number of Cycles) 100	300	700	1500
300° C soak rolled α annealed	100°-550° C	3.18%	9.86%	25.20%	64.0%
300° C soak rolled, β quenched annealed	"	0.04%	0.49%	1.23%	2.58%
600° C soak rolled α annealed	"	1.77%	4.48%	9.75%	20.22%
Cast, α annealed	"	0.01%	-0.02%	too small to measure	-
Swaged, α annealed	"	1.63%	4.70%	11.80%	-
α rolled	500°-550° C	2200 cycles showed 0.69% increase in length			

Comparison of Cycling Variations

Specimen	700 cycles Induction cycle 100°-550° C	700 cycles Woosher cycle 100°-550° C 2 min. cold, 20 min. hot	700 cycles Woosher cycle 100°-550° C 2 min. cold, 2 min. hot	2000 cycles Wig-Wag 20°-300° C 5 min. cold, 5 min. hot
α annealed	25% elongation	53%	41%	0.5%

General Data

300° C rolled uranium gives dimensional instability of 4 per cent in 100 cycles
600° C rolled uranium gives dimensional instability of 2.5 per cent in 100 cycles
700° C rolled uranium gives dimensional instability of 0. (?) in 100 cycles

SUMMARY AND CONCLUSIONS

Methods of Minimizing Dimensional Changes

I. Grain refinement for reducing surface roughening can be accomplished by:

a) Water quenching from β phase
b) Alloying additions such as Cr, Zr, Mo, Cb
c) Powder compacting in the high α range from powdered uranium

II. Grain random orientation reduces gross dimensional changes (longitudinal growth) to generally less than one-half of one per cent. Random orientation is a result of:

a) β heat treatment at approximately 700°-725° C
b) Casting with no mechanical work
c) Powder compacting

IV. Continuous processing of pile fuel would reduce thermal cycling distortion and may anneal out radiation changes.

V. Dimensional changes from thermal cycling are small when operating temperatures are less than 300° C.

Factors Affecting Dimensional Changes

I. Preferred orientation: The greater the degree of preferred orientation the greater the distortion.

II. Grain size: Surface roughening decreases as the grain size decreases. With a constant high degree of preferred orientation the finer the grain size the more rapid the growth. Clusters of small grains are as detrimental for surface distortion as large grains.

III. Temperature level: Generally, as the temperature level increases, the amount of distortion increases. The role of the recrystallization temperature in thermal cycling is in doubt. However, when the minimum temperature reaches 300° C the amount of distortion may vary. The effect of cycling in and out of the β phase has not been completely determined. Data on the effects of alloys and β treatment for thermal cycling in this range are incomplete. A large number of small fluctuations at high temperature causes considerable growth in rods having preferred orientation, but small changes in randomly oriented rods.

IV. Temperature difference: Distortion is directly proportional to the temperature difference.

V. Temperature gradient: It is believed that large temperature gradients, axial and radial, may contribute to growth and affect the warp problem, provided the gradient exists entirely within the α phase. The differences in physical properties between the α and β phases may cause some dimensional changes when both phases are present within one specimen. One theory bases gross dimensional changes from irradiation as resulting from gradients.

VI. Rate of thermal cycling: The data indicates that the distortion increases as the rate of cycling increases. The growth rate becomes constant as the holding time at temperature increases.

VII. Radiation: Recent work at KAPL has resulted in an apparent relationship between radiation and thermal cycling in which one MWD/T of pile exposure is equivalent to one thermal cycle. Irradiated specimens have both elongated and shrunk without apparent correlation.

VIII. Assembly of fuel element: The technique used in fuel element assembly can contribute to its distortion.

IX. Pretreatment of fuel element: The best pretreatment for operating in the α phase for pure or low alloy uranium is as follows: a β soak at 700°-725° C for complete transformation to the β phase, followed by a water quench, and a subsequent 1-2 hour α anneal at 575° C for stress relief.

Some Additional Problems

1. What are the effects of radiation on U-Cr, U-Mo, U-Cb, U-Zr (all ~ 99 at per cent U)?

2. Why do some fuel elements shrink and some elongate?

3. How can warping be eliminated? Through stress loading?

4. What are the physical properties of the above uranium alloys?

5. What is the effect of thermal cycling through phase changes? Can two phases be present under stable conditions?

6. What effect does radiation have on single crystals? Does it make them polycrystalline?

7. If grain boundary flow occurs at 300° C and a large percentage of the distortion occurs during cooling under 300° C, would the distortion caused by radiation which appears to be very similar to thermal cycling be annealed out at operating temperatures of about 500° C?

Sites Currently Contributing to Fuel Element Distortion and Fabrication Data

Site	Contribution
Battelle Memorial Institute	U-Mo, U-Cr alloys; basic stability data
Oak Ridge National Laboratory	Mostly for MTA program
Hanford Engineering Works	Assembly problems, effects of irradiation
Brookhaven National Laboratory	BNL pile element assembly
Knolls Atomic Power Laboratory	U-Cr alloy, radiation damage, theory
Chalk River	NRX pile problems, theory, effects of irradiation
Argonne National Laboratory	Principal source of data, alloys, radiation damage
Sylvania Electric Products	Powder compacting

Some References for Thermal Cycling and Fuel Element Distortion

BNL-67	Fuel Rod Testing
BR-737	Canning of Uranium Rods
EI-90	Wrinkled Tube Problem in NRX Pile
HW-18002	Dimensional Stability of Hanford Fuel Slugs
KAPL-281	Report of Visit to Chalk River
ORNL-712	Investigation of U-Al Reactor Slugs, Preparation of
PR-P-3	Changes in the Forms of X Rods Due to Irradiation
TID-65	Fuel Element Problems
ANL-FF-54	Dimensional Changes in Uranium Under Thermal Cycling (very good)
TID-203	Data on Shrinkage of Uranium Rods in NRX Reactor
TID-225	NRX Rods
TID-264	Comparison of Reactors and Observations
TID-288	Trip Report - Some Chalk River Slug Distortion
BMI-T-40	Jacketing of Uranium for High Temperature Service

ANL-4604	Reduction of Radiation Damage Through Zr Alloys
ANL-4580, 4399	Progress Report, Thermal Cycling Data
ANL-4316	Progress Report, Thermal Cycling Data, Metallurgy Division
ANL-4243	Progress Report, Thermal Cycling Data
ANL-4188	Progress Report, Thermal Cycling Data
BMI-62	Grain Refinement of Uranium by Alloying
HW-10475	Section A, Hanford Technical Manual
ORNL-170	Slug Ruptures in the ORNL Pile
NAA-SR-88	A Review of the Dimensional Stability of Natural Uranium Fuel Elements
DR-14	Uranium Metal for NRX Reactor and Future Systems
GE-KAK-2	Fuel Slug Designs

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