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Fig. 4b

NDA Memo 2136-2

THERMAL CYCLING TESTS ON U-10 w/o Mo FOR
THE ORNL FAST BURST REACTOR

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1. SUMMARY

One of the uncertainties concerning the use of U-10 w/o Mo in the ORNL Fast Burst Reactor is the thermal cycling behavior of this alloy. Accordingly, an experimental program was undertaken with the following objectives:

1. to determine whether transformation or distortion of gamma phase U-10 w/o Mo can occur during simulated fast burst reactor thermal cycling, and
2. should transformation occur, to establish the thermal cycling behavior of partially transformed U-10 w/o Mo.

Specimens were prepared by vacuum casting pins in coated graphite molds, homogenizing the castings at 1650 °F for 24 hr and centerless grinding and cutting to 0.158 in. diameter by 1½ in. long. The pins were sealed in evacuated Vycor tubes, heated rapidly both above and below the gamma transformation temperature and then cooled slowly to simulate reactor thermal cycling. Measurements of the time required to initiate transformation by conventional isothermal methods were employed to insure that the alloy material behaved as indicated in the literature. Observation of changes in appearance, dimensions, density, resistivity, and metallographic structure were used to obtain the desired information.

It was found that thermal cycling did not cause growth or distortion of either gamma phase or partially transformed U-10 w/o Mo pins. Transformation of gamma phase to $\alpha + \delta$ was not initiated by thermal cycling below the equilibrium gamma transition temperature.

2. INTRODUCTION

The design of the ORNL Fast Burst Reactor¹ is predicated upon the use of gamma phase U-10 w/o Mo as fuel material. This material is reported to possess excellent thermal cycling behavior.² However, it is not possible to extrapolate available information to anticipated conditions nor to ascertain the possible effects of thermal cycling upon the transformation kinetics of this alloy. An experimental program was undertaken, therefore, with the following objectives:

1. to determine whether distortion or transformation of gamma phase U-10 w/o Mo can occur during thermal cycling under simulated reactor conditions, and
2. should transformation occur, to establish the thermal cycling behavior of partially transformed (to $\alpha + \delta$) U-Mo.

3. FABRICATION OF SPECIMENS*

The specimens utilized in these tests were cylindrical pins 0.158 in. in diameter by 1.50 in. long. The pins were fabricated by the following process: The required quantities of depleted uranium and high purity molybdenum powder were charged into a graphite crucible (coated with magnesium zirconate) in a vacuum induction heating furnace. The melt was prepared at 2350°F and poured into a multiple cavity, coated graphite mold at 2100°F. The mold was cooled to 1650°F in 40 min after which it was quenched in water. The pins were then removed from the mold and solution heat treated (homogenized) in a vacuum furnace at 20 microns for 24 hr at 1650°F and quenched to retain gamma phase. The as-cast pins were approximately 0.22 in. in diameter by 3 in. long. These were cut to length and centerless ground to 0.158 in. diameter.

Chemical and spectrographic analysis of the finished pins as reported by National Lead Co. are given in Table 1.

A number of the finished pins were subsequently tempered at 870°F for 64 and 72 hr and water quenched to produce specimens which contained appreciable amounts of transformed ($\alpha+\delta$) material.

* Specimens were fabricated by National Lead Co., Nuclear Metal Division, Albany, New York.

Table 1 — Chemical and Spectrographic Analysis of Uranium-Molybdenum Pins

Element	Amount Present†	Element	Amount Present†
Mo	9.97%	Cr	20
C	719	Cu	12
O	25	Fe	125
N	43	Mg	N.D.*
H	3	Mn	15
Ag	<0.2	Ni	50
B	<0.25	P	50
Cd	<0.2	Si	70
		Zr	N.D.*

* N.D. — not detected.

† Results expressed as part per million unless otherwise noted.

4. EXPERIMENTAL MEASUREMENTS

Measurements of the specimen weight, density, length, diameter and electrical resistivity were made before and after all tests. A number of specimens were examined metallographically. Because of the reactive nature of the material, the specimens were wrapped in tantalum foil and sealed in evacuated ($<10^{-5}$ mm Hg) Vycor tubes prior to all high temperature tests.

Densities were determined at room temperature by the water displacement method. The method was found to be reproducible to within ± 0.02 g/cm³. Specimen weights were required for the density determination and were also used as a check on the possibility of oxidation of the specimens during the tests. The specimen weights before and after the high temperature test were within ± 0.2 mg.

Dimensions were measured by use of micrometers calibrated against standard gage blocks. The length and diameter were measured to the nearest 0.0001 in. and three readings at three points on the specimens were averaged. In general, the three values were within ± 0.0002 in. of the average.

Electrical resistivity data were obtained on the pins at room temperature before and after the tests. The current-potential method, employing a standard 0.001 ohm series resistance adjusted to within ± 0.02 percent, was used. This method has the advantage of minimizing the effects of contact and lead resistance. Potentials were measured on a Leeds and Northrup K-3 Precision potentiometer using a Minneapolis-Honeywell Null Balance Elektronik Galvanometer. A high capacity automobile storage battery provided a stable specimen current. Precautions were taken to eliminate or correct for stray and thermal emf's. The resistivities reported are the average of at least five determinations on each specimen. The individual values for any one specimen were within ± 0.2 percent of the average. Variation between specimens which were nominally the same was somewhat greater than ± 0.2 percent.

Specimens for metallographic examination were mounted in a conducting resin and wet ground through number 600 silicon carbide paper, using an oil

5. RESULTS AND DISCUSSION

Isothermal transformation studies were undertaken to ensure that the specimens utilized in the thermal cycling tests conformed to the reported^{3,4} kinetics for this alloy. Thermal cycling tests were performed in order to determine whether distortion or transformation occurred during cycling within the temperature ranges anticipated for the reactor fuel. Tests were conducted with specimens in both retained gamma and partially transformed conditions. Included were cycles in which the specimens were cycled entirely within the metastable range ($< 1070^{\circ}\text{F}$) for gamma phase, and cycles in which the specimens were heated into the gamma stable range ($> 1070^{\circ}\text{F}$).

5.1 ISOTHERMAL TRANSFORMATION STUDIES

The times required to initiate isothermal transformation of the retained gamma structure were determined at three temperatures by the quench and temper method. All specimens had been previously solution heat treated at 1650°F for 24 hr and water quenched. They were sealed in Vycor tubes and maintained at 770, 870, and 970°F , respectively, for measured times before being water quenched.

The tempering treatments were performed by placing the encapsulated specimens in cavities in a large aluminum soaking block. Six calibrated thermocouples located at various axial and radial positions in the block indicated a maximum temperature difference of about 2°F . The temperatures were recorded on a Minneapolis-Honeywell Electronik Recorder and checked periodically by a Rubicon Semi-Precision Potentiometer. Because of drift in the controller set points, the temperature during long tempering treatments could not be held to better than $\pm 10^{\circ}\text{F}$.

The time required to initiate transformation of the gamma phase in uranium-molybdenum alloys is dependent upon the technique used to detect transformation.⁴ Thus, metallographic examination generally indicates the shortest times; resistivity measurement intermediate times; and hardness measure-

ments longest times. Because of their convenience and sensitivity, room temperature resistivity measurements were adopted for following the transformation process in these and in the thermal cycling tests.

The room temperature density of the gamma phase U-10 w/o Mo specimens is 17.19 ± 0.02 g/cm³. Density and dimensional changes were insensitive to minor amounts of transformation.

Room temperature electrical resistivity data for isothermal transformations are presented in Figs. 1, 2, and 3. Transformation of the gamma phase is accompanied by a large decrease in resistivity. The average resistivity of 60 as-received pins was 71.37 ± 0.2 microhm-cm. Specimens tempered above 770°F for short times showed an initial rapid decrease of about 1 percent in resistivity, probably due to relief of residual stresses. Thereafter, the resistivity remained constant until transformation began, whereupon it decreased as much as 16 percent. No attempt was made to follow the transformation to completion.

The Time-Temperature-Transformation (TTT) curve for the U-10 w/o Mo specimens, based upon resistivity (i.e., following the initial decrease), is presented in Fig. 4. The times required to initiate transformation at 770, 870, and 970°F are approximately 75, 35, and 40 hr, respectively. The nose of the TTT curve is apparently in the vicinity of 870°F. This curve, although indicating somewhat longer times required to initiate transformation is in essential agreement with the data of Van Thyne and McPherson⁴ and of Saller et al.³

Photomicrographs of a series of specimens tempered at 870°F are shown in Fig. 5. The retained gamma structure of the solution-annealed and quenched, as-received and 2 hr specimens is shown in Figs. 5a and 5b. Fig. 5c shows the initiation of transformation after about 24 hr at 870°F. Fig. 5d shows the result of longer tempering treatment with additional transformation products evident.

This study confirmed the expectation that the cast and homogenized U-10 w/o Mo pins conformed to the reported transformation kinetics and that resistivity measurements could provide a sensitive and convenient means for detecting transformation in the thermal cycling tests.

5.2 THERMAL CYCLING TESTS

The U-10 w/o Mo specimens were thermally cycled, within the temperature ranges anticipated for the reactor fuel, to determine whether distortion or transformation occurred during cycling. Encapsulated specimens were plunged into a 1400°F furnace and heated until a thermocouple attached

to an adjacent dummy specimen indicated one of three desired upper cycle temperatures. The specimens were then withdrawn from the furnace and allowed to cool to about 150°F in 15 to 20 min before repetition of the cycle. The heating rate was essentially constant at 420°F/min up to about 1000°F and departed from a linear rate above that temperature. The average rate for specimens heated to 1260°F was about 300°F/min.

The temperature to which the specimens were heated during thermal cycling is subject to some uncertainty because of the transient conditions and the fact that the thermocouples were not attached to the actual test specimens. However, heat transfer calculations and a number of temperature calibration tests indicated that the actual specimen temperatures were about 60°F higher than the temperatures indicated by the dummy specimens. The reported temperature values have been corrected on this basis.

One test was performed to determine whether U-10 w/o Mo will transform as a result of interrupted heating. A number of specimens were tempered for 64 hr at 870°F. Two specimens were tempered for 8 hr at 870°F and water quenched. This cycle was repeated eight times, to total 64 hr of interrupted heating. A comparison was made of the amount of transformation which occurred as a result of the two treatments, based upon resistivity decrease.

The results of all thermal cycling tests are presented in Tables 2, 3, and 4. In most cases density, length, and diameter were measured before and after cycling to determine whether there was any distortion or growth. Resistivity measurements were made before and after cycling to determine whether there was any transformation. A number of specimens were examined metallographically after the tests. (See Fig. 6.) There was essentially no change in the microstructure of cycled specimens.

In terms of reactor performance there were no significant changes in density, length or diameter of either gamma phase or partially transformed specimens as a result of the thermal cycling tests. The resistivities of gamma phase specimens remained essentially unchanged (except for the initial 0.5 to 1 percent decrease probably due to relief of internal stresses), indicating that transformation was not initiated during thermal cycling. However, the results of one experiment (Table 4) indicate that transformation will initiate and proceed during thermal cycling if sufficient time at temperature is accumulated in the transformation range. The specimens partially transformed to $\alpha + \delta$ prior to thermal cycling (Table 3) exhibited slight decreases in resistivity. These may be attributed either to the occurrence of additional transformation during cycling or again to relief of quenching stresses during the slow cooling portion of the cycle.

Although these results are very encouraging, their limitations should be apparent. The thermal cycle associated with operation of the ORNL Fast Burst Reactor involves extremely rapid (~40 microsec) heating of the fuel, followed by relatively slow cooling to room temperature. Rapid heating produces large inertial stresses. The possibility of reproducing the rapid heating and consequent inertial stresses was considered¹ but was not pursued because of the complexity and cost of the experiment. The thermal cycle tests that were performed simulated the total time at temperature for typical portions of fuel and provided information on thermal cycling distortion and transformation in the absence of inertial stresses.

Table 2 — Thermal Cycle Test Results for Gamma Phase U-10 w/o Mo

Test No.	Specimen No.	No. of Cycles	Density Change, g/cm ³	Length Change, in. × 10 ³	Diameter Change, in. × 10 ³	Resistivity, microhm-cm	
						Before	After
1*	7	41	+0.1	Nil	-0.1	71.48	70.81
	9	361	Nil§	-0.2	-0.1	71.37	70.61
	12	760	Nil	-0.1	-0.2	71.33	70.45
	2	900	Nil	-0.5	-0.1	71.41	70.57
	10	900	Nil	+0.1	-0.1	71.52	70.67
	11	900	Nil	-0.4	-0.1	71.56	70.70
2†	63	1019	N.D.¶	-0.1	Nil	71.31	71.39
	64	1019	N.D.	-0.3	Nil	71.32	71.28
	65	1019	N.D.	-0.3	Nil	71.38	71.42
	66	1019	N.D.	+0.1	Nil	71.36	71.23
3‡	19	298	Nil	-0.2	-0.1	71.44	70.95
	29	298	Nil	-0.2	Nil	71.46	70.97
	4	1000	Nil	-0.1	-0.1	71.30	70.96
	14	1000	Nil	+0.7	-0.1	71.30	70.87
	16	1000	Nil	-0.4	-0.1	71.39	70.87
	23	1000	+0.1	-0.5	-0.3	71.47	70.73

* Test No. 1 — 150°F to about 1050°F, average heating rate = ~420°F/min, cooling time = 15 to 20 min.

† Test No. 2 — 150°F to about 960°F, average heating rate = ~420°F/min, cooling time = 15 to 20 min.

‡ Test No. 3 — 150°F to about 1260°F, average heating rate = ~300°F/min, cooling time = 15 to 20 min.

§ Nil for density change = < 0.01 g/cm³

Nil for length change = < 0.1 × 10⁻³ in.

Nil for diameter change = < 0.1 × 10⁻³ in.

¶ N.D. — not detected.

Table 3 — Thermal Cycle Test* Results for Partially Transformed U-10 w/o Mo

Specimen No.	No. of Cycles	Length Change After Cycle, in. $\times 10^3$	Diameter Change After Cycle, in. $\times 10^3$	Resistivity, microhm-cm		
				Initial Not Trans.	Trans. Avg.	Cycled
†	Not cycled	—	—	—	69.17	—
†	607	-0.2	Nil	71.18	—	68.87
†	607	-0.1	-0.1	71.40	—	68.36
†	607	-0.2	-0.1	71.37	—	68.27
‡	Not cycled	—	—	—	68.36	—
‡	1019	-0.1	-0.1	71.28	—	68.57
‡	1019	Nil	-0.1	71.39	—	68.35

* 150°F to about 960°F, average heating rate = ~420°F/min, cooling time = 15 to 20 min.

† Specimens were tempered at 870°F for 64 hr and water quenched prior to thermal cycling.

‡ Specimens were tempered at 870°F for 72 hr and water quenched prior to thermal cycling.

Table 4 — Effect of Interrupted Heating on Transformation

Specimen No.	Treatment	Resistivity, microhm-cm	
		Initial	Final
13	64 hr at 870°F, WQ	71.48	69.73
59	64 hr at 870°F, WQ	71.28	68.90
42	Eight 8-hr cycles at 870°F, WQ	71.39	68.91
43	Eight 8-hr cycles at 870°F, WQ	71.43	69.12

Fig. 1 — Room temperature resistivity data for specimens isothermally transformed at 870 °F

Fig. 2 — Room temperature resistivity data for specimens isothermally transformed at 770°F

Fig. 3 — Room temperature resistivity data for specimens isothermally transformed at 970 °F

Fig. 4 – TTT diagram for U-10 w/o Mo based upon initial resistivity decrease

NEG NO. 1228

(a) As-received; Spec. No. 34

NEG NO. 1232

(b) 2 hr at 870°F, WQ;
Spec. No. 15

NEG NO. 1233

(c) 25.2 hr at 870°F, WQ;
Spec. No. 17

NEG NO. 1239

(d) 72 hr at 870°F, WQ;
Spec. No. 76

Fig. 5 — Photomicrographs illustrating progress of isothermal transformation from gamma to alpha plus epsilon at 870°F. 200×.

NEG NO. 1237

Typical gamma structure, cycled 1019 times
between 150°F and 960°F; Spec. No. 64

NEG NO. 1238

Partially transformed structure, 72 hr at 870°F,
WQ, plus 1019 cycles between 150°F and 960°F;
Spec. No. 73

Fig. 6 — Effect of thermal cycling on microstructure of gamma and partially
transformed specimen; compare with Figs. 5a and 5c. 200X.

6. CONCLUSIONS

The following conclusions are based upon the results of these tests:

1. Based upon resistivity measurements, the Time-Temperature-Transformation curve for the cast and homogenized U-10 w/o Mo pins is in essential agreement with previously published TTT data for this alloy. It is noted, therefore, that neither the rather high carbon content nor the fabrication procedures has markedly affected the kinetics of transformation.
2. Thermal cycling, for about 1000 cycles, of the U-10 w/o Mo pins, either within the gamma metastable range or into the gamma range, did not cause distortion or growth of either retained gamma or partially transformed (~20%) pins.
3. It appears that although transformation can be initiated during thermal cycling, sufficient time at temperature is not accumulated during the anticipated reactor lifetime (~1000 cycles) for transformation to occur. Furthermore, as far as distortion and growth of fuel are concerned, there are no serious consequences even if the fuel should partially transform to $\alpha + \delta$.

7. REFERENCES

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