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MASTER
EVALUATION OF THORIUM-URANIUM ALLOYS
FOR THE UNCLAD-METAL BREEDER REACTOR

G. L. Copeland

OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

for the

U.S. ATOMIC ENERGY COMMISSION

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JUNE 1970

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ABSTRACT

The literature on thorium-uranium alloys was surveyed for information relative to the use of a Th-20% U alloy for the fuel element for the Unclad-Metal Breeder Reactor. The applicable physical, fabrication, and irradiation properties for a range of thorium-uranium alloy compositions are summarized. Several methods are available for manufacturing the fuel element, but the feasibility of most of the proposed methods has not been demonstrated for this alloy. Limited data indicate that the alloy will have the irradiation stability to reach the design life of 33,000 Mwd/metric ton at $\leq 650^{\circ}\text{C}$. The distribution of the phases in the alloy is determined by the fabrication technique and will influence the properties of the fuel element. Compatibility of the fuel with the sodium coolant was not considered for this report.

INTRODUCTION

This report presents the results of a survey of the literature on thorium-uranium alloys for information relative to fabrication and irradiation performance of the fuel element proposed for the Unclad-Metal Breeder Reactor (UMBR). The UMBR is a sodium-cooled breeder reactor proposed as a low-cost source of power and/or process heat for desalination.¹ The basic fuel element, shown in Fig. 1, is a hexagonal prism, about 18 in. long, that contains hexagonal coolant channels. Although several variations are proposed for the core, this report considers only the use of a Th-20% U alloy for that version of the reactor primarily intended for producing power. The coolant channels are to be 0.244 in. across the flats, and the web is to be 0.108 in. thick. The maximum fuel temperature is to be 650°C , and the average burnup is expected to be 33,000 Mwd/metric ton.

Fig. 1. Basic Fuel Element for UMBR.

The purposes of this report are to summarize the characteristics of the thorium-uranium alloy, evaluate the possible techniques for producing the fuel element, and summarize the irradiation performance data. Corrosion is not considered.

PHYSICAL METALLURGY

Phase Diagram

The phase diagram shown in Fig. 2 is taken from a recent report by Carlson and Stevens² and incorporates the contributions of several investigators.³⁻⁶ The liquid immiscibility loop is outside the composition range of interest to this study. The uranium-rich eutectic at 1085°C is important in the casting of alloys containing more than about 3% U. The eutectic, being the last liquid to freeze, is in the grain boundaries of the thorium-rich phase. With increasing uranium content,

Fig. 2. Phase Diagram for Thorium-Uranium. [Reprinted from O. N. Carlson and E. R. Stevens, A Compilation of Thorium Binary Phase Diagrams, IS-1752 (January 1968).]

the uranium-rich phase tends to become continuous. There are no compounds in the system.

Microstructure

The cast microstructure of the Th-20% U alloy consists of grains of the thorium-rich phase with the uranium-rich phase in the grain boundaries, as would be expected from the phase diagram. As shown in Fig. 3, the uranium phase is interconnected; however, the thorium does not appear to be completely enveloped by the uranium phase. This observation is supported by irradiation tests that indicate that the thorium phase controls the irradiation behavior at uranium concentrations up to about 25 wt % (ref. 7). Rolling tends to redistribute the uranium-rich phase, as shown in Figs. 4 and 5. The uranium-rich phase now appears as discrete small particles. This structure should offer very good irradiation performance within the range of conditions where thorium performs well.

Fig. 3. Microstructure of Th-20% U Alloy As Cast. Polished. 500X.

Fig. 4. Transverse Microstructure of Th-20% U After 94% Reduction. Annealed 1.5 hr at 750°C. Electrolytically etched. 500X.

Fig. 5. Longitudinal Microstructure of Th-20% U After 94% Reduction. Annealed 1.5 hr at 750°C. Electrolytically etched. 500x.

Metallography

Thorium and thorium-uranium alloys may be prepared for metallography by standard techniques of fine polishing. The specimen should be examined soon after final polishing to avoid staining from formation of the oxide film. Conventional etchants for thorium gave satisfactory results on the thorium-uranium alloys as cast, but the fine structure after working was more difficult to etch. Satisfactory results were obtained with the electrolytic etching procedure developed by Bentle.⁵ The polishing follows standard procedure with 1- μ m diamond paste as the final abrasive. The etchant solution is 95 vol % glycerine and 5 vol % HNO_3 . The etching conditions are 0.3 to 0.5 amp/cm² for 10 to 15 sec followed by a rinse in a solution of ethyl alcohol with 8 vol % glycerine.

Physical Properties

A survey of the mechanical and physical properties of thorium and thorium alloys was published recently by Hammond.⁸ Tables 1 and 2 list

Table 1. Physical Properties of Thorium

Property	Values
Crystallography	
Crystal structure $\leq 1375^{\circ}\text{C}$	Face-centered cubic
Lattice parameter, A	$a_0 (25^{\circ}\text{C}) = 5.0843 \pm 0.002$
Crystal structure $> 1375^{\circ}\text{C}$	Body-centered cubic
Lattice parameter, A	$a_0 (1450^{\circ}\text{C}) = 4.11 \pm 0.01$
Density, g/cm ³	
Theoretical	11.72
Calcium-reduced (as cast)	11.5 to 11.6
Arc-melted iodide	11.66
Elastic constants, 25°C	
Modulus of elasticity, psi	10.3×10^6
Shear modulus, psi	4.1×10^6
Poisson's ratio	0.27
Compressibility, cm ² /dyne	16.4×10^{-3}
Melting point, $^{\circ}\text{C}$	1750
Boiling point, $^{\circ}\text{C}$	3300 to 4500
Heat of fusion, kcal/mole	< 4.6
Heat of sublimation, kcal/mole	131
Heat of vaporization, kcal/mole	< 170
Vapor pressure 1757 to 1956°K	$\log P (\text{atm}) = -\frac{28780}{T (^{\circ}\text{K})} + 5.991$
Superconducting temperature, $^{\circ}\text{K}$	1.3 to 1.4
Magnetic susceptibility of iodide grade, cgs units	
20 $^{\circ}\text{C}$	0.54×10^{-6}
-183 $^{\circ}\text{C}$	0.64×10^{-6}
Work function, ev	3.39
Hall coefficient, cm ³ /coulomb	$-8.8 \times 10^{-5(a)}$
Emissivity	
1025 to 1425°C ($\lambda = 6670 \text{ A}$)	0.38
Molten ($\lambda = 6500 \text{ A}$)	0.40

^aThis value corresponds to a density of conduction electrons of 7.1×10^{22} electrons/cm³ or 2.4 electrons/atom.

From J. P. Hammond, Physical, Mechanical, and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).

Table 2. Properties of Thorium at Various Temperatures

Temperature (°C)	Thermal Conductivity ^a (cal sec ⁻¹ cm ⁻¹ °C ⁻¹)	Electrical Resistivity ^a (μΩ-cm)	Mean Coefficient of Thermal Expansion ^b (10 ⁻⁶ /°C)
25		16.95	
100	0.090	21.45	
200	0.093	27.35	
300	0.096	33.20	
400	0.100	38.85	
500	0.102	44.20	
600	0.106	49.20	
650	0.108		
700		53.80	
800		58.04	
900		62.00	
20-100			11.55
30-300			11.65
30-500			11.9
30-700			12.5
30-900			12.5
30-1000			12.65

^aO. N. Carlson et al., "The Metallurgy of Thorium and Its Alloys," pp. 74-106 in Proc. Intern. Conf. Peaceful Uses Atomic Energy, Geneva, 1955, Vol. 9, Columbia University Press, New York, 1956.

^bAmes Laboratory, Semi-Annual Summary Research Report in Physics for January-June 1959, IS-14 (October 1959).

From J. P. Hammond, Physical, Mechanical, and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).

the physical constants of thorium metal. Table 3 lists the thermodynamic functions.

The manner in which uranium additions affect these properties depends on the distribution of the uranium phase, which in turn is governed by the composition of the alloy and metallurgical treatment. Bentle⁵ has calculated the thermal conductivity for several thorium-uranium alloys. These are presented in Fig. 6. The densities of several thorium-uranium alloys are given in Table 4. The lower densities for the Ames Laboratory thorium were attributed to higher oxide content.⁹

Table 3. Thermodynamic Functions of Thorium

Temperature		Value of Function, ^a cal mole ⁻¹ °C ⁻¹				
°C	°K	C _p	C _v	S°	(H° - H° ₀)/T	-(F° - H° ₀)/T
-253	20	1.106	1.106	0.410	0.304	0.106
-223	50	4.048	4.042	2.770	1.801	0.969
-173	100	5.482	5.460	6.135	3.369	2.766
-123	150	5.965	5.927	8.460	4.164	4.296
-73	200	6.217	6.161	10.215	4.648	5.567
-23	250	6.392	6.318	11.623	4.980	6.643
25.00	298.16	6.532	6.441	12.760	5.220	7.540
27	300	6.538	6.446	12.799	5.228	7.571

^aM. Griffel and R. E. Skochdopole, "The Heat Capacity and Entropy of Thorium from 18 to 300°K," J. Am. Chem. Soc. **75**, 5250-5251 (1953).

From J. P. Hammond, Physical, Mechanical, and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).

Fig. 6. Thermal Conductivity of Thorium-Uranium Alloys. [Reprinted from G. G. Bente, A Physical Metallurgical Study of Thorium-Rich Thorium-Uranium Alloys, NAA-SR-2069 (1958).]

Table 4. Densities of Arc-Melted Thorium-Uranium Alloys^a

Alloy Composition	Density, g/cm ³	
	Iodide Thorium Alloys	Ames Thorium Alloys
Thorium	11.71	11.60
Thorium-5 w/o Uranium	11.88	11.79
Thorium-10 w/o Uranium	12.10	12.01
Thorium-15 w/o Uranium	12.33	12.30
Thorium-20 w/o Uranium	12.58	12.59

(a) Cold Reduced 80 per cent by swaging prior to measurement.

Reprinted from M. S. Farkas et al., "Development of Thorium-Uranium and Thorium-Uranium Base Alloys for Breeder Applications," pp. 468-497 in Proceedings of the Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, December 5-7, 1962, TID-7650, Book 2 (July 1963).

The effects of irradiation on the physical properties of thorium-uranium alloys also depend on the distribution of the uranium phase. Bowman and his co-workers¹⁰ estimated the thermal conductivity of thorium from measurements of electrical resistivity after cyclotron irradiation. The results, given in Table 5, show that the decrease in thermal conductivity tends to a maximum of 5 to 8%.

Thermal expansion coefficients of over $25 \times 10^{-6}/^{\circ}\text{C}$ were observed for an irradiated Th-11% U alloy.¹¹ This is more than twice the value usually found for unirradiated material. The increase was attributed to expansion and contraction of internal pockets of fission gas.

Mechanical Properties

Several laboratories have investigated the effect on mechanical properties of uranium additions to thorium and the effects of ternary and quaternary additions to the thorium-uranium alloys. The prime objective of most of this work was to increase the mechanical strength of the alloys. The tensile properties of thorium as a function of temperature are given in Figs. 7 and 8. Hammond⁸ summarized the properties

Table 5. Effect of 38-Mev Alpha-Particle Bombardment on the Thermal Conductivity of Thorium Wires^a

Temperature (°C)	Conductivity, $w\text{ cm}^{-1}\text{ }^{\circ}\text{C}^{-1}$		Decrease at Saturation (%)
	Unirradiated	Irradiated ^b	
300	0.402	0.372	7.5
500	0.431	0.406	5.8

^aComputed from the Wiedemann-Franz ratio and electrical-resistivity data. F. E. Bowman *et al.*, Cyclotron Irradiation Damage of Thorium, Stainless Steel, and Zirconium, NAA-SR-287 (April 1, 1954).

^bSaturation value, assuming no back annealing.

Reprinted from J. P. Hammond, Physical, Mechanical, and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).

Fig. 7. Changes in Some Tensile Properties of Thorium with Temperature of Testing. [Figure from O. N. Carlson *et al.*, "The Metallurgy of Thorium and Its Alloys," pp. 74-106 in Proc. Intern. Conf. Peaceful Uses Atomic Energy, Geneva, 1955," Vol. 9, Columbia University Press, New York, 1956; reprinted from J. P. Hammond, Physical, Mechanical, and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).]

Fig. 8. Effect of Temperature on Tensile Strength and Ductility of Wrought-Annealed and As-Extruded Calcium-Reduced Thorium. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

of thorium metal. The properties of thorium depend on the method of preparation and impurity content so that independent data cannot be easily compared for various effects. In general, the creep properties are good. To attain appreciable creep in Ames Laboratory thorium at 300°C, the stress must be within 2000 psi of the ultimate strength.

The impact strength varies with material and impurity content. One investigation¹² with Charpy V-notch specimens showed cast Ames Laboratory thorium to exhibit a gradual change from brittle to ductile fracture between 100 and 500°C. As shown in Fig. 9, Izod-type specimens showed a more narrow transition band centered at 50°C for Ames Laboratory thorium, whereas, iodide thorium showed no transition and increasing energy at lower temperatures.¹³

The effects of the interstitial solutes (carbon, nitrogen, and oxygen) are shown in Fig. 10. The normal ranges of these impurities normally found in calcium-reduced metal are indicated in the graph. Carbon, with a room-temperature solubility of 0.35%, provides solid-solution hardening. Oxygen and nitrogen appear to have a negligible effect in this range. Nitrogen in larger amounts moderately raises the tensile and yield strengths and the hardness of thorium.

Fig. 9. Effect of Temperature on Impact Strength of Thorium Showing the Ductile-to-Brittle Transition. [Figure from J. A. Milko, Metallurgy Div. Semiann. Progr. Rept. Oct. 10, 1953, ORNL-1625, p. 7; reprinted from J. P. Hammond, Physical, Mechanical, and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).]

Figure 11 shows the effects of uranium additions on the tensile properties of thorium at room temperature.¹⁴ The effects of uranium to 10 wt % as well as the effects of additional elements are shown in Table 6. Data from other investigators^{15,16} are given in Table 7. The strength of the alloys increases with increasing uranium content, especially at lower concentrations.

The creep strength, however, is lower for high-uranium concentration. As shown in Figs. 12 and 13, the creep resistance of thorium-uranium alloys decreases as the uranium content increases from 5 to 20 wt %. This decrease is attributed to the increasing amounts of the weaker uranium phase in the grain boundaries.⁹ The creep strengths of thorium-uranium ternary alloys are shown in Figs. 14 and 15. The strongest alloys in creep contained beryllium, molybdenum, or niobium. The strongest alloys in the tensile tests (10% Zr) were the weakest in creep. The disappointing creep properties of the zirconium alloys may be related to the phase transformation that occurs below 600°C or to the

Fig. 10. Effect of Some Interstitial Solutes Normally Found in Bomb-Reduced Thorium on the Room-Temperature Tensile Properties and Hardness of Iodide Thorium. Base metal compositions and metallurgical treatments: BMI - 0.012% C, 0.080% O; forged and hot rolled at 700°C, cold reduced 50%, annealed in argon 2 hr at 850°C. [Ref: R. M. Goldhoff et al., A Study of the Strengthening of Thorium by Alloying, Cold Work, and Aging, BMI-776 (Nov. 1, 1952).] ORNL - 0.015% C, 0.080% O; arc-melted, cold reduced 85%, vacuum annealed 0.5 hr at 650°C. (Ref: J. A. Milko, R. E. Adams, and W. O. Harms, "The Mechanical Properties of Thorium and High-Thorium Alloys," pp. 182-216 in The Metal Thorium, Proc. Conf. Cleveland, 1956, American Society for Metals, Novelty, Ohio, 1958.)

Fig. 11. Effect of Uranium Content on Tensile Properties of Commercial-Grade Thorium. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

Table 6. Tensile Properties of Thorium-Base Alloys

Composition (Balance Th), w/o	Test Temp.	0.2 % Offset Yield, psi	Tensile Strength, psi	Elongation %, in/in
Ames Thorium	Room	34,900	37,700	13
5 U	Room	27,900	39,300	23
10 U	Room	26,900	37,700	13.5
20 U	Room	32,100	42,000	30
10 U-1.5 Mo	Room	26,400	41,000	15
10 U-2 Nb	Room	22,900	35,400	22
10 U-10 Zr	Room	35,200	53,200	12
10 U-10 Zr-2 Nb	Room	39,200	60,400	14
Ames Thorium	600 C	13,800	15,700	26
5 U	600 C	11,600	15,950	44
10 U	600 C	13,800	17,970	41
20 U	600 C	11,200	15,160	50
10 U-1.5 Mo	600 C	11,220	17,700	43
10 U-2 Nb	600 C	12,550	18,000	37
10 U-10 Zr	600 C	19,600	28,400	44
10 U-10 Zr-2 Nb	600 C	19,200	25,640	48
Ames Thorium	700 C	4,280	8,090	58
5 U	700 C	6,590	11,700	51
10 U	700 C	8,800	10,300	44
20 U	700 C	6,700	8,170	58
10 U-2 Nb	700 C	8,400	10,800	66
10 U-10 Zr	700 C	15,400	20,000	39
10 U-10 Zr-2 Nb	700 C	12,100	13,910	77
10 U-0.1 Be	700 C	8,610	11,200	47

Reprinted from M. S. Farkas et al., "Development of Thorium-Uranium and Thorium-Uranium Base Alloys for Breeder Applications," pp. 468-497 in Proceedings of the Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, December 5-7, 1962, TID-7650, Book 2, (July 1963).

Table 7. Mechanical Properties of Bomb-Reduced Thorium-Uranium Alloys^a

Analyzed Composition (wt % U)	Strength, psi		Elongation in 1 in. (%)	Reduction of Area (%)	Hardness (VHN)
	0.2% Offset Yield	Tensile			
Unalloyed	19,400	31,700	46	50	79
1.00	25,600	38,400	37	51	104
5.08	27,400	42,300	38	44	105
10.2	30,000	45,000	35	50	
20.6	30,800	47,600	38	41	122
30.9	36,100	55,800	24	34	131
40.6	38,500	62,400	28	35	143
51.2	40,100			23	146
49.1	43,500	66,500	10	26	146

^a Annealed 2 hr at 850°C

From J. P. Hammond, Physical, Mechanical, and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).

Fig. 12. Creep Strength of Thorium-Uranium Alloys at 600°C.
 [Reprinted from M. S. Farkas et al., "Development of Thorium-Uranium and Thorium-Uranium Base Alloys for Breeder Applications," pp. 468-497 in Proceedings of the Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, December 5-7, 1962, TID-7650, Book 2 (July 1963).]

Fig. 13. Creep Strength of Thorium-Uranium Alloys at 700°C.
 [Reprinted from M. S. Farkas et al., "Development of Thorium-Uranium and Thorium-Uranium Base Alloys for Breeder Applications," pp. 468-497 in Proceedings of the Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, December 5-7, 1962, TID-7650, Book 2 (July 1963).]

Fig. 14. Creep Strength at 600°C of Th-10% U Alloys Containing Indicated Additions. [Reprinted from M. S. Farkas *et al.*, "Development of Thorium-Uranium and Thorium-Uranium Base Alloys for Breeder Applications," pp. 468-497 in *Proceedings of the Thorium Fuel Cycle Symposium*, Gatlinburg, Tennessee, December 5-7, 1962, TID-7650, Book 2 (July 1963).]

Fig. 15. Creep Strength at 700°C of Th-10% U Alloys Containing Indicated Additions. [Reprinted from M. S. Farkas *et al.*, "Development of Thorium-Uranium and Thorium-Uranium Base Alloys for Breeder Applications," pp. 468-497 in *Proceedings of the Thorium Fuel Cycle Symposium*, Gatlinburg, Tennessee, December 5-7, 1962, TID-7650, Book 2 (July 1963).]

larger quantities of the uranium phase contained in the grain boundaries of these alloys.⁹

Tests of hot hardness have been used to scope the effects on thorium and thorium-uranium alloys of a wide variety of additions.^{9,17,18} Hammond⁸ compiled Table 8, which lists the hot hardness of several alloys at 600, 750, and 800°C. While these tests usually give a good indication of tensile strength, they may or may not give a reliable indication of creep strength, since a different strengthening mechanism may be involved at the lower strain rates. For instance, neither the good creep resistance of the Th-10% U alloys containing beryllium, molybdenum, or niobium nor the poor creep resistance of the 10% Zr alloys could be predicted from these data.

Since swelling during irradiation is basically a creep phenomena, the most reliable indicator of expected irradiation performance is creep strength. The structure of the alloy is then important from two standpoints. Well dispersed, small, discrete particles of the uranium phase will maintain the creep strength and allow the recoil fission fragments to lodge in the isotropic thorium phase, which is more resistant to damage.

RADIATION PERFORMANCE

Swelling and Growth

The potential of thorium-uranium alloys for fuel elements has been recognized for some time. Carroll¹⁹ irradiated aluminum clad Th-1.3% U plates to about 1 at. % burnup. Although dimensional and density changes were not measured accurately, the plates appeared to grow uniformly and swell about 2%. The irradiation performance of thorium alloys has been reviewed by Bauer *et al.*,²⁰ Olsen *et al.*,²¹ and more recently by Leggett and Kemper.²² The most comprehensive irradiation experiments on thorium-uranium alloys were done at Argonne National Laboratory (ANL).⁷ They evaluated rolled plates, swaged rods, and chill-cast rods in various compositions and conditions. The results are presented in Tables 9, 10, and 11. The growth of all of the specimens containing less than 25% U

Table 8. Thorium-Base Alloys in Order of Decreasing Hardness^a

Alloy ^a	Composition (wt %)	Hardness (kg/mm ²)
600°C		
ORNL ^b	4 In	91
BMI ^c	10 U-0.5 Al	75.7
BMI ^d	10 U-10 Zr-2 Nb	73.2
BMI ^c	10 U-0.1 Be	69.1
BMI ^c	10 U-0.2 C	67.3
BMI ^c	10 U-10 Y	66.1
BMI ^c	10 U-1.5 Mo-0.2 C	64.7
ORNL ^b	2 In	63
BMI ^c	10 U-0.2 Be	62.3
ORNL ^b	10 Zr	59
ORNL ^b	5 Zr	59
BMI ^c	10 U-2 Nb-0.2 C	58
BMI ^c	10 U-4 Nb	56.6
BMI ^d	10 U-10 Zr	54.5
ORNL ^b	5 Zr-0.2 C	54
ORNL ^b	20 Zr	54
ORNL ^b	5 V-0.2 C	50
BMI ^c	10 U-2 Mo	49.3
ORNL ^b	4 In-5 Zr	49
ORNL ^b	2 In-0.2 C	49
ORNL ^b	2 Cr-0.2 C	47
BMI ^e	20 U	46.2
BMI ^e	5 U	45.4
BMI ^e	10 U	45.4
ORNL ^b	2 Mo-0.2 C	44
ORNL ^b	5 U-0.2 C	44
ORNL ^b	0.2 Al-0.2 C	43
ORNL ^b	8 Nb	43
BMI ^d	10 U-2 Nb	42.7
BMI ^d	10 U-20 Zr	42.4
BMI ^d	10 U-1.5 Mo	41.1
ORNL ^b	0.2 Be-0.2 C	40
ORNL ^b	5 Ti-0.2 C	40
BMI ^e	15 U	39.2
BMI ^d	10 U-25 Zr	39.2
ORNL ^b	0.2 C	38
ORNL ^b	5 Ti	37
ORNL ^b	20 U	36
ORNL ^b	0.4 Al	35
ORNL ^b	8 Cr	35
ORNL ^b	0.2 Al	32
ORNL ^b	5 V	32
ORNL ^b	5 U	30

Table 8 (continued)

Alloy ^a	Composition (wt %)	Hardness (kg/mm ²)
ORNL ^b	8 Mo	30
ORNL ^b	0.15 C	29
ORNL ^b	2 Nb-0.2 C	29
ORNL ^b	10 U	28
ORNL ^b	4 Mo	27
ORNL ^b	4 Cr	27
ORNL ^b	4 Nb	26
ORNL ^b	2 Nb	26
ORNL ^b	2 Mo	24
ORNL ^b	0.10 C	23
ORNL ^b	thorium	22
ORNL ^b	2 Cr	21
ORNL ^b	0.2 Be	21
750°C		
ORNL ^b	4 In	55
ORNL ^b	5 Zr	37
ORNL ^b	4 In-5 Zr	34
ORNL ^b	5 V-0.2 C	33
ORNL ^b	2 In	31
ORNL ^b	5 Zr-0.2 C	31
ORNL ^b	5 U-0.2 C	26
ORNL ^b	20 Zr	25
ORNL ^b	10 Zr	25
ORNL ^b	8 Nb	25
ORNL ^b	5 Ti-0.2 C	24
ORNL ^b	5 Ti	23
ORNL ^b	2 In-0.2 C	23
ORNL ^b	0.2 Be-0.2 C	20
ORNL ^b	5 V	20
ORNL ^b	2 Cr-0.2 C	20
ORNL ^b	0.2 Al-0.2 C	20
ORNL ^b	0.4 Al	19
ORNL ^b	0.2 Al	19
ORNL ^b	20 U	18
ORNL ^b	8 Cr	18
ORNL ^b	5 U	18
ORNL ^b	8 Mo	17
ORNL ^b	2 Mo-0.2 C	17
ORNL ^b	4 Nb	17
ORNL ^b	10 U	15
ORNL ^b	4 Mo	15
ORNL ^b	2 Nb	14
ORNL ^b	0.2 Be	14
ORNL ^b	0.2 C	13

Table 8 (continued)

Alloy ^a	Composition (wt %)	Hardness (kg/mm ²)
ORNL ^b	4 Cr	13
ORNL ^b	2 Mo	13
ORNL ^b	0.15 C	11
ORNL ^b	thorium	11
ORNL ^b	2 Cr	11
ORNL ^b	2 Nb-0.2 C	11
ORNL ^b	0.10 C	9
800°C		
BMI ^c	10 U-0.2 Be	29.6
BMI ^c	10 U-0.1 Be	27.3
BMI ^c	10 U-0.5 Al	26.9
BMI ^c	10 U-1.5 Mo-0.2 C	24.3
BMI ^e	5 U	24.0
BMI ^c	10 U-0.2 C	22.5
BMI ^d	10 U-1.5 Mo	22.4
BMI ^c	10 U-10 Y	22.4
BMI ^d	10 U-10 Zr	22.2
BMI ^e	10 U	22.1
BMI ^c	10 U-4 Nb	21.3
BMI ^d	10 U-2 Nb	20.3
BMI ^c	10 U-2 Nb-0.2 C	19.8
BMI ^d	10 U-10 Zr-2 Nb	17.2
BMI ^e	20 U	14.0
BMI ^d	10 U-25 Zr	11.8
BMI ^d	10 U-20 Zr	10.7
BMI ^e	15 U	10.6

^a Nominal composition prepared by arc melting.

^b Ames thorium, tested in hot-rolled condition. Ref: J. A. Burka and J. P. Hammond, Evaluation of Thorium-Base Alloys for High-Temperature Strength, ORNL-3777 (April 1965).

^c Ames thorium, tested in as-cast condition. Ref: M. S. Farkas, A. A. Bauer, and R. J. Dickerson, "Development of Thorium-Uranium and Thorium-Uranium-Base Alloys for Breeder Applications," pp. 468-497 in Proceedings of the Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, December 5-7, 1962, TID-7650, Book 2 (July 1963).

^d Iodide thorium, tested in as-cast condition. Ref: Ibid.

^e Ames thorium, tested in hot-rolled condition. Ref: Ibid.

Reprinted from J. P. Hammond, Physical, Mechanical and Irradiation Properties of Thorium and Thorium Alloys, ORNL-4218 (April 1968).

Table 9. Effects of Irradiation on Rolled Specimens

Spec. No.	Composition	Burnup, a/o	Length Change, %	Width Change, %	Thickness Change, %	Weight Change, mg	Density Change, %	Hardness Change, Rockwell 30T
3	Thorium (unalloyed)	0.12	-0.01	-0.28	-0.03	-0.4	0.08	26.5
1	Th-5 w/o U (natural U)	0.13	-0.02	-0.12	0	-0.1	0.08	15.5
2	Th-5.5 w/o U (93% enriched U)	1.4	0.11	0.40	0.06	0.3	-1.44	13.0

Reprinted from J. H. Kittel et al., Effects of Irradiation on Thorium and Thorium-Uranium Alloys, ANL-5674 (April 1963).

Table 10. Effect of Irradiation on Swaged Specimens

Spec. No.	U, w/o	Method of Preparation	Burnup, a/o	Irradiation Temp, °C		Length Change, %	Diameter Change, %	Hardness Change, R _A	Weight Change, mg	Density Decrease, %	% Δ Vol(a) a/o Burnup	Remarks
				Surface	Center							
H-10	0	Swaged at 25°C	1.2	76	88	-0.087	1.42	16.9	1.5	0.61	0.51	Bent during opening of capsule
H-14	0	"	1.5	82	96	0.18	0.68	16.3	1.4	1.09	0.77	
H-15	0	"	1.8	86	100	0.15	0.63	16.0	-0.1	1.45	0.97	
H-11	0	Swaged at 25°C, annealed at 700°C	0.26	75	87	0.24	-0.37	26.2	(b)	0.064	0.25	
H-12	0	"	0.34	81	95	0.14	0.47	34.3	(b)	0.85	2.5	
H-13	0	"	0.35	82	96	0.25	0.32	24.2	(b)	0.66	1.9	
H-6	0.13	Swaged at 25°C, annealed at 700°C	0.37	45	49	0.14	0.11	21.2	(b)	0.11	0.30	
H-5	0.13	"	1.6	45	49	0.47	0.63	26.2	2.1	1.07	0.69	
H-4	0.13	"	1.9	55	63	0.53	0.84	27.2	1.6	1.40	0.76	
H-3	1.39	Swaged at 25°C, annealed at 700°C	0.45	83	95	0.15	0.37	9.2	(b)	0.48	1.1	
H-2	1.39	"	0.69	98	115	0.21	0.84	16.5	(b)	0.70	1.0	
H-1	1.39	"	2.0	90	105	(c)	1.11	7.9	2.1	1.45	0.85	
H-9	5.53	Swaged at 600°C, annealed at 700°C	1.3	125	150	0.43	1.11	26.8	(b)	2.33	2.2	
H-8	5.53	"	2.8	110	130	0.17	1.74	15.9	1.0	3.56	1.3	
H-7	5.53	"	3.6	160	200	(c)	2.58	15.0	(b)	4.79	1.4	Fractured during opening of capsule

(a) Calculated from the expression $\% \Delta \text{Vol} = 100 \left(\frac{\rho_i}{\rho_f} - 1 \right)$, where ρ_i and ρ_f are the initial and final densities, respectively.

(b) Measurement not taken.

(c) Condition of specimen did not permit measurement.

Reprinted from J. H. Kittel et al., Effects of Irradiation on Thorium and Thorium-Uranium Alloys, ANL-5674 (April 1963).

Table 11. Effect of Irradiation on Cast Specimens

Spec. No.	U, w/o	Melt No.	Burnup, a/o	Midlength Irradiation Temp, °C		Length Change, %	Diameter Change, %	Weight Change, mg	Density Decrease, %	% Δ Vol(a) a/o Burnup	Remarks
				Surface	Center						
ANL-42-8-1	10.7	C16F-225	0.90	280	290	0.57	1.18	-1.7	1.24	1.4	Upper end swelled.
-4-1	9.43	-221	1.2	330	340	0.89	1.18	-0.6	2.31	2.0	
-6-1	9.43	-221	1.6	450	470	2.90	1.33	41.7	15.65 ^(b)	12	
-2-1	9.43	-221	2.2	650	680	3.25	4.59	-3.5	8.78	4.3	
-3-1	9.43	-221	2.9	370	380	1.87	1.18	-1.0	5.26	1.9	
-1-1	9.43	-221	3.9	510	530	2.24	1.33	-1.3	6.71	1.8	
-7-1	10.7	-225	4.0	420	440	1.97	1.46	-1.1	5.63	1.5	
-5-1	9.43	-221	4.0	540	560	2.27	0.91	-1.4	7.28	2.0	
CP-12-1	15.1	-218	0.16	605	650	0.03	0.20	1.4	0.34	2.1	Upper end swelled. Upper end swelled and broken off.
-11-2	15.1	-218	0.47	625	665	0.60	1.20	-0.2	1.80	3.9	
ANL-42-8-2	15.2	-224	1.0	290	300	0.52	1.75	-1.7	1.94	2.0	
-4-2	15.1	-218	1.4	390	410	0.82	1.45	-3.0	2.95	2.2	
-6-2	15.2	-224	1.8	520	540	3.81	2.87	56.3	3.56 ^(b)	2.1	
-2-2	15.1	-218	2.6	720	750	(c)	5.37	-550.8	11.12 ^(b)	4.8	
-3-2	15.1	-218	3.3	430	450	2.27	1.81	-1.6	5.74	1.8	
-7-2	15.2	-224	3.4	440	460	2.42	1.96	-1.2	7.93	2.5	
-1-2	15.1	-218	4.5	600	630	3.27	2.79	-1.9	9.45	2.3	
-5-2	15.1	-218	4.5	610	640	3.44	2.49	-1.5	10.32	2.5	
CP-12-4	20.4	-219	0.17	605	650	0.23	0.20	-1.0	0.28	1.7	Upper end swelled. Upper end swelled.
-11-5	20.6	-229	0.46	500	540	0.59	0.76	1.4	1.80	4.0	
ANL-42-8-3	20.6	-229	1.1	290	300	0.72	1.88	-0.8	1.85	1.7	
-4-3	20.4	-219	1.5	400	420	0.78	2.29	-3.9	2.48	1.7	
-6-3	20.6	-229	1.9	540	560	3.87	2.08	11.7	4.00 ^(b)	2.2	
-2-3	20.4	-219	2.7	740	770	3.76	7.82	-7.3	12.27 ^(b)	5.2	
-3-3	20.4	-219	3.5	440	460	2.99	2.16	-0.7	7.08	2.2	
-7-3	20.6	-229	3.5	450	470	2.87	1.43	-1.5	7.53	2.3	
-1-3	20.4	-219	4.5	610	640	2.91	3.35	-0.8	10.67	2.6	
-5-3	20.6	-229	4.7	620	650	5.69	2.98	-0.7	12.09	2.9	
ANL-42-8-4	25.2	-223	0.85	250	260	0.91	4.59	-1.4	1.78	2.2	Warped, slightly roughened surface.
-6-4	25.2	-223	1.8	510	530	-0.15	3.20	-0.6	3.45	2.0	
-4-4	25.3	-220	2.0	430	450	-0.51	2.22	-2.4	2.35	1.2	
-2-4	25.3	-220	2.5	620	650	2.49	14.98	21.3	12.30	5.6	
-3-4	25.3	-220	3.0	390	410	-2.25	4.67	-1.0	6.24	1.2	
-7-4	25.2	-223	3.4	430	450	2.22	4.32	-0.5	8.69	2.8	
-5-4	25.2	-223	4.2	580	600	4.72	3.74	-0.5	6.73	1.7	
-1-4	25.3	-220	4.7	640	670	6.63	5.68	1.3	15.25	3.9	
ANL-42-8-5	31.6	Y16F-190	1.7	490	510	-0.68	1.38	-2.0	4.11	2.5	Warped, roughened surface. Severely distorted. Severely distorted, sooty black surface. Severely distorted. Roughened surface. Severely distorted. Severely distorted.
-4-5	31.4	-189	3.4	690	720	5.05	5.64	-2.8	3.52	1.1	
-6-5	31.6	-190	4.1	860	900	17.2	12.0	45.9	24.81	8.0	
-2-5	31.4	-189	4.8	>1000	>1000	45.0	46.8	(c)	(c)	-	
-3-5	31.4	-189	6.5	690	720	18.1	11.5	4.2	26.86	5.6	
-7-5	31.6	-190	7.6	830	860	5.09	1.68	-0.1	22.22	3.7	
-5-5	31.4	-189	9.5	>1000	>1000	25.5	47.0	71.2	45.81	8.5	
-1-5	31.4	-189	10.1	>1000	>1000	27.5	33.3	60.1	46.36	8.4	

(a) Calculated from the expression $\% \Delta \text{Vol} = 100 \left(\frac{\rho_i}{\rho_f} - 1 \right)$, where ρ_i and ρ_f are the initial and final densities, respectively.

(b) Measurement taken after swelled end was removed.

(c) Condition of specimen did not permit measurement.

Reprinted from J. H. Kittel et al., Effects of Irradiation on Thorium and Thorium-Uranium Alloys, ANL-5674 (April 1963).

was fairly isotropic, and the specimens were not roughened on the surface or distorted. Distortion and roughening did occur in specimens of 25 and 31% U, probably in part because the uranium phase is more interconnected in these alloys and in part because of higher irradiation temperatures. Severe "mushrooming" occurred on the ends of some of these specimens, which were overheated because of a temperature stratification at the top of the NaK capsule.

Further work at ANL includes the irradiation of six thorium-uranium and thorium-uranium-plutonium fuel pins, sodium bonded to V-20% Ti jackets, in the CP-5 reactor. The capsule was removed in June 1967 after the instrumentation indicated a jacket failure. The design details and irradiation variables are given in Table 12. The burnups listed correspond to 3.1 to 3.6 at. % heavy metal. Neutron radiography later confirmed the jacket failure. Length changes measured from radiographs at 13.3 at. % (U + Pu) showed no elongation for the thorium-uranium alloys and 8% elongation for the thorium-uranium-plutonium pins.²⁴ Destructive evaluation of the assembly is planned and will be reported in the ANL Metallurgy Division monthly progress reports.

Argonne National Laboratory conducted transient experiments on Th-20% U fuel pins²⁵ in the Transient Reactor Test Facility (TREAT). Six fuel pins were run to a range of conditions ranging from no damage

Table 12. Status of Metal-Fuel Irradiations in CP-5

Specimen Number	Design Parameters ^a		Operating Conditions			
	Fuel Composition (w/o)	Effective Density (%)	Max kW/ft	Max Clad Temp (°C)	Burnup	
					a/o (U + Pu)	fiss/cc x 10 ^{-20b}
1N16	Th-20 U	74.7	8.7	590	25.1	12.1
4N19	Th-20 U	74.0	8.7	560	25.1	12.1
2N17	Th-10 Pu-10 U	74.1	9.2	590	28.6	13.5
5N20	Th-10 Pu-10 U	73.0	9.2	590	28.6	13.5
3N18	Th-10 Pu-20 U	73.5	8.7	560	17.0	12.4
6N21	Th-10 Pu-20 U	73.5	8.7	560	17.0	12.4

^aAll specimens are clad in 0.015-in.-thick V-20 a/o Ti alloy, with an OD of 0.198 in.

^bBased on effective density.

Reprinted from Argonne National Laboratory, Reactor Development Program Progress Report, June 1967, ANL-7349.

to extensive failure. Maximum cladding temperature reached 1330 to 1360°C, considerably in excess of the 1086°C solidus temperature. Since the fuel was still relatively intact at these temperatures of cladding failure, it appears that the continuous thorium matrix controlled the performance of the fuel under these transient conditions.

Atomics International (AI) irradiated Th-11% U specimens in NaK-filled capsules to burnups ranging from 0.3 to 1.5 at. % (refs. 26 and 27). They estimated the range of temperature to be 520 to 670°C. The specimens were cast and machined to size or cold reduced about 42% by swaging. The dimensional changes are presented in Table 13. Many of the specimens exhibited the mushrooming effect on the ends due to overheating caused by a drop in the NaK level. The density changes are somewhat higher than expected; examination of only the central sections avoided the mushroomed area. The high swelling may be due in part to irradiation temperatures, which may have been higher than reported. The difference in swelling per atom percent burnup varies greatly among experimental assemblies.

Figure 16 presents the swelling ratio for experimental assemblies 6 and 7 as a function of the estimated temperatures.²⁷ There appears to be a rapid increase in the swelling rate above about 1100°F (about 600°C). The swelling rates in general are higher than those reported by ANL and are believed to be due to the different uranium distributions, which will be discussed below. Further work by AI involved irradiating a full-sized Sodium Reactor Experiment (SRE) fuel element of Th-5.4% U in the SRE to about 0.13 at. % burnup. The results of dimensional measurements are in Table 14. The samples were in very good condition and looked essentially the same as when they were inserted.²⁸ A full core loading of Th-7.6% U was manufactured and irradiated as the second SRE core.²⁹⁻³² The fuel behaved well; however, the project was abandoned before appreciable exposure was attained (maximum burnup was about 0.5 at. %), and no detailed postirradiation examination was made.

Savannah River Laboratory (SRL) fabricated and irradiated two tubular Th-1.4 % U fuel elements clad with Zircaloy-2 for the heavy-water-moderated-and-cooled reactor concept.³³ These tubes were irradiated in the Heavy Water Components Test Reactor (HWCTR) at maximum core

Table 13. Dimensional Changes of Irradiated Specimens of Th-11% U

Measured Central Temperature (°F)		Calculated Surface Temperature (°F)		Burnup (total at. %) ^a	Maximum Increase in Diameter (%)	Increase in Volume (%) ^b	Specimen NAA-15-
Range	Average	Range	Average				
1100-1000	1059	899-820	866	0.3	1.6	0.47	5-5
1300-1110	1172	1060-905	957	0.3	0.8	0.73	5-1
1225-1195	1214	1000-975	991	0.3	1.1	0.86	5-2
1230-1215	1219	1005-992	995	0.3	1.5	1.9	5-3
				0.3	2.4	1.6	5-4
1135-1010	1071	925-825	876	0.55	0.7 ^c	2.9	7-6
1230-1160	1198	1005-950	979	0.60	1.4 ^c	4.7	7-5
1255-1115	1187	1025-910	970	0.63	1.3 ^c	4.1	7-4
1300-1090	1176	1060-890	961	0.65	d	d	7-3
1415-1163	1252	1150-950	1022	0.66	2.3 ^c	5.1	7-2
1495-930	1130	1220-760	924	0.68	4.4 ^c	7.7	7-1
	970 ^e		800 ^e	0.94	1.1 ^c	3.2	6-6
	1030 ^e		850 ^e	1.00	1.5 ^c	2.7	6-1
	1100 ^e		900 ^e	1.10	1.5 ^c	4.4	6-2
	1100 ^e		900 ^e	1.11	1.9 ^c	4.8	6-5
	1100 ^e		900 ^e	1.14	1.7 ^c	4.1	6-3
	1100 ^e		900 ^e	1.13	1.5 ^c	4.9	6-4

^aBased on chemical analyses of some specimens and heat balance on all specimens.

^bBased on density measurements of central sections.

^cBased on micrometer measurements.

^dSpecimen could not be removed from capsule.

^eBrief thermocouple life required that these temperatures be calculated.

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

Fig. 16. Density Change of Th-11% U Specimens as a Function of Irradiation Temperature. [Reprinted from J. E. Gates, G. E. Lamale, and R. F. Dickerson, The Examination and Evaluation of Irradiated Thorium-11 w/o Uranium Specimens, BMI-1334 (April 1959).]

temperatures of 465°C to maximum exposures of 3500 Mwd/metric ton enriched uranium (approx 0.37 at. % burnup). This test was interrupted when the HWCTR operation was terminated. The 10-ft-long elements were described as satisfactory with no distortion or warping. Postirradiation measurements indicated about 0.8% vol change in the region of highest burnup and temperature.

Battelle Memorial Institute Pacific Northwest Laboratory (BNWL) has irradiated "button-head" tensile specimens of Th-1, -4, and -5.4% U to burnups of 1.5 at. % in unmonitored NaK capsules at less than 250°C (ref. 22). Little or no warping occurred, and the specimens retained their original appearance. They have been irradiating tubular Th-2.5% U-1% Zr under water-cooled power reactor conditions in the

Table 14. Dimensional and Density Changes for Extruded and Swaged Th-5.4 wt % U

Slug Position	Slug No.	Warp (0.001 in.)	Length (in.)	Diameter (in.)	Density (g/cc)		Change (%)		
					Pre	Post	Diameter (ΔD)	Length (ΔL)	Density ($\Delta \rho$)
Nominal		3 max. single throw	6.000 ± 0.010	0.750 ± 0.002					
1	D-19	Less than 3	6.011	No measured change	11.90	11.89	No measured change	0.2	0.1
2	G-19		6.002		11.85	11.82		0	0.3
3	E-26		6.010		11.90	11.85		0.4	0.4
4	I-21		6.011		11.90	11.88		0.3	0.2
5	L-33		6.012		11.90	11.90		0.2	0
6	D-25		6.013		11.90	11.81		0.2	0.8
7	G-23		6.005		11.85	11.83		0.1	0.2
8	J-31		N.R.		11.90	11.86		N.R.	0.3
9	H-10		6.009		11.90	11.87		0.2	0.3
10	G-3		6.004		11.85	11.83		0.1	0.2
11	F-32		6.006		11.90	11.85		0.1	0.4
12	B-29		N.R.		11.90	11.90		N.R.	0
Average			6.006		11.89	11.86		0.2	0.3
Maximum			6.013		11.90	11.90		0.3	0.8
Minimum			6.002		11.85	11.81		0	0

Fuel Cluster MC-1-1

Rod No. 528

Channel R-44

Extruded and Swaged Th - 5.4 wt % U

Burnup - Average, 1000 Mwd/MTU; Maximum, 1300 Mwd/MTU

Reprinted from J. L. Ballif et al., Engineering Evaluation of a Mixed Alloy Fuel Element Irradiated at Elevated Temperatures in the SRE, NAA-SR-3888 (June 1960).

Engineering Test Reactor (ETR). These fuel elements have been clad with and bonded to Zircaloy-2 by the coextrusion process.³⁴ An interim evaluation of the test indicates that the behavior of these elements has been excellent to burnups of as much as 2.1 at. % (ref. 35). Irradiation data and swelling at burnups to 2.4 at. %, as measured from density measurements in the ETR basin, are presented in Table 15. Dimensional changes from an in-cell examination are presented in Table 16 and compared to the swelling as measured from density measurements in the ETR basin.³⁶ After exposures up to 21,000 Mwd/metric ton, the specimens are still showing near the minimum theoretical swelling, and indications are that the goal exposure of about 26,500 Mwd/metric ton can be attained easily.

The swelling rate of thorium containing up to 20% U as a function of temperature for irradiation data and postirradiation anneals is

Table 15. Irradiation Data on Th-2.5% U-1% Zr Tubular Fuel Elements

GEH-10	Percent ^a Fuel Swelling	Burnup		Surface Temp., °C	Maximum Core Temp., °C	Current Specific Power, W/g	Surface Heat Flux, ² Btu/(hr)(ft ²)	Thermal ^b Cycles
		Fissions/cm ³ , MWd/T	at. %					
65	3.9	7.2×10^{20} (21,300)	2.4	295	425	31	4.5×10^5	419
64	2.2	5.1×10^{20} (15,100)	1.7	285	390	24	3.4×10^5	358
71	1.8	4.2×10^{20} (12,400)	1.4	295	445	34	5.0×10^5	219
72	0.1	1.5×10^{20} (4,300)	0.49	300	490	43	6.2×10^5	103
70	-0.5	1.3×10^{20} (3,900)	0.45	295	470	39	5.6×10^5	103
84	0.1	0.9×10^{20} (2,500)	0.29	290	440	33	4.8×10^5	58

a. Fuel swelling is determined by weighing the elements under water at the end of each reactor cycle.

b. A thermal cycle is defined as a change of reactor power from less than 10 to 90% or greater and back to less than 10%.

Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).

Table 16. Dimensional Changes in Th-2.5% U-1% Zr Fuel Elements

Element No., Exposure	ΔL , ^a in.	Measuring Position	ΔOD , ^b in.	ΔID , ^c in.	Δ Air Weight, ^d g	% $\Delta V/V$ ^e Cell	% $\Delta V/V$ Basin
GEH-10-65 (18,600 MWd/T)	+0.062	End	+0.012	-0.001	-0.10	3.3	3.4
		Center	+0.016	0			
		End	+0.011	0			
GEH-10-62 (13,000 MWd/T)	+0.039	End	+0.006	0	+0.70	2.1	1.7
		Center	+0.012	0			
		End	+0.007	0			
GEH-10-71 (9,400 MWd/T)	+0.033	End	+0.006	+0.001	+0.30	1.5	1.3
		Center	+0.009	-0.001			
		End	+0.004	0			

a. Original length, ~ 8.5 in.

b. Original outer diameter, ~ 1.76 in.

c. Original inner diameter, ~ 1.06 in.

d. Original weight, ~ 2170 g

e. Original volume, ~ 205 cm³

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shown in Fig. 17. These data indicate that up to about 650°C, the ratio of the percentage of volume change to burnup in atomic percent, the so called "R" value, is between 1.5 and 2. This R value of about 2 represents the theoretical minimum that could be expected for accommodation of solid fission fragments in the thorium lattice. That AI data tend to lie above the other data may be due to the actual temperatures being higher than the estimates made by AI or due to the larger particles of uranium in the AI material, which would retain more of the fission fragments. The theoretical minimum R value for solid fission fragments in uranium is about twice that for thorium.

Fig. 17. The Temperature Dependence of Swelling in Thorium and Thorium Alloys. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

The probability, $\bar{F}$, of escape of recoil fission fragments with a range r ($r \approx 6.6 \mu\text{m}$ in uranium) in spherical particles of diameter D has been given as³⁷

$$\bar{F} = \frac{3}{4} \left(\frac{2r}{D} \right) - \frac{1}{16} \left(\frac{2r}{D} \right)^3. \quad (1)$$

Kittel et al.⁷ list a relationship for a slab geometry that is typical of the uranium phase in cast thorium-uranium alloys. The probability of recoil escape from particles with slab geometry and thickness t is

$$\bar{F} = r/2t, \quad t \geq r, \quad (2)$$

$$\bar{F} = 1 - t/2r, \quad t \leq r. \quad (3)$$

A plot of this function for uranium particles is shown in Fig. 18. This curve does not take into account the probability of a fission fragment leaving one particle and lodging in another. A small percentage of the fission fragments will remain in the $< 1\text{-}\mu\text{m}$ -thick network of uranium at the grain boundaries in the ANL alloys, whereas, up to 15% could remain in the $10\text{-}\mu\text{m}$ -diam particles in the AI alloys. This difference may

Fig. 18. Probability of Escape of Fission Recoils from Uranium Particles with Slab Geometry as a Function of Particle Thickness. [Reprinted from J. H. Kittel et al., Effects of Irradiation on Thorium and Thorium-Uranium Alloys, ANL-5674 (April 1963).]

account for some of the extra swelling in the AI alloys. Most of the other alloys that have been irradiated had either very fine particles of uranium or the slab-shaped network at the grain boundaries.

In the temperature range above about 650°C the swelling behavior is not so well defined. The excess swelling, or that above the minimum R value of approximately 2, is due to the formation and growth of bubbles of fission gas. The diffusion of fission gases to the bubbles is controlled by vacancy mobility. The resistance that the matrix offers to the growth of the bubbles depends on the creep strength of the alloy at the temperature of the experiment. In view of the wide spread in creep properties of thorium and thorium alloys, the spread in R values above 650°C is not too surprising. Carefully controlled experiments are needed to define the swelling behavior of thorium and thorium alloys in the range of temperatures around 650°C and higher. A limited number of capsule experiments are under way at BNWL to provide some of these data.³⁸

Based on the fact that the melting point of an alloy provides a first crude estimate of vacancy mobility and hence of the diffusion of fission gas, Leggett proposed that the higher melting point of thorium may be largely responsible for its improved resistance over uranium to swelling at high temperatures.³⁹ He points out that the 950°C annealing temperature is the same homologous temperature as is 575°C for uranium. If data at this same homologous temperature are normalized to unit burnup, the respective R values for thorium and uranium are 24 and 21. The isotropic nature of thorium does contribute significantly to its irradiation stability since it obviates the mechanical volume changes brought about directly by the fission process in alpha uranium.⁴⁰ Post-irradiation annealing of specimens of Th-1 and 5.4% U irradiated to 1.5 at. % burnup at less than 300°C corroborated the effect of alloy melting point.²² The results of postirradiation anneals at several temperatures up to 900°C are given in Table 17. After the final treatment, the 1 and 5.4% U alloys had swelled, 3.2 and 9.0% per atom percent burnup, respectively. However, Fig. 19 shows that, on a homologous temperature scale, both alloys showed the same swelling behavior.

Table 17. Density and Hardness Changes in Thorium-Uranium Alloys During Postirradiation Annealing

Material	Condition	at.% Burnup	R_A	Density, g/cm ³	% Swelling $\frac{\rho_i - \rho_f}{\rho_f} \times 100$	"R" % Swelling at.% Burnup	H_T
Th-1 wt% U (mp $\approx$ 1990 °K)	As irradiated	1.5	62	11.51	--	--	--
	725 °C/100 hr	↓	59	11.33	1.6	1.1	0.50
	825 °C/100 hr		54	11.30	1.9	1.3	0.55
	900 °C/100 hr		24	10.98	4.8	3.2	0.59
Th-5.4 wt% U (mp $\approx$ 1830 °K)	As irradiated	1.3	58	11.67	--	--	--
	725 °C/100 hr	↓	50	11.58	0.8	0.6	0.54
	825 °C/100 hr		40	11.32	3.1	2.4	0.60
	900 °C/100 hr		32	10.45	11.7	9	0.64

H_T = Homologous temperature - Annealing temperature in °K divided by the solidus temperature in °K

ρ_i = As-irradiated density

ρ_f = As-annealed density

Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).

Fig. 19. Swelling Behavior of Thorium-Uranium Alloys During Post-irradiation Annealing. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

In summary, the Th-20% U alloys of interest in this review may be expected to perform well under the expected operating conditions of $< 650^{\circ}\text{C}$ for a maximum exposure of 40,000 Mwd/metric ton (about 4.3 at. % burnup). The swelling should be isotropic and only slightly more than the minimum expected from solid fission fragments, assuming that the uranium phase will be in either small particles or a slab geometry. These conditions correspond closely to cast specimen ANL-42-1-3 irradiated for Kittel *et al.*,⁷ which swelled 10.7% to an R value of 2.6.

Mechanical Properties

The only reported tensile data on irradiated thorium-uranium alloys are from BNWL.²² The stress-strain curves are given in Fig. 20, and the pertinent facts concerning the samples are in Table 18. The elastic

Fig. 20. Effect of Irradiation on the Stress-Strain Behavior of Thorium-Uranium Alloys. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

Table 18. Irradiation History and Mechanical Properties of Thorium-Uranium Alloys

Specimen Number	wt% U	Flux $\times 10^{-14}$	nvt $\times 10^{-20}$	Days	% Burnup	Average Hardness Rockwell B	Strength, psi	Elongation, %	Yield Offset, psi	Red. in Area, %
13T-132	1.0	--	--	--	0	--	39,400	38.8	31,600	--
T 121	1.0	1.3	4.88	44.2	0.18	--	71,700	9.6	69,200	--
T 122	1.0	1.5	13.31	101	0.6	83	83,000	3.5	79,500	4.9
T 123	1.0	1.0	22.65	252	1.5	92	Broke in the elastic region outside the gage length			
P2T-424	4.0	--	--	--	0	--	41,900	33.1	30,100	--
441	4.0	0.48	0.39	9.2	0.08	--	68,700	15.6	67,500	--
412	4.0	0.28	0.706	28.4	0.15	--	72,200	15.0	68,600	--
41-T 413	4.0	0.31	3.22	120	0.6	74	85,000	5.4	79,000	7.2
E-7-2	5.4	--	--	--	0	--	39,400	35.0	28,700	--
A 91	5.4	0.32	0.25	9.2	0.07	--	63,200	13.8	61,600	--
C12	5.4	0.21	4.48	249	1.3	88	81,500	4.1	75,900	11.6

Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).

modulus of 11×10^6 psi for these alloys was essentially the same as that for unalloyed thorium. At the slow strain rate of 0.005 min^{-1} a measurable amount of ductility remained even after 1.3 at. % burnup. Figure 21 shows the variation of ultimate strength and elongation with burnup. Since these alloys consist of uranium particles in a thorium matrix, it is apparent that the thorium matrix accounts for their behavior.

Bement⁴¹ has collected the changes in tensile properties and hardness of thorium upon irradiation. These results are presented in Tables 19 and 20 and Fig. 22. The yield and tensile strengths increase dramatically with burnup. The elongation decreased with increasing burnup: brittle fracture occurred at less than 4% elongation after burnups of greater than 1 at. %. Only limited data are available on strength and ductility at high temperatures after irradiation. Leggett³⁹ evaluated the tensile properties of thorium at high temperatures after relatively low burnups. These results are in Table 21. The brittle fracture at 815°C was attributed to collection of fission products at the grain boundaries during testing. The absence of data on mechanical properties at elevated temperatures after reasonable exposure is a

Fig. 21. Dependence of Ultimate Tensile Strength and Total Elongation Upon Burnup in Irradiated Thorium-Uranium Alloys. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).

significant gap in the knowledge of the irradiation behavior of thorium alloys.

FABRICATION OF THORIUM-URANIUM ALLOYS

Thorium alloy fuels may be manufactured by powder metallurgy or by conventional techniques such as casting to shape or fabricating shapes from an ingot by any of the common routes for metal processing. Due to the extreme reactivity of the metal, care must be exercised to avoid contamination of the metal by reactions with crucibles, etc. Hazards are involved due to both the reactivity of the metal, especially in the powdered or molten state, and to the toxic and radiological properties of the metal. The hazards associated with production and manufacture of thorium have been covered thoroughly by Albert⁴² and others. Several review articles summarize most of the fabrication experience with the thorium-uranium alloys.^{22,43-45}

Melting and Casting

The high melting point and extreme chemical reactivity of thorium present special problems for the melting and casting of alloys. These

Table 19. Tensile Properties of Unirradiated and Irradiated Thorium

Sample Number	Source	Thermal nvt x 10 ⁻²⁰	Burnup, at. %	Strength (x 10 ⁻³ psi)				Total Elongation, % in 1 in.
				Lower Yield	Upper Yield	0.2%	Ultimate	
A. Unirradiated								
CT-1	BMI ^(c)	--	--	--	--	16.7	24.4	41
CT-2	BMI	--	--	--	--	16.4	24.6	50
CT-6	BMI	--	--	--	--	17.5	26.5	57
CT-9	HAPO ^(d)	--	--	--	--	21.0	31.7	55
CT-12	HAPO	--	--	--	--	22.3	29.9	(a)
B. HAPO Irradiations								
1. As-Irradiated								
EZ	BMI	1.5	0.0014	37.8	34.4	--	37.8	44
GZ	BMI	1.6	0.0017	38.2	35.9	--	38.2	34
DZ	HAPO	1.6	0.0017	40.3	36.5	--	40.3	(a)
FZ	HAPO	1.8	0.0023	39.8	36.5	--	39.8	32
EW	BMI	6.9	0.054	--	--	42.3	43.7	8.3
GW	BMI	6.6	0.050	44.2	44.0	--	44.6	(a)
DW	HAPO	6.4	0.046	44.4	43.2	--	44.4	17
FW	HAPO	6.6	0.050	53.0	49.5	--	53.0	(a)
EX	BMI	8.1	0.077	--	--	44.2	44.6	24
GX	BMI	7.1	0.058	--	--	44.5	44.9	29
DX	HAPO	7.7	0.070	51.1	50.5	--	51.1	(a)
FX	HAPO	8.4	0.085	49.5	48.6	--	49.5	(a)
2. Annealed at 750 °C for 1 hr								
IZ	BMI	1.9	0.0026	--	--	19.6	27.8	55
HZ	HAPO	2.1	0.0033	--	--	18.1	28.4	38
AY	BMI	6.9	0.054	--	--	25.7	34.5	39
9Y	HAPO	6.8	0.053	--	--	29.7	38.7	(a)
IX	BMI	8.6	0.088	--	--	27.2	36.1	(a)
HX	HAPO	8.4	0.085	--	--	33.0	42.5	36
3. Annealed at 750 °C for 100 hr								
KZ	BMI	2.2	0.0037	--	--	15.2	24.0	--
KZ	HAPO	2.3	0.0042	16.8	16.0	--	25.0	--
KW	BMI	6.5	0.049	--	--	19.6	28.4	43
JW	HAPO	7.0	0.056	--	--	19.6	29.3	40
KX	BMI	8.4	0.085	--	--	23.7	33.5	36
JX	HAPO	8.9	0.095	--	--	25.7	34.4	46
C. MTR Irradiations								
28	BMI	9.9	0.12	50.9	49.3	--	50.9	18
27	HAPO	12	0.18	56.8	56.2	--	56.8	13
30	HAPO	31	0.92	--	--	69.1	74.9	2.4
26	HAPO	35	1.2	--	--	--	48.9 ^(b)	<1 ^(a)
25	BMI	45	1.6	--	--	--	55.9 ^(b)	<1 ^(a)
29	BMI	50	1.9	--	--	69.1	75.6	3.2

(a) Sample broke outside of 1-in. gage length

(b) Premature failure in range of elastic strain

(c) BMI Material (~400 ppm °C)

(d) HAPO Material (~700 ppm °C)

Reprinted from R. D. Leggett and R. S. Kemper (eds.),
Status of Thorium Fuel Technology, BNWL-861 (August 1968).

Table 20. Changes in Hardness with Irradiation

Source	Thermal Flux, nvt x 10 ⁻²⁰	Burnup, at. %	Average Hardness, Rockwell A		Range	Variance, s ²
BMI	0	0	(7) ^(a)	15.7	14.2-16.6	0.45
BMI	50	1.9	(11)	44.5	42.0-45.5	0.91
HAPO	0	0	(7)	27.2	25.8-28.9	1.23
HAPO	12	0.18	(12)	48.4	42.5-51.0	5.92
HAPO	31	0.92	(8)	52.7	51.0-54.3	1.61
HAPO	35	1.2	(6)	53.0	50.0-58.6	16.0

(a) Number of readings taken

Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).

problems have generally been handled by special melting techniques such as induction melting or arc melting in either a vacuum or inert atmosphere. Homogeneity of thorium-uranium castings is also of concern due to the difference in melting points, difference in densities, and very low mutual solid solubilities of the two metals. Another item of concern in casting of intricate shapes is hot tearing due to the low melting point of the uranium-rich eutectic in the system for alloys that contain more than about 5% U. A wide variety of thorium-uranium alloys have been cast into button and ingot shapes. All of the irradiation data have been obtained from cast specimens, some of which were hot and/or cold worked before irradiation.

Induction Melting

Several furnace designs used to melt thorium by induction in vacuum or an inert atmosphere are shown in Figs. 23, 24, and 25. For routine operation, furnaces of the tilt-pour type should offer more reliable service and minimize problems of reaction with the crucible. The power for these furnaces is generally provided by 50 to 200 Kw, 700 to 3000 Hz motor-generator sets. The lines of magnetic force from the induction coils induce stirring in the melt. Stirring of the melt is substantial unless graphite or another electrical conductor is used for crucibles or heaters around the crucibles.²²

Fig. 22. Change in Tensile Properties and Hardness with Irradiation for Unalloyed Thorium. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), *Status of Thorium Fuel Technology*, BNWL-861 (August 1968).]

Table 21. Room Temperature and Elevated Temperature
Tensile Properties of Irradiated Thorium

Sample Number	Testing Temperature, °C	Thermal nvt x 10 ⁻²⁰	Burnup, at.%. .	Strength x 10 ⁻³ , psi		Total Elongation, % in 1 in.
				0.2% Yield	Ultimate	
<u>Medium Carbon (~400 ppm)</u>						
<u>BMI Material</u>						
CT-6	RT	0	--	17.5	26.5	57
CT-8	815	0	--	~7	~17	~43
EX	RT	8.1	0.077	44.2	44.6	24
GX	RT	7.1	0.058	44.5	44.9	29
AX	590	7.3	0.060	~24	~27	~22
CX	815	7.6	0.065	~20	~21	~9
<u>High Carbon (~700 ppm)</u>						
<u>HAPO Material</u>						
CT-10	590	0	--	~9	~20	~92
CT-4	815	0	--	~8	~16	~26
DX	RT	7.7	0.070	50.5 ^(b)	51.1	(a)
FX	RT	8.4	0.085	48.6 ^(b)	49.5	(a)
9X	590	7.3	0.060	~22	~23	~24
BX	815	7.5	0.065	~19	~19	~16

(a) Sample broke outside of 1 in. gage length.

(b) Upper yield strength

Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).

Fig. 23. Bottom-Pour Induction Furnace and Mold Assembly Wholly Contained in a Water-Cooled Melting Chamber. [Reprinted from R. D. Legett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

Fig. 24. Internally Water-Cooled Tube-Type Bottom-Pour Furnace with Induction Coil Outside of Nonconducting Tube (Tube is Typically Micarta or Quartz). [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

Fig. 25. Tilt-Pour Induction Furnace for Semicontinuous Vacuum Melting. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

Arc Melting

The consumable-electrode process for arc melting was developed for zirconium for the Bureau of Mines at Albany, Oregon. In the mid-1950s this process was selected by National Lead Company of Ohio for the scale-up of the Ames Laboratory process to a semiworks production process at Fernald, Ohio.²² This type of melting furnace is pictured in Fig. 26. The atmosphere during melting is inert gas or vacuum. The alloy to be melted is in the form of a rod that is the negative electrode of a direct current power source. The positive side of the power source is the water-cooled copper crucible. An arc is struck with the bottom of the crucible and melts the electrode. The electrode is advanced to maintain a constant gap between the electrode tip and the molten pool.

Fig. 26. Consumable Arc-Melting Furnace. [Reprinted from R. D. Leggett and R. S. Kemper (eds.), Status of Thorium Fuel Technology, BNWL-861 (August 1968).]

Only a small pool of metal adjacent to the tip of the electrode is molten. This minimizes interactions with the crucible and loss of alloying elements by vaporization. The metal is usually melted at least twice to ensure complete solution of alloying elements. The large ingot is sometimes extruded to form the electrode for the subsequent melting.

Nonconsumable-electrode arc melting furnaces of the type shown in Fig. 27 have been used extensively for small quantities of alloys for research. The alloy constituents are placed on the water-cooled copper hearth and melted several times. The atmosphere is usually inert gas. Shapes may be cast in this furnace by replacing the copper hearth with a similar one with a hole in the center. A water-cooled copper mold (generally) is placed underneath the hole. The button which solidified on the previous hearth is then placed in the drop-casting hearth and heated by the arc. The button is slowly melted and stirred with the arc until only a thin skin of solid metal is adjacent to the hearth. When the

Fig. 27. Laboratory-Size Inert-Gas Nonconsumable Arc-Melting Furnace.

alloy is ready for casting, the power is increased, and the arc is concentrated over the hole so that the skin melts and allows the molten alloy to drop into the mold.

Another type of nonconsumable-electrode arc melting furnace used for larger melts is the fixed-electrode furnace shown in Fig. 28. The fixed-electrode furnace is similar in principle to the consumable-electrode furnace in that only a portion of the charge is molten at one time. Preparation of material to be melted is simplified since chips or chunks of the alloy are fed into the molten pool from a hopper.

Fig. 28. Fixed-Electrode Furnace. 1. Vacuum-tight hopper shell; 2. feed hopper; 3. vacuum port; 4. crucible; 5. tungsten electrode; 6. sight glass; 7. vacuum-tight gland; and 8. power connection. (Reprinted from R. A. Beall *et al.*, *Cold-Mold Arc Melting and Casting*, U.S. Bur. Mines Bull. 646, U.S. Bureau of Mines, Washington, D.C., 1968.)

Another type of arc furnace that is capable of producing fairly large ingots or shape castings is the "skull furnace" developed by the Bureau of Mines at Albany for titanium castings.⁴⁶ This is a furnace with a water-cooled crucible that operates with a "skull" of frozen metal against its surface, but keeps the main mass of the metal molten so that it can be poured from the crucible into a mold. The heat input is from an arc from either a permanent or consumable electrode.

Figure 29 is a drawing of a typical consumable-electrode skull furnace. A permanent-electrode furnace is similar except that chunks of the alloy would be charged into the skull for melting or fed into the skull by a hopper arrangement similar to that shown in Fig. 28. The permanent electrode has the advantage of not requiring consolidation and shaping of the charge into a consumable electrode. However, consumable electrodes have the advantages of higher power input for faster melting and more superheat, more refining and gas removal as the metal is melted and sprayed into the crucible, no contamination of the melt from erosion of a permanent electrode, and the capability of operating in a vacuum. Permanent electrodes generally must be operated in an atmosphere of inert gas to promote arc stability. A dual installation with consumable electrodes cast in a permanent-electrode furnace for reclamation of scrap and addition of alloying elements would offer an efficient production setup. Shape castings would then be made with the more desirable operating conditions of a consumable-electrode skull furnace.

Crucible and Mold Materials

The chemical reactivity of thorium metal creates contamination problems during prolonged contact of the liquid alloy with crucible and mold materials. During arc melting, this problem is usually alleviated by the use of water-cooled copper crucibles and molds to maintain a film of solid metal next to the crucible or mold. Materials that have been used for crucibles in induction melting and mold materials include stabilized zirconia, beryllia, graphite, and graphite washed with BeO or MgZrO_3 (ref. 22). Five-hundred-pound tilt-pour vacuum induction

- A Power connection
- B O-ring
- C Consumable electrode
- D Viewing ports
- E Tilting lever
- F Tilting crucible
- G Mold

Fig. 29. Schematic Outline of a Typical Skull Furnace. (Reprinted from R. A. Beall et al., Cold-Mold Arc Melting and Casting, U.S. Bur. Mines Bull. 646, U.S. Bureau of Mines, Washington, D.C., 1968.)

furnaces with stabilized zirconia crucibles and graphite molds were used by National Lead Company of Ohio; zirconia pickup averaged 400 ppm. Bottom pouring crucibles of beryllia were used in early work at Ames Laboratory; beryllium pickup was about 150 ppm. Work at Nuclear Metals, Inc., indicated that BeO was superior to zirconia as a crucible material and to MgZrO_3 as a wash material on graphite.⁴⁷ They indicate that washed graphite crucibles may be used satisfactorily if superheat is held to a minimum and if 200 to 400 ppm of carbon pickup is tolerable.

Thorium dioxide crucibles have not been used in recent work for melting thorium. In very early work, Marden and Rentschler⁴⁸ reported thoria crucibles to be the most satisfactory refractory for melting thorium. No contamination was observed. Thorium, even after melting, did not stick to the thoria crucibles as prepared for their work. Thoria crucibles have been used for melting of other high-temperature reactive metals. Since thoria has the highest melting point and is the most stable to reduction of all the refractory-metal oxides, it resists attack and is compatible with many reactive metals.⁴⁹ One limitation to thoria is that it is subject to thermal shock. Fabrication, properties, and use of these thoria ceramics have recently been reviewed by Hepworth and Rutherford⁵⁰ and by Ryshkewitch.⁵¹

Another possibility for shape castings is expendable graphite molds made from powder mixtures developed for casting reactive metals.⁵² These would probably need to be coated for use with thorium alloys.

Homogeneity of Ingots

Gravity segregation of alloy constituents can be a problem when castings are cooled slowly. For example, 100-lb melts poured into pre-heated, washed graphite molds have yielded billets ranging from 6.7% U at the top to 12.7% U at the bottom. Identical 10% U melts poured into cold, heavy-walled graphite molds yielded billets with variations of only $\pm 0.2\%$ U (ref. 22). Work at Nuclear Metals, Inc. indicated that induction melting and pouring into 1 3/8-in.-diam molds yielded ingots varying from ± 0.1 to ± 0.3 wt % in alloys containing 5 to 15% U (ref. 47). They reported homogeneity in arc castings of the compositions to be inferior

to that in the induction castings. Homogeneity was not improved by a secondary arc melt. Kemper et al.,³⁴ produced Th-2.5% U-1% Zr ingots of up to 200 lb in a consumable-electrode arc furnace (Fig. 26) with excellent uranium homogeneity. The attainment of homogeneity in thorium-uranium castings depends on obtaining a reasonably fast cooling rate. A 3/4-in.-diam by 2-in.-long Th-19% U alloy drop cast into a water-cooled copper mold from a permanent-electrode arc furnace (Fig. 27) and hot rolled into 0.030-in. sheet yielded uranium contents along the length of 19.0, 18.4, 18.4, 18.8, 19.0, 18.8, and 19.1% U.

Hot Tearing

Hot tearing is of concern in casting shapes with inside corners when a low melting eutectic is present as is the case with the Th-20% U alloy. We drop cast into a water-cooled copper mold an H-shaped section of this alloy that weighed 366 g and had a 1/4-in.-web thickness to determine if hot tearing would be a problem in this "worst case" configuration. The results are shown in Figs. 30 and 31. Although there are some

Fig. 30. Cross Section of H-Shaped Casting. As polished. 5x.

Fig. 31. Inside Corner of H-Shaped Casting. As polished. 100x.

small cracks in the inside corners, the results are very encouraging. The small amount of cracking observed could probably be eliminated by proper mold design.

Powder Metallurgy

Although we uncovered no reports of fabrication of thorium-uranium alloy by powder metallurgy, we found that the powder metallurgy of thorium has been investigated extensively.^{22,43,53} The processing techniques for thorium powder will be presented briefly below. Both uranium and thorium powders may be produced from massive metal by the hydride process or reduced directly into powder form.

The hydride process for thorium consists of reacting chunks of cleaned metal with hydrogen at 1 atm pressure at 600 to 650°C to form ThH_2 (ref. 53). During this treatment, the charge swells but does not break up. The temperature is lowered to 250°C to form the higher hydride, ThH_4 . The charge exfoliates and breaks up during this operation. The temperature is raised to 500°C, still under 1 atm H_2 , to decompose the charge to ThH_2 . Finally, the ThH_2 is decomposed by heating to 700°C

and reducing the pressure to less than 500 $\mu\text{m Hg}$. The resulting powder can be broken down to pass a 30-mesh screen. A finer powder can be obtained by repeating the hydride process or by ball milling the hydride before it is decomposed.

The hydride process for uranium consists of reacting small pieces of uranium metal with hydrogen at temperatures as low as 100 to 150°C; the maximum reaction rate occurs at about 225°C (ref. 54). The reaction is exothermic but self-controlling, since temperatures above 225°C reduce the reaction rate. The UH_3 formed is a fine powder of less than 400-mesh screen size. The hydride powder is then decomposed above 225°C in vacuum. At about 430°C, the hydride can be decomposed even at atmospheric pressure. The high surface activity of the finely powdered uranium causes sintering of the fine particles when decomposition occurs at higher temperatures. The agglomerates or sinter cakes formed in this way are brittle and may be mechanically broken into particles of the desired size. Any uranium particle obtained in this way is actually an agglomerate of much finer particles.

Cold Compacting and Sintering^{22,43,53}

Thorium powders are ductile and compact readily during pressing. The coarse fractions of powder exhibit better cold compatibility than the finer fractions. Densities up to about 90% of theoretical are obtained using pressures up to 75 tsi. Sintering at temperatures of 1400°C gives densities slightly lower than can be obtained by hot pressing. The finer powders yield the highest densities after sintering. Sintering at higher temperatures has very little advantage for most powders. Densities up to 98% of theoretical are obtained with -60 mesh powders. Samples sintered in vacuum increase more in oxide content than comparable samples hot pressed in vacuum. The sintered compacts may be further densified by low-temperature rolling, coining, or extrusion.

Hot Compacting^{22,43,53}

Thorium powders are generally hot pressed at 625 to 650°C for 10 to 15 min at 12 to 15 tsi. The finer powders reach higher density than do

the coarse powders but also increase more in oxide content during the hot-pressing operation. Chips or lathe turnings typically pick up an additional 300 to 500 ppm O, whereas the finer hydride or electrolytic powders gain an additional 1000 to 2000 ppm O under hot-pressing conditions (25 to 30 μ m Hg pressure). Any of the powders may be hot pressed to 98 to 100% of their theoretical density.

Forming

The face-centered cubic structure of thorium suggests good ductility for forming operations, and this has been demonstrated for a wide variety of operations. Since there are no phase transformations up to 1400°C, the range of working temperatures is quite large. Most of the hot-working operations are performed between 600 and 950°C. Uranium, however, is generally hot worked in the alpha range at 600 to 640°C. Not enough data are available to fully assess the effects of higher working temperatures on the small, dispersed particles of uranium in the thorium alloys.

Extrusion^{20,43}

Thorium has been extruded at temperatures from 593 to 954°C. The metal may be canned in copper or extruded bare after it has been heated in a salt bath.

The copper canning protects the thorium from oxidation during heating and extrusion, prevents the spread of contamination, provides good lubrication, and prolongs die life. The temperatures are limited to about 790°C to prevent alloying of the thorium and copper. The copper is removed after extrusion by pickling in nitric acid. Cold finishing operations make the extrusions usable without need for machining.

The expense of canning and decanning may be eliminated by the use of salt baths for heating the bare thorium billets. Proper facilities are required since radioactive scale may be released. Mixtures of barium, potassium, and sodium chloride salts with proper melting points have been used to heat thorium to 650 to 950°C. Very little attack of the metal occurs on heating. The salt acts as the lubricant during

extrusion. Die life is relatively short and depends on the salt temperature and reduction ratio used. A die life of 12 to 15 extrusions has been reported for commercial extrusions of 5-in.-diam billets. The extruded surface is somewhat striated and rough. A conical die is preferred over a flat die.

Thorium-enriched uranium fuel elements have been coextruded with Zircaloy claddings by SRL in cooperation with Nuclear Metals, Inc.,³³ and by BNWL.³⁴ The uranium contents were 1.4 and 2.5%, respectively, and results were excellent in both cases.

Rolling^{22,55}

Thorium ingots are easily rolled at 740 to 850°C with reductions of 25 to 30% per pass. The metal may be clad in copper or rolled from a salt bath as for extrusion. The metal may also be cold rolled as-cast up to about 25% reduction before edge cracking begins. After edge trimming, the metal has been successfully cold reduced as much as 98% with no intermediate anneals. Hot working improves the cold-rolling properties of the metal. Reductions of 80 to 90% can be made on material that has been hot rolled with no serious edge cracking. Foil 0.001 in. thick has been cold rolled from 0.020 in. hot-rolled sheet with no intermediate anneals.

Although quantitative results are not available, the presence of 20% U distributed uniformly in the thorium appears to have very little effect on the rolling properties. The principal effect is probably the strengthening due to the small amount of uranium in solution in the thorium.

Swaging and Drawing^{22,43}

Thorium can be swaged easily with conventional equipment. Good quality thorium can be swaged to any amount of reduction without intermediate anneals. Very good surface finishes are obtained with standard swaging procedures.

Thorium rods and wires have been produced successfully by drawing. Copper cladding or plating and solid-film lubricants have been used.

The low coefficient of strain hardening limits the total amount of reduction per pass. Wire as small as 0.003 in. in diameter has been made from arc-melted iodide thorium.

Extruded thorium tubes can be drawn by standard techniques of tube drawing. Only mandrel drawing is used because of the difficulty of lubricating the inside surface of tubing and because of the low strength of the metal. A total of 80 to 90% reduction in area can be obtained without annealing. The reduction per pass should be about 25% at the start and lowered to less than 10% on further passes.

Again, the principal effect of the uranium is probably the increased strength due to the uranium in solid solution. The particles of the uranium-rich phase probably have very little influence when they are well-distributed.

FABRICATION OF UNCLAD-METAL BREEDER REACTOR FUEL ELEMENT

This section summarizes the methods that might be used economically to fabricate fuel elements for the UMBR. The reference design of the fuel element (Fig. 1) and some alternate designs that would offer fabrication advantages are considered. Generalized flowsheets are presented for each method, and the obvious advantages and disadvantages are discussed. Detailed cost analyses were not made; however, relative costs of the processes may sometimes be estimated from the flowsheets. The status of technology applicable to each process for thorium-uranium alloy is also of importance. Every technique considered feasible is presented, although several are obviously impractical.

Reference Hexagonal Design

Several techniques may possibly be used to fabricate the reference fuel element, shown in Fig. 1, which consists of hexagonal coolant channels in a hexagonal sheath. The feasibility of these methods has not been demonstrated for the thorium-uranium alloy.

Cold-Binder Extrusion of Alloyed Powders

This method, briefly investigated at the Y-12 Plant of Union Carbide Nuclear Division,⁵⁶ entails extrusion of alloyed powders and a binder into individual hexagonal tubes. Following partial sintering and removal of the binder, the tubes are bundled into the overall hexagonal shape, then bonded together during final sintering. Figure 32 is a simplified flowsheet for this process. It is of utmost importance to obtain alloyed powder with the desired characteristics. The powder should contain a uniformly distributed and reproducible uranium content. The powder should also have a reproducible distribution of particle sizes and be of the required purity. The proposed method for making the powders, outlined in Fig. 33, consists of (1) blending uranium and thorium powders (or their hydrides) and sintering to form a thorium-uranium alloy and (2) hydriding and dehydriding the alloy to form fine powder.

A binder must be found that does not react with the powders and is easily removed after extrusion. Obtaining high density and good bonds between the tubes may also be a problem. One advantage is that the tubes

Fig. 32. Flowsheet for Cold-Binder Extrusion of Alloy Powders.

Fig. 33. Flowsheet for Production of Thorium-Uranium Alloy Powders.

may be long enough for more than one fuel element. Dimensional control may be a problem unless a mechanical drawing or similar operation is performed after initial sintering of the tubes.

Cold Pressing of Alloyed Powders

Short cross sections of the element would be cold pressed from alloyed powders as outlined in Fig. 34. The pieces would probably have to be about 1/2 to 1 in. long to obtain reasonable cold-pressed density. A lubricant would undoubtedly be required during pressing.

Fig. 34. Flowsheet for Production of Fuel Element by Cold Pressing and Sintering of Cross Sections.

A lubricant would have to be found that would not react with the powders and would be easily removable during sintering. A warm coining operation might also be required after sintering. Assuming that 1-in.-long sections of suitable density and purity could be produced, the fuel element would require that 18 of these be stacked together. The flow of sodium through coolant channels formed in this manner might present an erosion and turbulence concern. If this problem exists, it might be solved by securely bonding the sections together and smoothing the channels by broaching. The bonding might be accomplished by pressure bonding, brazing, welding at the edges, or securely clamping the sections together with an external sheath.

Casting

Direct casting of the finished fuel element would probably be the most economical fabrication method. The feasibility of casting to shape depends on (1) finding suitable mold materials and (2) having sufficient superheat for fluidity to fill the mold, yet cooling rapidly enough to avoid gravity segregation in the casting. Figure 35 is a flowsheet for producing castings. A consumable electrode skull arc-melting furnace (Fig. 29) operating in vacuum would be used for pouring the final castings. The electrodes could be cast in a similar permanent-electrode furnace. Ideally, full-sized elements could be cast in one piece. If it proved necessary to cast shorter cross sections, these could be stacked to form the full-length element, subject to the comments above.

Fig. 35. Flowsheet for Arc Melting and Casting Cross Sections.

Electrochemical Machining

Electrochemical machining (ECM) is the controlled removal of metal by anodic dissolution in an electrolytic cell. The workpiece is the anode, and the tool is the cathode. The current is carried by an electrolyte pumped through the tool and passing between the tool and workpiece. The application of this technique to thorium-uranium alloys

would depend on selection of an electrolyte compatible with the alloy for the time involved. This process, outlined in Fig. 36, would machine a full-sized fuel element from a solid round or hexagonal billet. Fabrication of such a billet will be discussed below. A machine capable of removing about 10 in.² would require 10,000 amp and operate at 10 to 12 v. Initial costs would be about \$120,000 for the machine and tooling. The 18-in. length would require 2 1/2 hr of fully automatic operation and would consume about 250 kwhr electricity. The obvious disadvantage of a process such as this would be the necessity for reclaiming about 65% of the alloy from the electrolyte for reprocessing.

Fig. 36. Flowsheet for Production of Fuel Element by Electrochemical Machining (ECM).

Extrusion with Sacrificial Metal

This technique has been developed at BNWL for production of honeycomb fuel elements and similar shapes. Figure 37 is a flowsheet for production of a honeycomb fuel element with internal cladding. For the UMBR element, the starting billet could be produced by casting or powder metallurgy with copper sacrificial metal. Both external and internal shapes would be hexagonal. One of the potential problem areas would be the removal of the sacrificial copper from the interior of the 18-in.-long sections of the fuel element. Possible solutions include drilling to expose the copper to the leaching solution along the entire length or switching to a sacrificial metal that could be removed by another method.

Fig. 37. Flowsheet for Sacrificial-Metal Extrusion to Form Fuel Element. (Courtesy of Battelle Memorial Institute Pacific Northwest Laboratory.)

Alternate Designs Inside Hexagonal Sheath

This section includes fabrication routes for some fuel element designs that offer fabrication advantages over the reference hexagonal design. While these elements may compromise some of the heat flow or nuclear advantages of the hexagonal coolant channels, the economic advantages may be sufficient to justify their consideration. The fabrication of these elements involves the proven methods of hot rolling and forming of billets into plates of the desired thickness and shape. The plates may then be assembled into a fuel element by a variety of techniques.

The billets may be fabricated by arc melting as in Fig. 38 or, if desirable, by powder metallurgy as in Fig. 39. Arc melting in a skull-type furnace (Fig. 29) is probably the preferable method for casting

Fig. 38. Flowsheet for Production of Alloy Ingots by Arc Melting.

Fig. 39. Flowsheet for Production of Ingots from Alloy Powder.

billets. Again, the two-step process of nonconsumable- and then consumable-electrode melting shows promise for high-quality ingots. The billets may be cast into the shape desired for hot rolling or may be cast round and formed (for example, by extrusion) into flat billets suitable for rolling. For round billets, a standard cold-mold consumable-electrode furnace (Fig. 26) could be used. If production of alloy powder of suitable quality proves feasible, billets could be hot pressed either round for further forming or rectangular for direct rolling. The density of the material as pressed would probably be sufficient for rolling, but greater density could be obtained by extrusion if necessary.

Figure 40 is the flowsheet for production of plates from these billets and assembly into fuel elements. The hot rolling of thorium alloys is a well-established process. The plates may be joined in a variety of ways to form a fuel element. The plates may be mechanically fastened, welded, or brazed to the outer hexagonal sheath. The plates might be joined independently of the sheath by welding or brazing the assembly. The joining technology of thorium-uranium alloys is not developed; however, it appears likely that joints of modest integrity are feasible. Cracking during welding of Th-2.5% U electrodes was reported by BNWL,³⁴ but this problem was solved by the addition of 1% Zr.

Several of the fuel element designs that fit into the hexagonal channel for the reference design are presented below. These three are

Fig. 40. Flowsheet for Production of Fuel Element from Plates.

examples that illustrate the assembly procedure. A large number of variations in plate design and assembly are possible. The simplest plate to fabricate is the flat plate (Fig. 41) fitted into a rigid hexagonal sheath. The plates may be brazed into the slots in the sheath or welded into place by passing electron-beam welds around the periphery of the sheath in several places along the 18 in. length. Designs that include a forming step for the plates are also shown in Fig. 41. In these cases, the plates may be joined at the nodes by brazing or welding to form self-supporting structures, or they can be held together mechanically by the sheath or some clamping device. Grooves for insertion of a braze wire or a tongue-and-groove type of joint for mechanical holding could be formed into the plates during the final forming operation. As shown in Fig. 42, the plate thickness probably could be decreased where the nodes join to equalize the distance for heat transfer. An alternate method of assembly, shown in Fig. 43, consists of electron-beam welding the plates together as each plate is placed on the assembly. Each of these assembly methods seems to be practical for remote operation.

Fig. 41. Sketch Illustrating Several Plate Designs in a Hexagonal Sheath.

Fig. 42. Examples of Methods of Joining Plates at the Nodes.

Fig. 43. Joining Plates into Hexagonal Prism Array by Electron-Beam Welding.

These are only examples of the many plate designs that would fit the hexagonal fuel element sheath. Other variations could also be considered with a different sheath configuration. Also, if a pin-type element were feasible, this would be a very simple shape to fabricate by extrusion and/or swaging of a billet.

CONCLUSIONS AND RECOMMENDATIONS

Several techniques appear to be available for fabrication of the UMBR fuel element. The best technique will have to be chosen on the basis of both economics and properties of the element. Irradiation tests indicate that the Th-20% U alloy has the irradiation stability required to reach the design life of 33,000 Mwd/metric ton at $\leq 650^{\circ}\text{C}$; however, 650°C appears to be on the threshold where increased swelling due to fission gas could occur.

The Th-20% U alloy will consist of the thorium-rich phase and uranium-rich eutectic no matter how it is produced. However, the distribution of the phases may be different for various production methods. The distribution of the phases will be important in determining the irradiation, mechanical, and corrosion performances of the alloy. Alloys with a finely distributed uranium phase have been shown to perform better under irradiation than alloys with a more coarse distribution of uranium. Thus, any test specimens should be produced by techniques that yield a distribution of the phases similar to that which may be in the actual fuel elements.

Early in the development of the fabrication procedures for the fuel element, the basic feasibility of casting to shape, forming from an ingot (cast or powder metallurgy), and powder metallurgy production should be determined. Based on these results a prime method should be selected and the major effort applied there.

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