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IRRADIATION OF U-Mo BASE ALLOYS

By

M. P. JOHNSON
W. A. HOLLAND

ATOMICS INTERNATIONAL

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PREFACE

The results reported herein represent work performed under AEC contract in fiscal years 1961 and 1962. Subsequent to the initiation and examination of these NAA-38 irradiation tests, an extensive evaluation of U - 10 Mo alloys, by both AI and APDA, has shown this alloy to be highly sensitive to radiation and thermal conditions. Under certain conditions of fission rate and temperature, this alloy distorts severely. These conditions are only sometimes achieved and recorded during the irradiation in test reactors.

A broader analysis of this problem, based on more recent data, has been performed and will be issued in a later topical report.

CONTENTS

TABLES

FIGURES

ABSTRACT

A series of experiments was designed to assess the suitability of uranium-molybdenum alloys as high-temperature, high-burnup fuels for advanced sodium cooled reactors. Specimens with molybdenum contents between 3 and 10%* were subjected to capsule irradiation tests in the Materials Testing Reactor, to burnups up to 10,000 Mwd/MTU at temperatures between 800 and 1500°F.

The results indicated that molybdenum has a considerable effect in reducing the swelling due to irradiation. For example, 3% molybdenum reduces the swelling from 25%, for pure uranium, to 7% at ~3,000 Mwd/MTU at 1270°F. Further swelling resistance can be gained by increasing the molybdenum content, but the amount gained becomes successively smaller. At higher irradiation levels, the amount of swelling rapidly becomes greater, and larger amounts of molybdenum are required to provide similar resistance. A limit of 7% swelling, at 900°F and an irradiation of 7,230 Mwd/MTU, requires the use of 10% molybdenum in the alloy. The burnup rates were in the range of 2.0 to 4.0 x 10^{13} fission/cc-sec.

Small ternary additions of silicon and aluminum were shown to have a noticeable effect in reducing swelling when added to a U - 3% Mo alloy base. Under the conditions of the present experiment, 0.26% silicon or 0.38% aluminum were equivalent to 1 to 1-1/2% molybdenum.

The Advanced Sodium Cooled Reactor requires a fuel capable of being irradiated to 20,000 Mwd/MTU at temperatures up to 1500°F in metal fuel, or equivalent in ceramic fuel. It is concluded that even the highest molybdenum contents considered did not produce a fuel capable of operating satisfactorily under these conditions. The alloys would be useful, however, for less exacting conditions. The U - 3% Mo alloy is capable of use up to 3,000 Mwd/MTU at temperatures of 1300°F before swelling becomes excessive. The addition of silicon and aluminum would increase this limit to at least 3,500 Mwd/MTU, and possibly more if the alloy were heat treated to provide a fine dispersion of second phase.

*All compositional percentages are weight percent, unless otherwise stated.

I. INTRODUCTION

Uranium-molybdenum binary alloys, and uranium-molybdenum base ternary alloys containing aluminum or silicon, were studied as prospective high-temperature, high-burnup fuels for Advanced Sodium Cooled Reactors (ASCR). Fuels for ASCR require high thermal conductivity, high uranium density, high elevated-temperature strength, relatively high resistance to irradiation damage, and resistance to corrodents that may be encountered during fabrication, storage, and reactor service. A large amount of prior research and development on metallic fuels had dealt with unalloyed uranium[1] and binary alloys containing small amounts of molybdenum.[2] Such fuels were known to fall short of ASCR requirements.

This program was initiated to evaluate binary U-Mo alloys having up to 10 wt% Mo, and ternary U - 3 wt% Mo base alloys with small additions of either silicon or aluminum, as ASCR fuels. It is known that molybdenum tends to stabilize the high-temperature gamma phase, and that this phase has a higher resistance to irradiation damage than alpha uranium.[3,4] In equilibrium at low temperatures, the alloys being considered would have a structure consisting of $\alpha + \gamma'$, the γ' being an ordered form of the gamma phase. The approach to equilibrium, $\gamma \rightarrow \alpha + \gamma'$, is retarded by the addition of molybdenum, and takes some hundreds of hours at 900°F for complete transformation, in the case of the 10 wt% alloy. Opposing this tendency toward equilibrium is the effect of neutron irradiation, which causes a reversal of the transformation reaction by localized thermal agitation of the alloy and reforming the high-temperature gamma phase.[5,6] This effect of reversion to the metastable γ phase during irradiation could have a large effect in the promotion of swelling resistance. To favor this reaction, high fission rates and high molybdenum contents in the region of 10 wt% are required. A series of experiments was designed to show the effect of molybdenum. The fission rate varied over only a relatively small range of 2.0 to 4.0 x 10^{13} fissions/cc-sec.

As a further method of reducing swelling, it has been suggested that, if the gas bubbles were nucleated on a fine enough scale, the surface tension forces would restrict the swelling.[7] Silicon and aluminum additions tend to form intermetallics as a fine dispersion with suitable treatments.[8] Their effect in providing nuclei in the binary U - 3 wt% Mo was observed in this series of experiments.

ASCR operating criteria, at the time these experiments were planned, required fuel capable of operating at center temperatures of 1200 to 1300°F, and a minimum burnup of 20,000 Mwd/MTU at a fuel surface heat flux of 1×10^6 BTU/hr-ft^2. The experiments were designed to determine whether the materials irradiated would show promise as ASCR fuels.

II. EXPERIMENTAL METHODS

A. MATERIAL

All fuel materials for these irradiations were produced by vacuum induction melting and casting into 3/8-in. diameter by 6-in. long fuel pins. These pins were subsequently cut into specimens 1-1/2 in. in length. The chemical analyses of the individual irradiation specimens are shown in Tables I and II. The specimens were irradiated either in the "as-cast" or "gamma heat treated" condition. Material was not removed from the cast surface of any of the slugs. The gamma heat treatment consisted of heating to 1560°F in evacuated Vycor capsules, holding for 20 hr, and water quenching.

B. DESCRIPTION OR IRRADIATION ASSEMBLIES

Figure 1 is an illustration of the irradiation capsule assembly. Each experiment contained a column of twelve 3/8-in. diameter by 1-1/2-in. long specimens. The column of fuel slugs was supported by a split, perforated metal basket with an ID 0.144 in. greater than the diameter of the slugs. Four 0.072-in. diameter stainless steel tubes, oriented longitudinally and arranged symmetrically in the basket, centered the column of slugs and provided an annular space near the fuel for fuel-surface thermocouples.

The specimen assembly was centered in an 0.900-in. OD stainless steel capsule and NaK bonded. Four longitudinal, 0.095-in. diameter tubes containing cobalt wire flux monitors were attached to the outside of each assembly. Each assembly contained eight 1/16-in. OD, MgO insulated, stainless steel sheathed, Chromel-Alumel thermocouples. Seven thermocouples were located near the fuel surface, and one was located near the center of the fuel. The central thermocouple was protected by a tantalum foil sheath. The location of each of the thermocouples, in relation to the fuel slugs, is shown in Figure 2.

The details of the irradiations assemblies are presented in NAA-SR-Memo 3215.[9]

C. IRRADIATION HISTORY

The NAA-38-2 and -4 assemblies were irradiated in the A-29-SE position, and NAA-38-3 and -5 in the A-28-SE position, of the MTR. Nominal unperturbed neutron flux was 0.75×10^{14} nv, and irradiation time was three MTR cycles for

TABLE I

CHEMICAL ANALYSIS OF NAA-38-2, -3, -4, AND -5 FUEL ALLOYS

Irradiation	Slug No.	Nominal Mo (wt %)	Mo (wt %)	Si (wt %)	Al (wt %)	C (wt %)
NAA-38-2	1	10.0	10.65			0.08
	2	7.5	8.50			
	3	5.0	5.14			0.06
	4	3.0	2.91			
	5	10.0	10.65			0.08
	6	10.0	10.36			0.08
	7	7.5	8.53			
	8	7.5	8.53			
	9	5.0	5.14			0.06
	10	3.0	2.91			
	11	5.0	5.56			0.06
	12	3.0	2.86			0.06
NAA-38-3	1	10.0	10.65			0.01
	2	7.5	8.50			
	3	5.0	5.14			0.06
	4	3.0	2.91			
	5	10.0	10.65			0.08
	6	10.0	10.36			0.08
	7	7.5	8.53			0.10
	8	7.5	8.53			
	9	5.0	5.14			0.11
	10	3.0	2.91			
	11	5.0	5.56			0.06
	12	3.0	2.86			0.06
NAA-38-4	1	3.0	2.90			
	2	10.0	9.75			0.07
	3	10.0	9.75			0.07
	4	3.0	2.89			0.05
	5	3.0	4.13	0.26		
	6	3.0	2.80		0.38	
	7	10.0	9.75			0.07
	8	3.0	2.90			
	9	3.0	4.07	0.26		0.11
	10	3.0	2.80		0.38	0.09
	11	3.0	4.16	0.26		
	12	3.0	2.80		0.38	
NAA-38-5	1	3.0	2.90			
	2	10.0	10.62			0.05
	3	10.0	10.62			0.05
	4	3.0	2.89			0.05
	5	3.0	4.00	0.26		
	6	3.0	2.80		0.38	
	7	10.0	10.62			0.05
	8	3.0	2.89			0.05
	9	3.0	4.07	0.26		0.11
	10	3.0	2.90			0.05
	11	3.0	4.16	0.26		
	12	3.0	2.80		0.38	

TABLE II

TYPICAL IMPURITY CONTENT OF NAA-38-2, -3, -4, AND -5 FUEL ALLOYS
(Impurities in ppm)

Al	Be	Bi	B	Cd	Ca	Co	Cr	Cu	Fe	Hg	Li	Mg	Mn	Ni	Nb	Pb	Si	Sn	Ti	V	Zn	Nominal Composition (wt %)
<25	<0.5	<20	2	<1	<5	<5	5	<5	35	<10	<50	4	20	40	<40	1	200	<1	5	<1	<50	U - 3 Mo
<25	<0.5	<20	3	<1	<5	<5	5	<5	30	<10	<50	4	10	40	<40	3	175	4	<5	<1	<50	U - 5 Mo
<25	<0.5	<20	4	<1	<5	<5	5	10	30	<10	<50	2	10	10	<40	2	300	4	<5	<1	<50	U - 7.5 Mo
<25	<0.5	<20	4	<1	<5	<5	5	5	50	<10	<50	2	20	10	<40	4	200	4	<5	<1	<50	U - 10 Mo
*	<0.5	<20	4	<1	5	200	5	5	100	<10	<50	8	10	20	<40	20	-	1	10	<1	<50	U - 3 Mo - 0.1 Al
50	<0.5	<20	10	<1	5	400	5	100	800	<10	<50	15	10	40	<40	15	*	2	20	1	<50	U - 3 Mo - 0.5 Si

*Beyond range of spectrographic standard (> 1000 ppm)

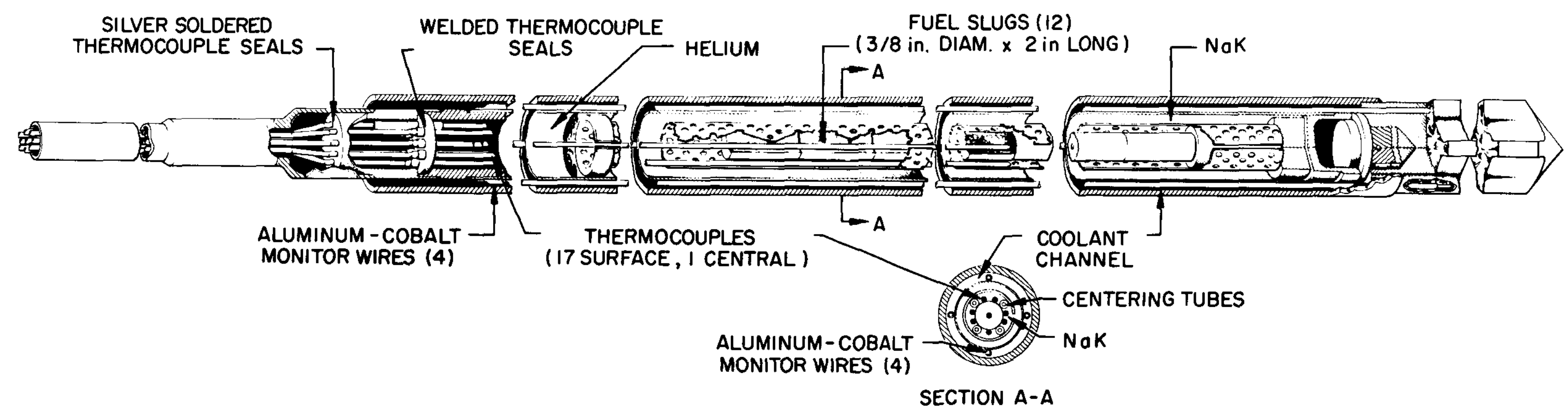

Figure 1. Diagram of Irradiation Capsule Assembly

Slug Composition NAA-38-2 and 3	Slug Number	Thermocouple Location	Slug Composition NAA-38-4 and 5
U – 10 Mo (gamma heat treated)	1		U – 3 Mo
		Tc 5	
U – 7.5 Mo (gamma heat treated)	2		U – 10 Mo (gamma heat treated)
U – 5 Mo	3		U – 10 Mo
U – 3 Mo	4	Tc 1, 4, 6 (Surface) Tc 8 (Central)	U – 3 Mo
U – 10 Mo (gamma heat treated)	5		U – 3 Mo – 0.5 Si
U – 10 Mo	6		U – 3 Mo – 0.1 Al
		Tc 2	
U – 7.5 Mo	7		U – 10 Mo
U – 7.5 Mo (gamma heat treated)	8		U – 3 Mo
U – 5 Mo (gamma heat treated)	9	Tc 7	U – 3 Mo – 0.5 Si
U – 3 Mo	10		U – 3 Mo – 0.1 Al (–4) U – 3 Mo (–5)
U – 5 Mo	11		U – 3 Mo – 0.5 Si
		Tc 3	
U – 3 Mo	12		U – 3 Mo – 0.1 Al

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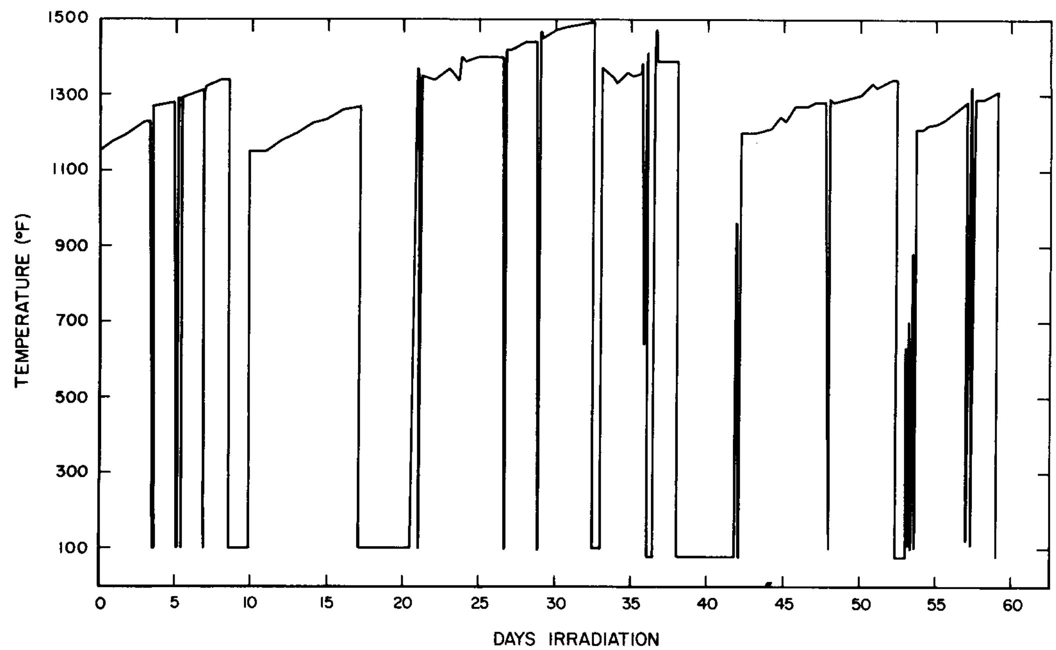
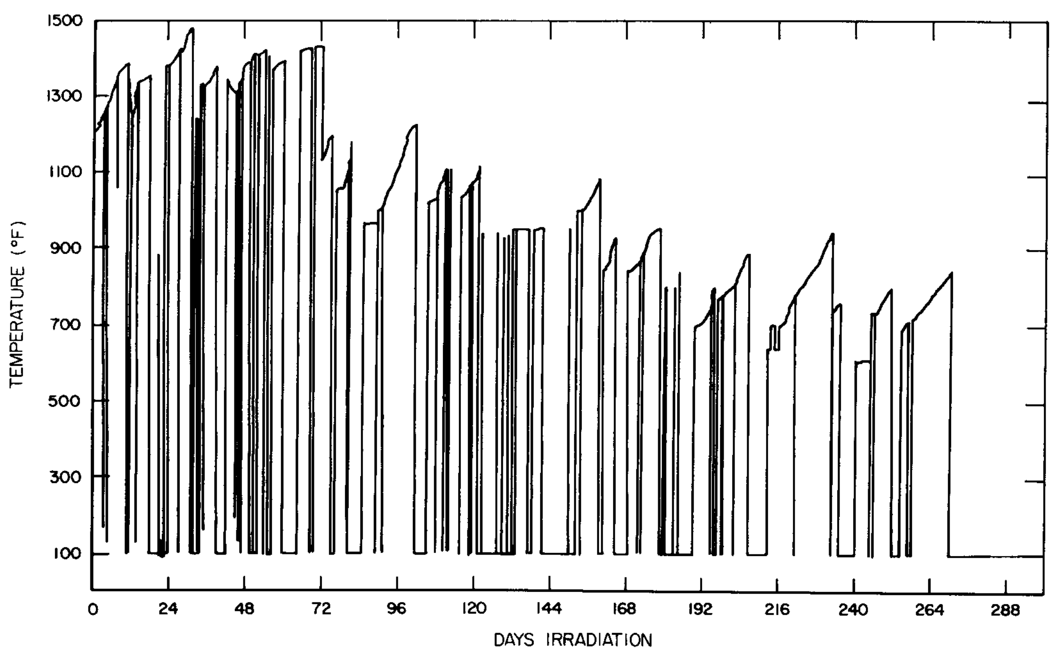
Figure 2. Slug Identity and Thermocouple Placement



a. NAA-38-2



b. NAA-38-3

Figure 3. Temperature Data on Point No. 8
(Fuel Center Thermocouple)

NAA-38-2 and -5, and ten cycles for NAA-38-3 and -4. The target maximum central fuel temperature was 1300°F for all assemblies.

The actual temperature data, taken by continuously recording the outputs from eight thermocouples, are presented, in part, in Figures 3 and 4 and in Table III, in three forms:

1) Central fuel temperature, as measured by Thermocouple No. 8.

2) Typical surface temperature profiles along the column of fuel.

3) The average surface temperature of selected individual fuel slugs.

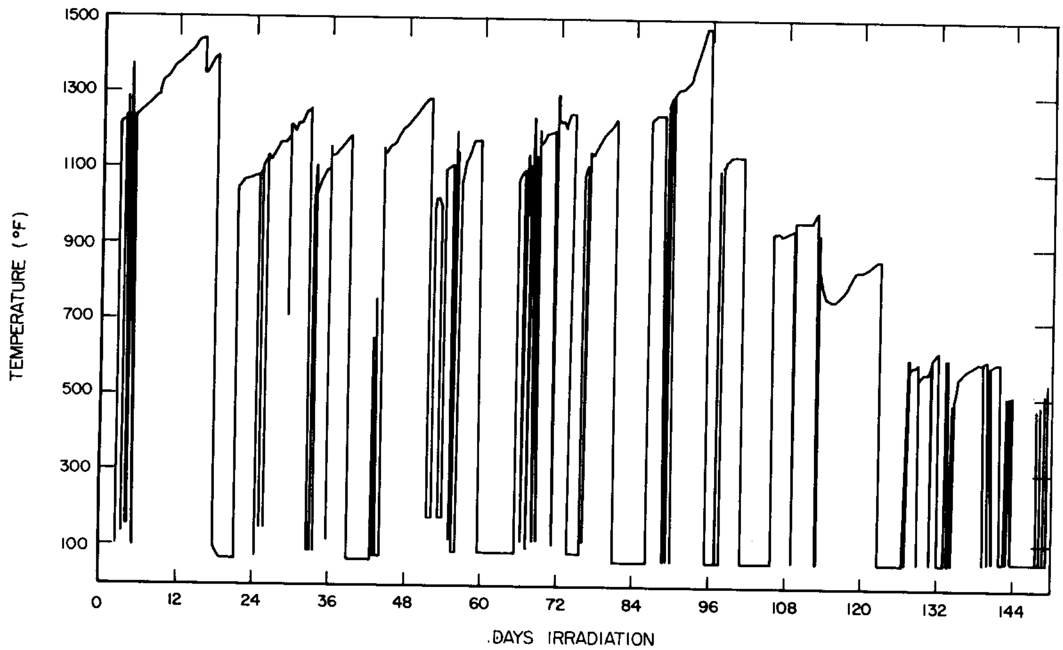
The temperature readings of Thermocouple No. 8 (central fuel) are given, in Figures 3a, b, c, and d, for the NAA-38-2, -3, -4, and -5 assemblies, respectively. Figure 3c does not show the last 40 days of irradiation, during which time the temperatures were about the same as for the last 25-day period shown. It should be noted that the temperatures in these figures are not maximum central fuel temperatures. Due to continual repositioning of control rods during reactor operation, there is a continuous upward shift of the position of maximum neutron flux, and hence temperature. This effect can be observed in Figures 4a and 4b.

Several comments will aid in the interpretation of Figures 3a, b, c, and d.

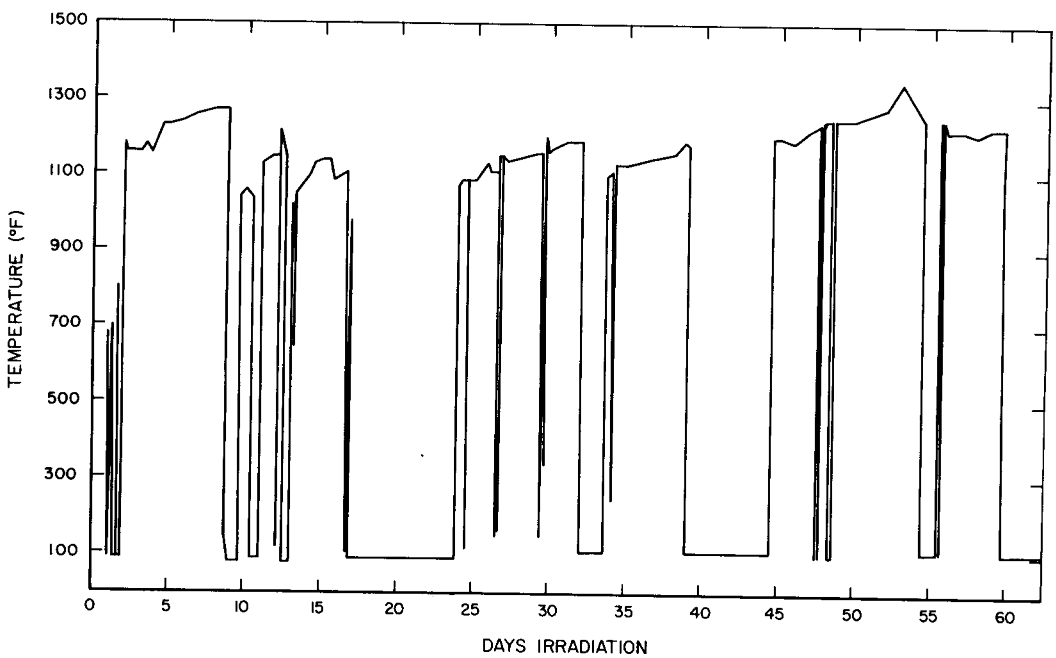
1) The short periods of abrupt temperature decrease, to approximately 100°F, are due to unscheduled scrams. The low temperature periods of about 1 to 1-1/2 days duration are midcycle refuelings, and the low temperature periods of 3 to 4 days duration are end-of-cycle shutdowns.

2) The gradual increase in temperature (saw-tooth effect), beginning with each new cycle and midcycle refueling, is due to shift in control-rod position.

3) The range of central fuel temperatures at the position of Thermocouple No. 8 for NAA-38-2 and -5, and in the first 100 irradiation days for NAA-38-3 and -4, is typical for capsules without temperature control. The lower temperatures for NAA-38-4, after 100 days irradiation, are the effect of a change in core loading pattern at MTR during this period.

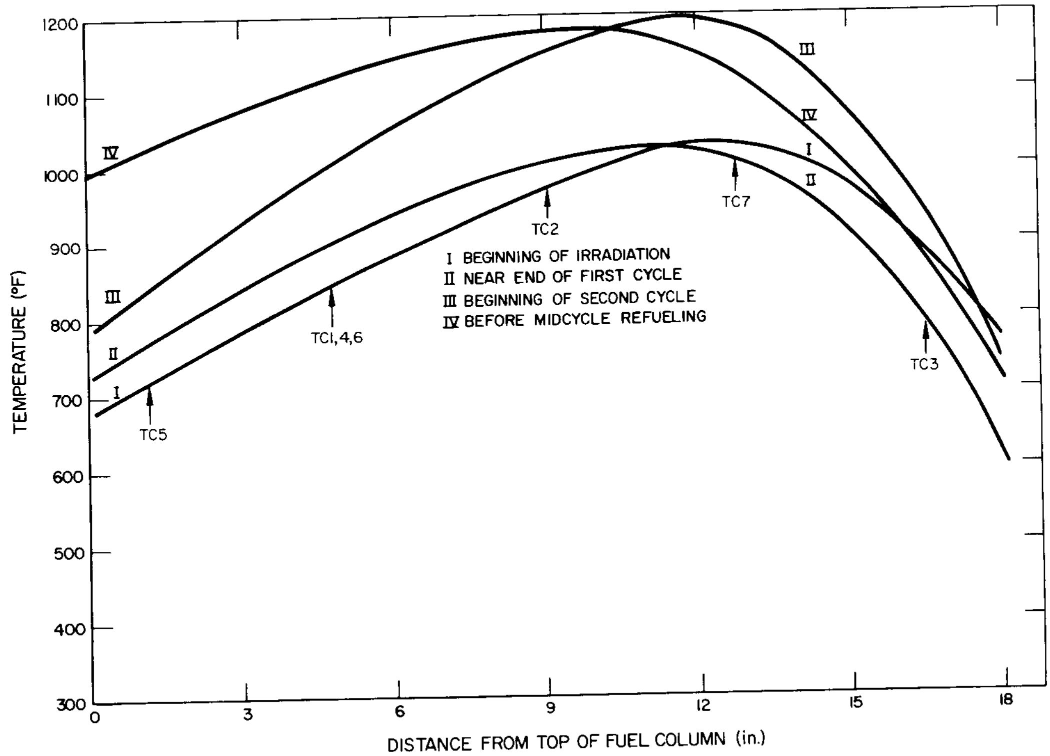


c. NAA-38-4

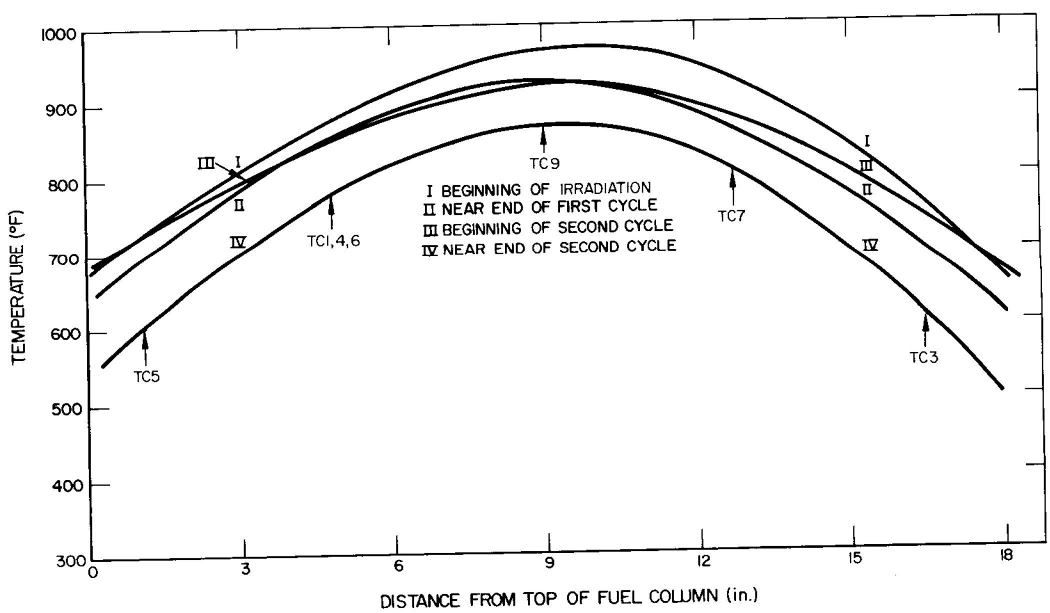


d. NAA-38-5

Figure 3 (Continued). Temperature Data on Point No. 8 (Fuel Center Thermocouple)



a. NAA-38-2



b. NAA-38-5

Figure 4. Surface Temperature Profile

TABLE III

SURFACE TEMPERATURE VARIATIONS DURING IRRADIATION

Irradiation	Slug No.	Nominal Composition (wt %)	Surface Temperature (°F)		
			Average	Low	High
NAA-38-2	1	U – 10 Mo	805	640	1010
	9	U – 5 Mo	1070	1000	1265
	12	U – 3 Mo	750	585	940
NAA-38-3	4	U – 3 Mo	825	515	1240
	6	U – 10 Mo	930	600	1215
	9	U – 5 Mo	930	610	1275
NAA-38-5	1	U – 3 Mo	735	585	885
	6	U – 3 Mo – 0.1 Al	965	860	1045
	12	U – 3 Mo – 0.1 Al	690	490	770
NAA-38-4	1	U – 3 Mo	670	495	880
	6	U – 3 Mo – 0.1 Al	1015	820	1160
	7	U – 10 Mo	1010	830	1150
	12	U – 3 Mo – 0.1 Al	450	225	585

Figure 4a shows surface temperature profiles along the fuel column for different stages of irradiations for NAA-38-2. Comparison of Curves I, II, III, and IV in Figure 4a clearly shows the effect of control rod withdrawal on the position of the peak temperature. Also, note the difference in temperature levels between the first cycle (Curves I and II) and the second cycle (Curves III and IV). The performance of the thermocouples in this experiment was excellent. All couples operated well throughout the irradiation, except for No. 7, which failed a few days before the end of the irradiation.

Similar temperature profile data are presented, in Figure 4b, for NAA-38-5. In this experiment, thermocouple performance was poor. No reliable readings were obtained from No. 7, and readings from No. 5 were questionable. A considerable temperature spread was indicated by No. 1, 4, and 6, which were at the same elevation. Since the readings of No. 4 agreed well with the surface temperatures determined from the central thermocouple at the same elevation (No. 8), and from ΔT considerations, its readings were favored in drawing the curves. In general, the temperature profiles for this experiment are considered less reliable than those for NAA-38-2. The profiles were established from the readings of the operable thermocouples, and interpolations through regions where thermocouples had failed were made on the basis of the best smooth curve consistent with the available knowledge of flux profile in this position.

With the above reservations as to the validity of the temperature profiles for NAA-38-5, it appears, from Figure 4b, that less shifting of the flux peak and less axial tilting of the flux was experienced than in the NAA-38-2 experiment.

Similar profile curves are not presented here for NAA-38-3 and -4 assemblies, mainly because the longer irradiation time for these experiments requires the examination of a very large number of curves for an accurate picture of temperature history.

The operation of thermocouples in Capsule 3 was generally good, and all couples were working throughout the irradiation. However, not all the readings were regarded as reliable. This can probably be attributed to displacement of thermocouples due to fuel swelling or asymetrical flux distribution.

With Capsule 4, considerable uncertainty exists in the actual temperature profile in the lower portion of the capsule. Only Thermocouple No. 3 was operable in this region, and this is strongly suspected of giving low readings.

TABLE IV

NAA-38-2 REDUCED BURNUP DATA

Pin No.	Nominal Composition (wt % Mo)	Burnup[a]			Average Central Temp. (°F)	Burnup Rate[b] Fissions/cc-sec ($\times 10^{-13}$)	Increase in Diameter[c] (%)	Increase in Length[d] (ΔL)(in.)	Decrease in Density[e] (%)	Increase in Volume[f] (%)	Material Condition
		Mwd/MTU	at. %	Fissions/cc ($\times 10^{-19}$)							
1	10.0	2550	0.27	10.50	1070	2.80	0.2	0.13	0.1	0.5	γ Heat Treated[g]
2	7.5	2750	0.29	11.90	1190	3.17	0.0	0.23	0.8	0.2	γ Heat Treated[g]
3	5.0	2950	0.31	13.40	1290	3.57	1.1	0.70	2.5	2.8	As-cast
4	3.0	3030	0.32	14.85	1370	3.96	5.6	3.17	14.5	14.3	As-cast
5	10.0	3470	0.36	14.30	1500	3.81	0.6	0.48	1.3	1.8	γ Heat Treated[g]
6	10.0	3590	0.38	14.80	1540	3.94	0.4	0.67	1.6	1.5	As-cast
7	7.5	3720	0.39	16.10	1530	4.28	0.8	0.73	2.5	2.4	As-cast
8	7.5	3640	0.38	15.75	1470	4.20	0.9	0.56	2.0	2.3	γ Heat Treated[g]
9	5.0	3440	0.36	15.60	1400	4.16	1.1	1.71	6.4	3.8	γ Heat Treated[g]
10	3.0	2960	0.31	14.50	1270	3.86	1.4	2.11	7.2	4.8	As-cast
11	5.0	2610	0.27	11.85	1130	2.91	2.9	0.71	2.8	6.5	As-cast
12	3.0	2490	0.26	12.20	1070	3.25	3.1	1.11	3.0	7.3	As-cast

(a) Calculated data based on the radiochemical analyses of Pins 2, 6, and 7 and relative gamma scans of the individual slugs.

(b) Calculated data based on the assumption that the accumulated burnup occurred in 0.7 of total reactor residence time.

(c) Average of 10 diameter measurements at 0 and 40° intervals.

(d) Measured.

(e) Measured.

(f) Calculated from measured dimensional changes.

(g) Held at 850°C for 20 hours and water quenched.

D. EXAMINATION PROCEDURE

All postirradiation examinations were conducted at the Components Development Hot Cell (CDHC) at Atomics International. At CDHC, the capsules were opened according to standard practices checked out in a dummy cell with a dummy capsule. In general, the following procedure was utilized:

1) Outer tube was cut and removed, to allow removal of dosimeter wires.

2) Inner tube was cut, to allow removal of NaK and fuel slugs.

3) NaK was digested in butyl alcohol.

4) Perforated basket was forced apart, to permit visual examination of slugs.

Slug length and diameter measurements were made by dial gage micrometers after slugs were thoroughly cleaned in an ultrasonically-agitated methanol solution. Wet and dry weights were taken, in order that slug densities could be determined. After all measurements were made, the slugs were scanned by a gamma ray spectrometer, to determine the relative intensity of gamma radiation on the slug surfaces. Samples for metallographic study and radiochemical determination were taken before slugs were put in storage casks for future reference and eventual reprocessing.

E. BURNUP MEASUREMENTS

The burnup and temperature data in Tables IV, V, VI, and VII are not absolute, but were calculated on the basis of the gamma activity of individual slugs in the capsule and the measured temperature and radiochemical analysis of selected slugs, according to the following procedure:

1) The surface activity of each slug was determined with a gamma spectrometer at four points spaced 90° apart around a circumference. When the side having maximum activity had been found, the slug was scanned along its length and peak R.M.S. values were recorded.

2) Samples from both ends and the middle of the assembly were selected for radiochemical analysis.

3) Assuming that the relative gamma scan intensity of the slugs is a measure of burnup, burnup values were determined for all the slugs from the radiochemical analysis and the gamma scan data by direct proportion.

4) Average slug temperatures were determined by proportion from calculated burnup data and the central temperature at Slug 4.

TABLE V

NAA-38-3 REDUCED BURNUP DATA

Pin No.	Nominal Composition (wt % Mo)	Burnup[a]			Average Central Temp. (°F)	Burnup Rate[b] Fissions/cc-sec ($x\ 10^{-13}$)	Increase in Diameter[c] (%)	Increase in Length[d] (%)	Decrease in Density[e] (%)	Material Condition
		Mwd/MTU	at. %	Fissions/cc ($x\ 10^{-19}$)						
1	10.0	7230	0.76	29.80	900	2.65	0.27	0.43	6.20	γ Heat treated[f]
2	7.5	7650	0.80	33.10	955	2.94	0.13	0.60	9.85	γ Heat treated[f]
3	5.0	7230	0.76	32.80	1010	2.92	6.24	10.74	19.80	As-cast
4	3.0	8320	0.88	40.75	1060	3.62			**	As-cast
5	10.0	9500	1.00	39.20	1180	3.49	12.70	*	28.30	γ Heat treated[f]
6	10.0	9880	1.04	40.70	1230	3.62	18.20	10.35	34.00	As-cast
7	7.5	10080	1.06	43.60	1260	3.88	31.00	*	45.50	As-cast
8	7.5	10190	1.07	44.10	1270	3.92	24.90	*	47.00	γ Heat treated[f]
9	5.0	9800	1.03	44.50	1210	3.96			**	γ Heat treated[f]
10	3.0	8730	0.92	42.70	1110	3.80			**	As-cast
11	5.0								**	As-cast
12	3.0								**	As-cast

(a) Calculated data based on the radiochemical analyses of Pins 1, 2, and 5.

(b) Calculated data based on the assumption that the accumulated burnup occurred in 0.7 of total reactor residence time.

(c) Average of 10 diameter measurements at 0 and 40° intervals

(d) Measured.

(e) Measured.

(f) Held at 850°C for 20 hours and water quenched.

* Ragged edges, no measurement.

** Sample disintegrated.

TABLE VI

NAA-38-4 REDUCED BURNUP DATA

Slug No.	Nominal Composition (wt %)			Burnup[a]			Average Central Temp. (°F)	Burnup Rate[b] Fissions/cc-sec ($\times 10^{-13}$)	Increase in Diameter[c]		Increase in Length[d] (%)	Decrease in Density[e] (%)	Material Condition
	Mo	Si	Al	Mwd/MTU	at. %	Fissions/cc ($\times 10^{-19}$)			Min %	Max %			
1	3			5370	0.57	26.3[g]	790	2.07	8.5	23.4	10.0		As-cast
2	10			7480	0.79	30.85	930	2.425	2.1	2.7	1.3	6.0	γ Heat Treated[f]
3	10			7950	0.84	32.82	990	2.583	2.7	3.7	−1.9	7.2	As-cast
4	3			4690	0.49	22.98[g]	1080	1.808	6.1	22.6	7.9		As-cast
5	3	0.5		8050	0.85	39.40	1180	3.142	12.5	14.1	7.4		As-cast
6	3		0.1	9130	0.96	44.70	1340	3.518	25.9	34.2	6.7		As-cast
7	10			10350	1.09	42.7	1280	3.36	9.1	28.8	5.3		As-cast
8	3			7510	0.79	36.8[g]	1100	2.898	21.9	27.2	2.7		As-cast
9	3	0.5		9150	0.96	44.76	1340	3.518	27.5	34.2	7.3		As-cast
10	3		0.1	9380	0.99	45.9	1380	3.617	42.1	48.0	8.4		As-cast
11	3	0.5		8450	0.89	41.4	1220	3.26	2.9	31.8	2.0	13.6	As-cast
12	3		0.1	7640	0.80	37.41	1120	2.945	2.4	17.6	0.6		As-cast

(a) Calculated data based on the radiochemical analyses of Slugs 2, 6, and 7 and relative gamma scans of the individual slugs.

(b) Calculated data based on the assumption that the accumulated burnup occurred in 0.7 of total reactor residence time.

(c) Average of 5 measurements at maximum and minimum diameters.

(d) Measured

(e) Measured

(f) Held at 850°C for 20 hours and water quenched.

(g) Inconsistent and unreliable gamma scan data.

TABLE VII

NAA-38-5 REDUCED BURNUP DATA

Pin No.	Nominal Composition (wt %)			Burnup[a]			Average Central Temp. (°F)	Burnup Rate[b] Fissions/cc-sec (x 10-13)	Increase in Diameter[c] (%)	Increase in Length[d] (%)	Decrease in Density[e] (%)	Increase in Volume (f) (%)	Material Condition
	Mo	Si	Al	Mwd/MTU	at. %	Fissions/cc (x 10-19)							
1	3			1990	0.21	9.76	850	2.56	0.2	0.18	0.6	+0.7	As-cast
2	10			2280	0.24	9.40	940	2.46	0.1	−0.39	0.0	−0.3	γ Heat Treated[g]
3	10			2470	0.26	10.20	1020	2.67	0.2	−0.013	0.2	+0.4	As-cast
4	3			2740	0.29	13.42	1160	3.52	0.8	0.66	3.8	+2.0	As-cast
5	3	0.5		2790	0.29	13.67	1180	3.58	0.7	0.47	2.2	+1.9	As-cast
6	3		0.1	2830	0.30	13.85	1190	3.63	2.4	0.21	3.6	+4.7	As-cast
7	10			2950	0.31	12.15	1215	3.18	0.3	0.33	0.8	+0.7	As-cast
8	3			2920	0.31	14.32	1230	3.74	2.6	1.07	5.9	+6.9	As-cast
9	3	0.5		2780	0.29	13.63	1175	3.57	1.1	0.53	2.2	+2.9	As-cast
10	3			2560	0.27	12.53	1065	3.28	1.2	0.33	2.5	+2.8	As-cast
11	3	0.5		2370	0.25	11.61	1000	3.02	0.6	0.335	1.3	+1.5	As-cast
12	3		0.1	2190	0.23	10.72	930	2.81	0.6	−0.366	0.4	+1.0	As-cast

(a) Calculated data based on the radiochemical analyses of Pins 2, 6, and 7 and relative gamma scans of the individual slugs.

(b) Calculated data based on the assumption that the accumulated burnup occurred in 0.7 of total reactor residence time.

(c) Average of five measurements.

(d) Measured.

(e) Measured.

(f) Calculated from measured dimensional changes.

(g) Held at 850°C for 20 hours and water quenched.

III. RESULTS

A. NAA-38-2

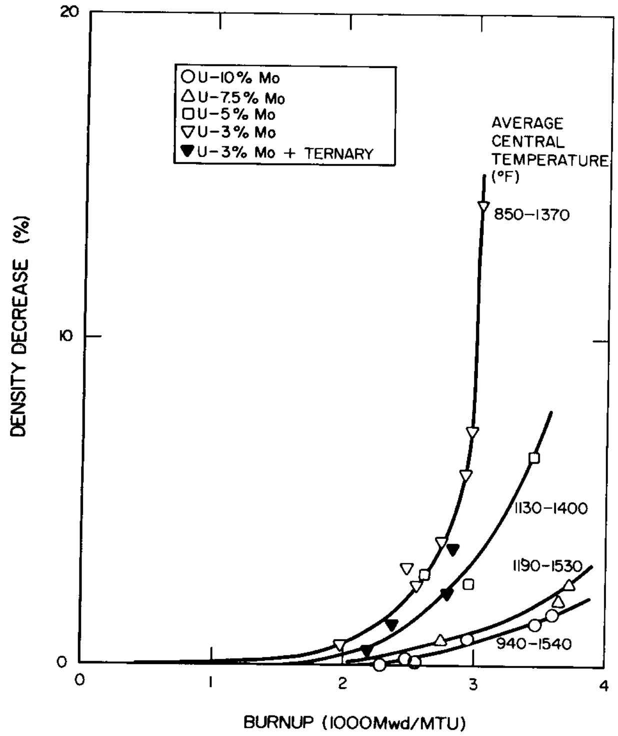
This capsule was irradiated to fuel burnups of 2500 to 3700 Mwd/MTU and center temperatures of 1070 to 1540°F. None of the slugs suffered extensive damage except that No. 4, a U - 3 wt% Mo specimen irradiated to 3030 Mwd/MTU, showed a density decrease of 14.5%. This is considerably more than the other two 3% Mo slugs in the capsule, although it had only a slightly increased burnup. This suggests that the slug was on the point of catastrophic swelling. A similar situation occurred with the three 5% Mo slugs, although at a lower level of damage than the 3% Mo specimens. Thus, there is a fairly sudden increase in damage between Specimens 9 and 3 for a small increase in burnup.

The 7.5% Mo and 10% Mo specimens all show much less swelling than do the lower percentage molybdenum specimens. Although the density change increases with increasing burnup, there is no sudden change apparent in this capsule. The heat treatment given to the U - 10% Mo specimens appears to have had little effect on the damage resistance of the alloy. This can be seen from a comparison of Slugs 5 and 6. Both of these specimens, however, were running well above the $\alpha + \gamma' \rightarrow \gamma$ phase-change temperature, and so the initial heat treatment would be expected only to affect the initial period of the irradiation.

B. NAA-38-3

This capsule was a high burnup version of Capsule 2,. and again the effect of composition and burnup is evident.

The burnup of more than 8000 Mwd/MTU proved to be beyond the capability of the lower molybdenum compositions. All of the 3% Mo specimens, and all but one of the 5% Mo specimens, disintegrated. With the remaining specimens, it is possible to see the increased damage resistance with increasing molybdenum content (Specimens 1, 2, and 3). The rapid increase in damage or density decrease occurring at this level of burnup is shown by comparison between the two 10% Mo specimens (Specimens 5 and 6) and the two 7.5% Mo specimens (Specimens 7 and 8). In both cases, the specimens were beginning to swell rapidly, much the same as the 3 and 5% Mo specimens did in Capsule 2.



a. Capsules 2 and 5

b. Capsules 3 and 4

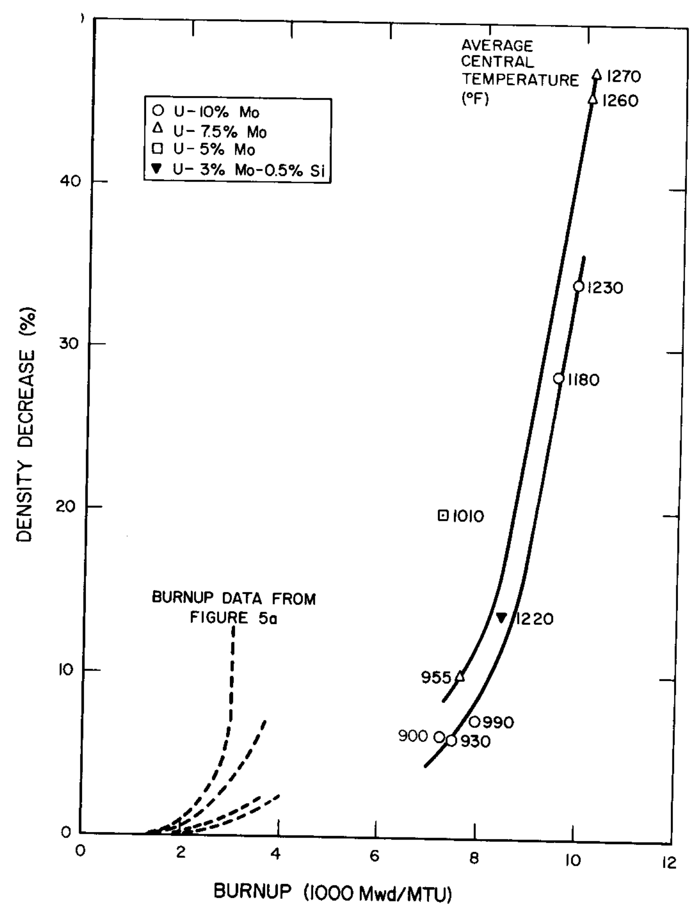


Figure 5. Amount of Swelling as Function of Burnup

C. NAA-38-4

This capsule was designed to show the effect of silicon and aluminum additions on the basic U - 3% Mo composition. Three U - 10% Mo specimens were included for comparison.

As with Capsule 3, the burnup was too high for all the 3% Mo specimens, except Slug 11, to yield accurate density decreases. Comparison of the diameter changes, however, showed a slight improvement resulting from the additions. Specimens 1, 4, and 8, of the basic composition, all show greater changes than corresponding alloyed slugs. It is not possible to differentiate between the silicon and aluminum; and, in fact, Specimens 6 and 9 showed almost identical dimensional changes at the same burnup.

D. NAA-38-5

This low-burnup version of Capsule 4 showed similar trends, although the damage was very much smaller, and differences were barely distinguishable. No specimen was swollen badly; but, as in Capsule 2, there were signs that the damage would increase rapidly at higher burnups. This can be seen from a comparison of Specimens 3 and 8, where a small increase in burnup of No. 8 resulted in a disproportionately large density decrease.

E. COMPOSITE RESULTS

A comparison of all data from the four capsules has been made in Figures 5, 6, and 7. Figure 5a shows the density change, as a function of burnup for each composition, in the low-burnup capsules. Figure 5b is a similar plot for the two high-burnup capsules. It is at once evident that the U - 3% Mo specimens have reached their limit of burnup at 3000 Mwd/MTU. Specimens having the same composition, with aluminum or silicon additions, all fall below this line; but there is insufficient data to predict the life of this fuel with any certainty. Apart from Slug No. 11 (NAA-38-4), which shows only 13.6% density decrease after 8450 Mwd/MTU burnup, the increase in life would be expected to be only of the order of 1000 Mwd/MTU. Slug No. 11 is the only 3% Mo specimen of the high burnup series that could be measured for density decrease. The others in the capsule were badly swollen and cracked, and not much reliance can be placed on the measured diameter changes. The performance of Slug No. 11 is even more remarkable, when it is seen that the temperature of the irradiation was higher than the low-burnup series in Capsule 5.

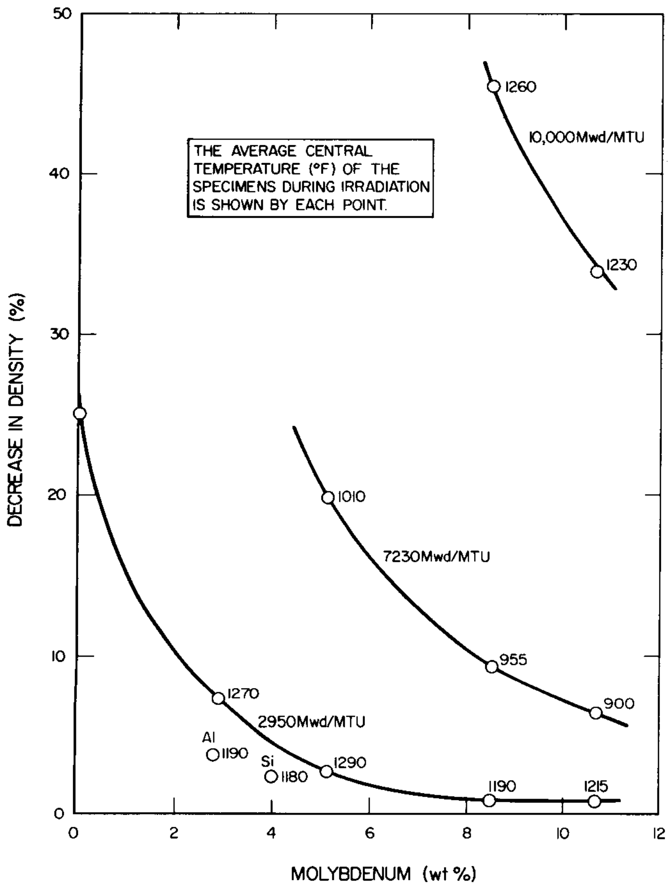


Figure 6. Effect of Molybdenum Content on Swelling of Uranium Alloys

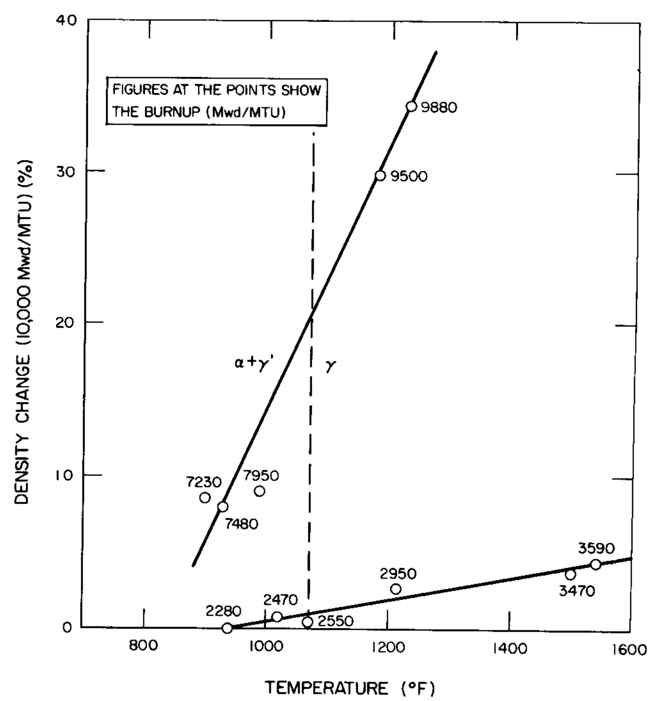


Figure 7. Density Change vs Temperature for U - 10 Mo

There are four specimens of U - 5% Mo, but only one of these is from a high burnup capsule, and it is interesting to note that the swelling for this slug is less than would be expected from an extrapolation of the low-burnup slugs. The U - 7.5% Mo and U - 10% Mo curves could also be interpreted in a similar manner, although there is no justification for this with the evidence available. The reason for this apparent anomaly may possibly be found in the temperatures at which the capsules were running. Generally, the temperature of the low-burnup slugs (Figure 5a) was directly proportional to the burnup received. Figure 5a is therefore a plot of density change against a function of both burnup and temperature. The high-burnup slugs (Figure 5b), being in a different capsule, have a different correspondence between burnup and temperature; and usually the temperatures are lower than in the corresponding low-burnup capsule. Consequently, the swelling is less than would be expected from a straight extrapolation. If swelling is plotted against temperature, as for the U - 10% Mo specimens in Figure 7, it can be seen that burnup is the dominant effect; but the limited data precludes complete separation of the two factors.

The effect of composition can be seen in Figure 6, where density change for a given burnup is plotted against the percentage of molybdenum in the alloy. Three irradiation levels were used, 2950, 7230, and 10,000 Mwd/MTU. Where there was no specimen irradiated to exactly this burnup, the nearest specimen to it was taken; and the density change was normalized to the standard value. Since the density change is not a linear function of burnup, this procedure leads to some error, but the changes necessary are very small. It is seen that, at the low burnup, there is initially a rapid decrease in the irradiation damage as the molybdenum content is increased. For low burnups, there is little to be gained by further increases in alloying additions above about 7 or 8% Mo. At the higher level of burnup, however, there is a significant improvement gained by increasing the molybdenum content beyond 8%. If the curve does level out, it is at some higher molybdenum content not considered in this experiment. The significance of the aluminum and silicon additions can be seen clearly from the two isolated points below the low burnup curve. The molybdenum content of these two specimens was different from the nominal 3%; but, in both cases, the additions are equivalent to 1 to 1-1/2% Mo.

IV. DISCUSSION

A considerable amount of data has been accumulated in this series of irradiations, and an attempt has been made to assess the influences of burnup and composition on the dimensional changes taking place. Due to the complexity of this type of experiment and the interdependence of the many variables, great care must be taken in trying to isolate the effect of any particular variable.

A common form of presentation is to plot density change as a function of burnup, as in Figure 5. Even this is not as simple as it may seem. Not only burnup, but also temperature, is being taken into account; since, in an experiment of this type, where many slugs are in one capsule, the temperature obtained is proportional to the total burnup. This fact arises because temperature is proportional to power generation in a capsule where the geometry and heat barrier are the same for all specimens. The power produced is a direct function of fission rate, which in turn is proportional to burnup, since all the specimens are in pile for the same time. Thus, if the density change of specimens in a particular capsule is plotted against burnup, the high burnup specimens are also the ones operating at high temperature. The interpretation of the data is thus complicated by the interdependence of the two factors. Further confusion arises when data from two or more capsules is plotted on the same graph. In the second capsule, the relationship between burnup and temperature can be expected to be different, and this can account for an apparent lack of continuity between the two sets of data. It is sometimes possible to separate such effects by the use of a multiple-correlation technique. In the present experiment, this is not possible. The proportionality between burnup and temperature in a particular capsule means that we can only consider these two variables as a single combined function, and not as two independent variables.

The possibility of correlating swelling with burnup rate is dismissed in a similar way, since burnup rate is simply calculated by dividing burnup by the time in pile, and is another measure of burnup for a particular capsule.

The composition of the specimens is a truly independent variable, and so we can judge the effect of molybdenum content on the swelling resistance of the alloy. To get a true picture, however, we must take care to compare only compositions having the same burnup.

Figure 6 includes a point representing pure uranium. This was taken from a similar capsule irradiation, NAA-38-1.[1] The material used in this experiment was high-purity uranium, and the data used in Figure 6 was obtained by extrapolating the measured density changes of the cast and beta-quenched specimens to a burnup of 2950 Mwd/MTU. This extrapolation, although not very accurate, serves to show the large improvements obtained by the addition of quite small amounts of molybdenum, especially at low burnups. With increasing burnup, more and more molybdenum is required to provide the same level of resistance.

V. CONCLUSIONS

The swelling resistance of uranium is markedly increased by molybdenum additions; 3 wt% Mo reduced the swelling from about 25% to 7% for 2950 Mwd/MTU burnup at about 1270°F central temperature. Larger additions reduce the swelling further, but the effect saturates at a constant swelling with 7 or 8% Mo at low burnups.

The minimum amount of molybdenum required to produce a given swelling resistance increases with the burnup under the conditions of fission rates experienced in these tests. At 7000 Mwd/MTU, slightly more than 10% would appear to be necessary before the swelling vs burnup curve flattens out. At 10,000 Mwd/MTU, large amounts of swelling occur, even with 10% Mo, and it would be necessary to use even higher molybdenum contents to produce the same amount of swelling resistance.

For all compositions, the swelling first increases slowly with increasing burnup, but eventually a point is reached where rapid swelling occurs. This point varies for each composition, being at higher burnups for the higher molybdenum alloys. For the 3% alloy, it is at about 3,000 Mwd/MTU; while, for the 10% alloy, it is at about 9,000 Mwd/MTU.

The temperature effect is significant; but, with the data available, it has not been possible to separate this unequivocally from the burnup effect.

Although some specimens were in the γ phase, no difference could be seen in the behavior of these and those in the $\alpha + \gamma'$ phase.

The burnup rate effect could not be separated from the temperature or total burnup effects, in the 10% molybdenum specimens in these irradiations.

The aluminum and silicon additions have a significant effect on swelling resistance, and can be regarded as equivalent to about 1 to 1-1/2 wt% molybdenum. Distinction, as to the relative merit of the two additions, is not warranted by the results of these irradiations.

None of the alloys considered is regarded as a potential fuel material for the Advanced Sodium Cooled Reactor. In no case was an alloy capable of operating satisfactorily at burnups of 10,000 Mwd/MTU and temperatures up to 1500°F.

The U - 3% Mo alloy would be a useful fuel, up to burnups of 3,000 Mwd/MTU and temperatures of 1300°F. The addition of small amounts of aluminum or silicon can be expected to raise this limit to at least 3,500 Mwd/MTU. The specimens used in these irradiations were not heat treated, to produce a very fine dispersion; and, if this were done, the improvement might be much more marked.

REFERENCES

1. D. G. Harrington and J. B. Newton, "Capsule Irradiation of Unalloyed Uranium at High Temperature," NAA-SR-7098 (April 1962)

2. B. R. Hayward and L. E. Wilkinson, "Radiation Behavior of Metallic Fuels for Sodium Graphite Reactors," NAA-SR-3411 (1959)

3. G. W. Greenwood, J. Nuc. Mat., 6 (1962) p 26

4. A. A. Shoudy, W. E. McHugh, and M. A. Silliman, "The Effect of Irradiation Temperature and Fission Rate on the Radiation Stability of Uranium - 10 wt% Molybdenum Alloy," Symposium of Radiation Damage, I. A. E. A., Venice (May 1962)

5. S. T. Konobeevski et al., First Geneva Conference, 8 (1955) P/681

6. M. L. Bleiberg, L. I. Jones, and B. Lustman, "Phase Changes in Pile Irradiated Uranium Base Alloys," J. App. Phys., 27 (1956) p 1270

7. G. W. Greenwood, A. J. E. Foreman, and D. E. Rimmer, J. Nuc. Mat., 1 (1959) p 305

8. D. Kramer, J. Nuc. Mat., 4 (1961) p 281

9. D. G. Harrington, "MTR Metal Fuel Tests for Advanced Sodium Graphite Reactors," NAA-SR-Memo 3215 (January 1959)