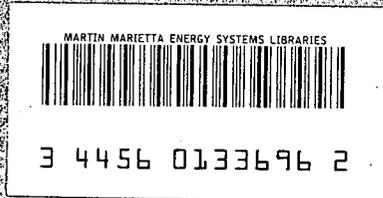


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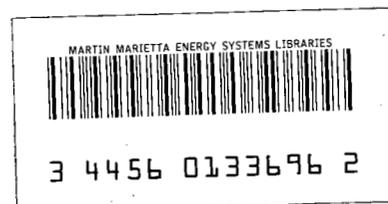
Metals and Ceramics Division

PROPERTIES OF THORIUM, ITS ALLOYS, AND ITS COMPOUNDS*

Sigfred Peterson, R. E. Adams, and D. A. Douglas, Jr.

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ABSTRACT

Physical and mechanical data pertinent to nuclear application have been reviewed for thorium, its alloys, and its ceramic compounds. Thorium-base fuels are superior to uranium at elevated temperatures. Although thorium oxide is well characterized, the effect of incorporated uranium oxide on its properties is very poorly known. Data are incomplete on thorium carbides, but these compounds are being actively investigated along with their uranium substitution products.

INTRODUCTION

Reactor fuels based on thorium or its compounds are important for fuel economy in advanced converters and breeders. The development of such fuels requires considerable physical, mechanical, and other data on potential materials. Our purpose has been to survey the literature on these materials, report selected data, and indicate the areas where further data are needed.

THORIUM AND ITS ALLOYS

Although thorium metal has potential application as a nuclear fuel base, up to now it has been used only as the Core II loading for the Sodium Reactor Experiment. It has been proposed for some power reactors, and considerable development on thorium metallurgy and properties has been carried out. The results of this research have been summarized in several reviews;¹⁻⁴ the most comprehensive have covered available data up to the period of about 1958 to 1960. We have examined some of the more recent data on thorium and its alloys as an aid in planning additional research directed toward establishing the feasibility of thorium-fueled reactors.

Physical Properties of Thorium

In many respects the physical and mechanical properties of thorium compare favorably with those of uranium. The physical properties are compared in Table 1. The isotropic face-centered cubic crystal structure of thorium is a significant advantage, since the orthorhombic crystals of uranium expand unequally in different directions. This anisotropy is responsible for growth effects and internal stresses that lead to accelerated swelling when uranium is irradiated in the temperature range of 400 to 550°C. The phase transformation in uranium effectively prohibits its operation as a fuel above about 660°C, since fission gases are released from the lattice during atomic rearrangements that

Table 1. Physical Properties of Thorium and Uranium.

	Pure Thorium	Uranium (β transformed)
Crystal structure	face-centered cubic	orthorhombic
Lowest transformation temperature	1400°C	661°C
Melting point	1750°C	1130°C
Thermal conductivity coefficient, $w \text{ cm}^{-1} \text{ } ^\circ\text{C}^{-1}$		
at 100°C	0.38	0.28
400°C	0.42	0.35
650°C	0.45	0.42
Thermal expansion coefficient, $10^{-6}/^\circ\text{C}$ (average)		
25-200°C	11.0	15.5*
25-650°C		19
25-1000°C	12.5	
Density, g/cm^3	11.7	19.0
Elastic constants (25°C)		
Modulus of elasticity, psi	10.4×10^6	29.3×10^6
Shear modulus, psi	4.1×10^6	12.0×10^6
Poisson's ratio	0.27	0.22

*Average values for beta-transformed uranium values for the three directions are [100], 27.4×10^{-6} ; [010], 0; [001], 23.2 for the range 0 to 300°C.

occur during transformation. The phase transformation of thorium is at about 1400°C, and it is decreased only slightly by many alloying elements, as shown by Bannister.⁵

The thermal conductivity of thorium is about 30% greater than that of uranium at 100°C and about 8% greater at 650°C. The density of thorium is significantly less than that of uranium metal.

A selection from Ames Laboratory's⁶ values of the heat capacity of iodide thorium follows:

Temperature, °C	25	100	200	400	600	800	1000
Heat capacity, $\text{cal mole}^{-1} \text{ } ^\circ\text{C}^{-1}$	6.56	6.75	6.99	7.49	7.99	8.62	9.54

Darnell and McCollum⁷ have studied the vapor pressure of thorium in equilibrium with ThO₂ and found by extrapolation the following relation for the vapor pressure of pure metal

$$\log P(\text{atm}) = -27,960/T + 5.575.$$

The predominate vapor species above liquid solutions of ThO₂ in thorium was ThO, and its vapor pressure was described as

$$\log P(\text{atm}) = -(22,200 \pm 700)/T + (4.70 \pm 0.31).$$

The solid solubility of ThO₂ in thorium was 3.5% at 2090°K, and 8.2% at 2450°K.

Thorium has elastic and mechanical properties at room temperature somewhat inferior to those of uranium. The mechanical properties of thorium metal are dependent on impurity and alloy content and on the textures and cold work developed during fabrication treatments. The purest thorium, made by decomposition of thorium iodide, has when annealed tensile strengths of 16,000 to 20,000 psi, yield strengths from 5000 to 10,000 psi, and considerable ductility. Commercial thorium, produced by bomb-reduction of thorium fluoride, contains carbon and other interstitial impurities which strengthen thorium, as shown in Fig. 1. In the cast or wrought and annealed condition this metal has good ductility and tensile and yield strengths varying upward from about 25,000 and 18,000 psi, respectively. Thorium work hardens rapidly, and tensile strengths are increased 50% and yield strengths doubled by cold reductions of 50% or less. Ductility is reduced by cold work, but not excessively so. Representative data¹ are shown in Table 2.

Thorium metal is readily fabricated by casting, powder metallurgy, extrusion, rolling, and other methods. Cold worked material starts to recrystallize and anneal at slightly above 500°C, although temperatures up to 700°C may be required to ensure complete annealing for slightly worked material.

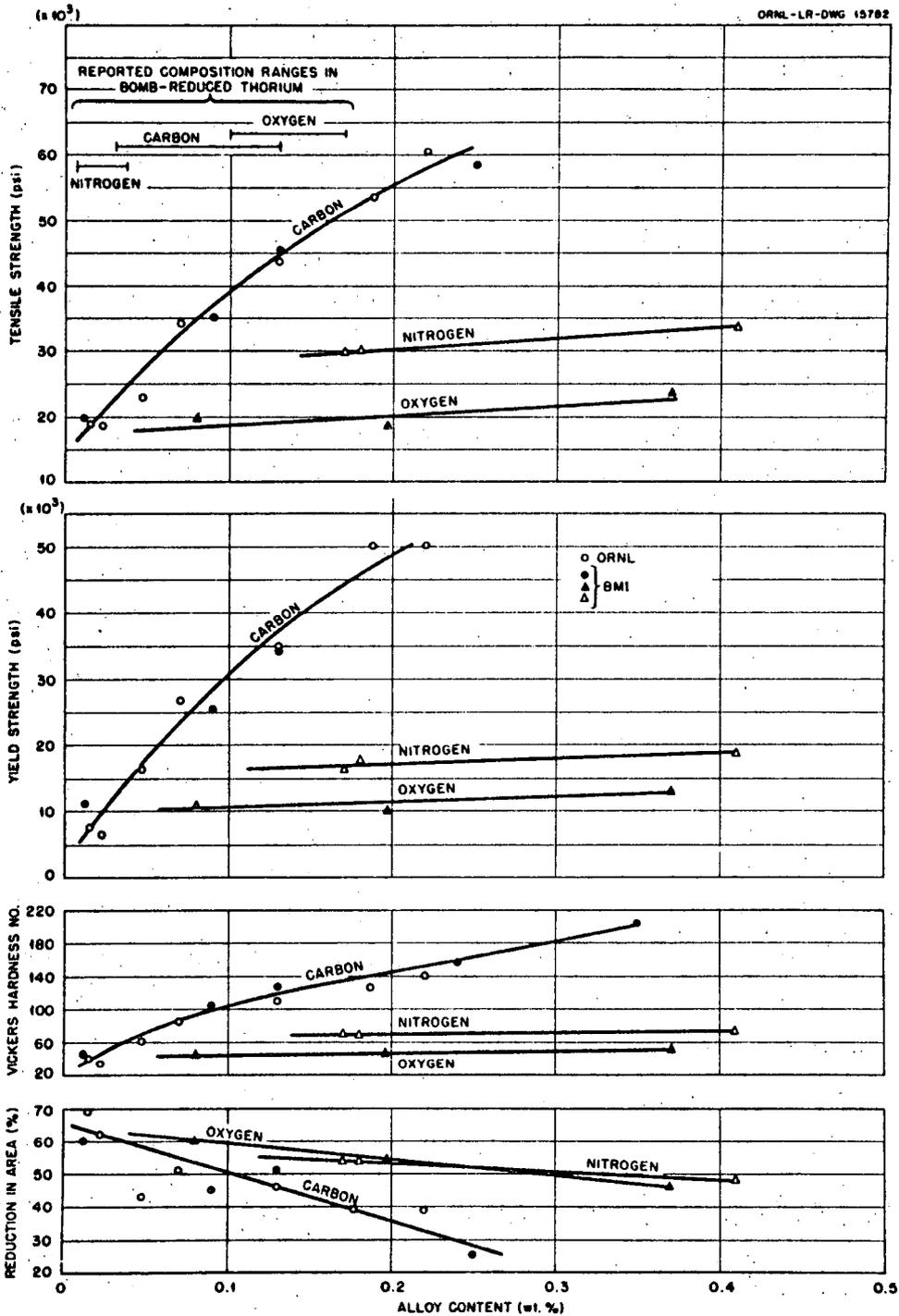


Fig. 1 Effect of Some Interstitial Solutes Normally Found in Bomb-Reduced Thorium on the Room-Temperature Tensile Properties and Hardness of Iodide Thorium. (Base metal compositions and metallurgical treatments: BMI - 0.012% C, 0.080% O; forged and hot rolled at 700°C, cold reduced 50%, annealed in argon 2 hr at 850°C. ORNL - 0.015% C, 0.080% O; arc-melted, cold reduced 85%, vacuum annealed 0.5 hr at 650°C.)

Table 2. Typical Properties of Wrought-Annealed and Cold-Worked Thorium at Room Temperature

Condition When Tested	Tensile Strength (psi)	Yield Strength, 0.2% Offset (psi)	Elongation (%)	Gage Length (in.)	Reduction in Area (%)
Iodide					
Wrought-annealed sheet ^a	17,300	6,900	36	2	62
Wrought-annealed sheet ^b	19,700	11,200	44	1	60
Bomb-Reduced					
Extruded-annealed rod ^c	30,000	21,700	51	2	74
Extruded-annealed rod ^d	34,400	27,600	51	2	73
Wrought-annealed rod ^e	33,700	26,300	55	1.4	69
Extruded-annealed rod ^f	38,500	31,900	48	2	69
Wrought-annealed sheet ^g	39,600	30,300	--	--	52
Cold rolled 37.5% rod ^h	49,000	45,400	20	1.4	61
Cold rolled 25% sheet ⁱ	58,700	54,800	11	1	39
Cold rolled 50% sheet ⁱ	65,400	61,600	5	1	16

^aAverage of 4 lots, cold reduced 85%, annealed 0.5 hr at 650°C.

^bForged and hot rolled at 700°C, cold rolled 50%, annealed 2 hr at 850°C.

^cAverage of 4 lots, 0.05% C, annealed 0.5 hr at 750°C after extrusion.

^dAverage of 4 lots, 0.07% C, annealed 0.5 hr at 750°C after extrusion.

^eAverage of 4 lots, 0.06 to 0.085% C, reduced 95% by extrusion, cold reduced 37.5%, annealed 0.5 hr at 750°C.

^fAverage of 4 lots, 0.09% C, annealed 0.5 hr at 750°C after extrusion.

^gForged and hot rolled at 700°C, annealed 1 hr at 750°C, 0.11% C.

^hAverage of 3 lots, 0.06 to 0.085% C, reduced 72% by extrusion, cold rolled 37.5%.

ⁱ0.11% C, forged and hot rolled at 700°C prior to cold rolling.

The modulus of elasticity¹ of thorium is about 10.4×10^6 psi at 25°C and it decreases linearly with increasing temperature to about 7×10^6 psi at 500°C. Slightly higher values of 11.6×10^6 psi at 25°C decreasing by 7400 psi/°C to 300°C were more recently reported by Livesey⁸ as the result of dynamic measurements. For uranium, values of 27×10^6 at 25°C and 23.4×10^6 at 300°C have been reported.²

Thorium Alloys

Considerable research has been done on alloying thorium to improve the mechanical properties for use in reactor fuel elements. Many potential hardeners were found, some of the most effective being C, Zr, Al, In, Mo, and U. Detailed information on effects of specific elements and metallurgical factors involved can be found in earlier reviews.¹⁻³ More recent studies of thorium have been directed toward improvement of high-temperature properties, and we shall briefly summarize the data from several recent studies on potential fuel alloy systems.

Solid solution hardening, precipitation hardening, and dispersion hardening have all been considered for improving high-temperature properties of thorium. For exploratory studies, hot hardness has been a frequent method of evaluation, but hot tensile tests and creep tests have also been used. The results of separate studies are not directly comparable, probably because of differing impurity constituents in the base thorium and slightly differing experimental techniques.

Thorium-Uranium Alloys

Uranium is an essential constituent in thorium fuel alloys because it is added to provide a fissionable isotope and because it is a product of neutron irradiation. The solubility of uranium was reported by Rough and Bauer⁹ as about 1.8% at 1100°C, 1.1% at 600°C, and 0.7% at 25°C. Essentially similar results were reported by Murray¹⁰ and Bente,¹¹ but a maximum solubility limit of 2.7% at 1343°C has been suggested by Bannister.⁵ Bente's results predicted somewhat higher solubility at 1250°C. Uranium in excess of the solubility limit

exists as free uranium, which at levels in excess of about 15% is present as a grain boundary network regardless of fabricating techniques used.¹² Below about 15% U, the composition of the alloy and the metallurgical treatments will govern the size and distribution of the uranium phase, which in turn affect the physical properties and irradiation performance. High-temperature heat treatment tends to cause agglomerations of uranium and decrease strength.¹³ Battelle¹² has studied the effect of casting and fabricating techniques on hot hardness and creep strength of alloys containing up to 20% U. Limited data indicated that Th-5% U had greater creep strength at 600 and 700°C than alloys containing greater amounts of uranium. Kittel¹⁴ found superior irradiation performance on alloys containing 15% U or less.

Creep data by Atomics International,¹⁵ shown in Table 3 for annealed, swaged, and swaged and thermally cycled Th-9% U, show that the strengthening effects of mechanical work are retained at 600°C.

Table 3. Creep Data from Th-9% U Tested at 600°C

Alloy Condition	Stress (psi)	Strain Rate (%/hr)
Annealed	4000	0.017
Annealed	4500	0.033
Swaged	6000	0.010
Swaged	7000	0.026
Swaged	8000	0.060
Swaged and thermally cycled	5000	0.0035
Swaged and thermally cycled	5700	0.0060
Swaged and thermally cycled	7000	0.017

Ternary Alloys Containing Uranium

Battelle Memorial Institute¹² also investigated high-temperature mechanical properties of fabricated ternary alloys containing 10% U. Relative behavior of the alloys was the same at 600 and 700°C. Of the alloys tested in creep, those containing 0.5% Be, 1.5% Mo, or 2% Nb had creep strengths appreciably above that of the binary 10% U alloy; some appeared to be very slightly stronger than the binary Th-5% U alloy. Alloys containing both 10% U and 10% Zr were somewhat inferior to the binary 10% U alloy in creep strength but had significantly higher tensile and yield strengths at both 600 and 700°C. Hot hardness test results were also reported for cast specimens containing the various ternary additions to Th-10% U. Alloys containing 10% Zr, 0.1% Be, 0.2% C, and 1.5% Mo were significantly harder than the 10% U base alloy at 600°C and below.

Cole and Wilkinson,¹³ of Advanced Technology Laboratories (ATL) also studied ternary additions to thorium containing 5 or 10% U. Tensile and stress-rupture strengths of the base alloys at 600 and 800°C could essentially be doubled. Most noticeable improvement was attained with 2 and 5% Zr, although additions of Nb, Mo, C, Al, and Be also significantly improved strength. Solid solution hardening and dispersion hardening were obtained. The mechanical properties were sensitive to the size and distribution of the uranium-phase particles, and careful control of melting and heat-treatment procedures was utilized to achieve optimum structures and properties. Carbon at levels of 0.2 and 0.25% only slightly improved the properties of the binary Th-U alloys at 600°C and above but was very effective at 400°C and below. However, carbon was detrimental in alloys that also contained zirconium.

Of the alloys tested by ATL, superior strengths were obtained for quenched and aged alloys. Table 4 shows typical results including the effects of extended aging treatments. The data indicate that overaging has no detrimental effects. The extended aging treatments did not significantly alter the microstructure.

Table 4. Effects of Aging Treatments on the Strength
of Heat Treated Th-U-Zr Alloys^a

Alloy	Aging Conditions		Ultimate Tensile Strength (psi)	Yield Strength (psi)
	Time (hr)	Temperature (°C)		
Th-5% U-2% Zr	2	850	12,900	11,190
	100	800	7,700	7,250
	170	850	8,200	7,530
	2	850	8,350	7,585
	200	800		
Th-5% U-5% Zr	2	850	13,230	11,730
	100	800	11,800	4,800
	170	850	14,115	12,460
	2	850	12,050	10,400
	200	800		
Th-10% U-2% Zr	2	850	9,015	7,870
	100	800	10,500	8,900
	170	850	8,450	7,725
	2	850	10,550	9,230
	200	800		
Th-10% U-5% Zr	2	850	7,900	7,350
	100	800	9,250	7,975
	170	850	11,150	7,330
	2	850	8,000	6,650
	200	800		

^aTested at 800°C.

High-Temperature Strengthening

Burka and Hammond¹⁶ at ORNL studied the effects of many binary and some ternary additions to arc-melted thorium and evaluated results by hot hardness traverses on hot rolled plate. Carbon was found to be principally effective at below 600°C, although 0.2% C in combination with Cr, Mo, U, or Be appeared to enhance hot hardness at 600°C. Best hardening alloy additives at 600 and 750°C were 2 or 4% In and 5% Zr. The hardness of the various alloys at 600 and 750°C is shown in Table 5. Indium was less effective as a hardener in alloys prepared by powder metallurgy.¹⁷ Murray¹⁸ has studied the solubility of indium in thorium and found a eutectic at 8.9% In and 1160°C. The solubility decreased to 2.5% at 1000°C and 0.85% at 800°C. Indium, of course, is not a desirable alloying element for thermal reactor fuel.

Burka and Hammond^{17,19} also investigated the hardening of thorium by fine particle dispersions of ThO₂. The compacts, fabricated by extrusion of ball milled ThH₄ and ThO₂ powders, showed high strength and hardness and considerable ductility at 800°C. The experiments also indicated that control of the oxide and carbon pickup during the milling was quite difficult.

Compatibility with Cladding Materials

We have not yet surveyed information on the compatibility of thorium with potential clad materials, but little appears available, particularly for the temperature range of power reactors. Hanford²⁰ has successfully clad tubular Th-2.5% U-1.0% Zr by coextrusion with Zircaloy-2 and is now testing the material under irradiation in pressurized water. Zircaloy-2 and the fuel alloy interdiffuse slightly at 700°C, but the materials are compatible in the operating temperature range of water-cooled power reactors.

Table 5. Thorium-Base Alloys in Order of
Decreasing Hardness at 600 and 750°C

600°C		750°C	
Alloy (wt %)	DPH	Alloy (wt %)	DPH
4 In	91	4 In	55
2 In	63	5 Zr	37
10 Zr	59	4 In-5 Zr	34
5 Zr	59	5 V-0.2 C	33
5 Zr-0.2 C	54	2 In	31
20 Zr	54	5 Zr-0.2 C	31
5 V-0.2 C	50	5 U-0.2 C	26
4 In-5 Zr	49	20 Zr	25
2 In-0.2 C	49	10 Zr	25
2 Cr-0.2 C	47	8 Nb	25
2 Mo-0.2 C	44	5 Ti-0.2 C	24
5 U-0.2 C	44	5 Ti	23
0.2 Al-0.2 C	43	2 In-0.2 C	23
8 Nb	43	0.2 Be-0.2 C	20
0.2 Be-0.2 C	40	5 V	20
5 Ti-0.2 C	40	2 Cr-0.2 C	20
0.2 C	38	0.2 Al-0.2 C	20
5 Ti	37	0.4 Al	19
20 U	36	0.2 Al	19
0.4 Al	35	20 U	18
8 Cr	35	8 Cr	18
0.2 Al	32	5 U	18
5 V	32	8 Mo	17
5 U	30	2 Mo-0.2 C	17
8 Mo	30	4 Nb	17
0.15 C	29	10 U	15
2 Nb-0.2 C	29	4 Mo	15
10 U	28	2 Nb	14
4 Mo	27	0.2 Be	14
4 Cr	27	0.2 C	13
4 Nb	26	4 Cr	13
2 Nb	26	2 Mo	13
2 Mo	24	0.15 C	11
0.10 C	23	Thorium	11
Thorium	22	2 Cr	11
2 Cr	21	2 Nb-0.2 C	11
0.2 Be	21	0.10 C	9

Summary

The data on physical and mechanical properties of thorium metal suggest that it may have considerable potential as a reactor fuel. A preliminary analysis of creep data, high-temperature strength, and hot hardness data suggests that at temperatures above about 500°C thorium is significantly stronger than uranium. Even at temperatures of 400 to 550°C, anisotropy causes internal stresses under thermal cycling or neutron irradiation, which decrease the creep strength of uranium significantly below that obtained at isothermal conditions in the absence of irradiation. Since thorium, with its isotropic structure, may not be subject to such internal stresses, its properties may not be degraded to the same extent. However, the effect on creep strength of fission events during irradiation of thorium has yet to be established.

Recent developments with uranium alloy fuels have emphasized the importance of microstructural control for improving irradiation behavior of the metal, and data indicate that fine particle dispersions are most effective in increasing dimensional stability of fuel alloys at high temperature. Similar methods may be effective with thorium and should be investigated.

Thorium fueled reactors will likely have greatest economy if initial uranium concentrations are low and if generated ^{233}U contributes substantially to the burnup. Thus, long-time operation of the fuel will be required. Experiments are needed to establish the radiation performance of thorium in which burnup of generated ^{233}U is substantial.

Mechanical property data suggest that thorium fuels may be able to operate at considerably higher temperatures than uranium. If such abilities are to be utilized, consideration must be given to compatibility of the fuel with potential clad materials.

CERAMIC COMPOUNDS OF THORIUM

Many thorium compounds exist with the high-temperature stability required for useful ceramic bodies. These include ThBe_{13} , ThB_4 , ThB_6 ,

ThC, ThC₂, ThN and higher nitrides, ThO₂, and several phosphides, silicides, and sulfides. Time does not permit reviewing here the state of knowledge of the properties of most of these except to say that for the most part it is incomplete and unverified. The literature on ceramic compounds of thorium is rampant with guesses, misquotations, and incomplete qualifications, so the original sources must be traced for all data. This we have not done for this brief review, but we are preparing an extensive collection of data on thorium ceramic compounds.²¹ Our attention here will emphasize the compounds of most immediate nuclear application.

Thorium Dioxide

Thorium dioxide is without doubt the best characterized ceramic compound of thorium. Although this partly stems from its study for nuclear purposes, a great deal of information exists because of the non-nuclear usefulness of the material. Since thoria is the highest melting and the most stable to reduction of all the refractory oxides, it is a superior crucible material for the melting of reactive metals. Thoria is generally prepared in powder form by the thermal decomposition of a purified salt, usually the oxalate. This powder can be consolidated by usual ceramic fabrication techniques, such as slip casting, pressing and sintering, or hot pressing. The fabricability and ceramic properties can often be related to conditions of preparation of the starting salt and firing. Fabrication, properties, and uses of thoria ceramics have recently been reviewed by Hepworth and Rutherford²² and by Ryshkewitch.²³

Thorium dioxide exists up to its melting point as a single cubic phase with the fluorite crystal structure, isomorphous and completely miscible with UO₂. Unlike UO₂, thoria does not dissolve oxygen to a measurable extent. Therefore it is stable to high temperature in oxidizing environments. On prolonged heating to 1800 to 1900°C in vacuum it blackens with loss of oxygen, although the loss is insufficient to be reflected in chemical analysis or lattice-parameter measurement.

On reheating in air to 1200 or 1300°C the white color is restored. When uranium dioxide is incorporated in thoria, the lattice can take up extra oxygen in proportion to the uranium content.

Table 6 summarizes the more important physical and mechanical properties of thoria, along with analogous properties of uranium dioxide taken from the compilation by Belle.²⁴ In some cases the data are a small selection of what are available. The heat capacity is known with precision from near absolute zero up to over 900°C, and other thermodynamic functions have been derived from it.^{25,26} Less precise data exists^{27,28} up to 2400°C. The heat and free energy of formation are from a compilation²⁹ that extends up to 2000°C, although it is not based on the best and latest data. The lack of a value for the heat of fusion illustrates the state of confusion on properties of thoria. A value of 21.4 kcal/mole has been attributed to Lambertson, Mueller, and Gunzel.³⁰ These authors needed a value for a theoretical interpretation of their UO₂-ThO₂ melting point data and guessed it by taking 3R as the entropy of fusion. Mechanical properties up to 900°C and higher are given in the compilation by Ryshkewitch²³ and elsewhere, but these should be taken as examples of particular specimens and not as values to be expected. The actual values depend on density, porosity, grain size, and other qualities dependent on fabrication and vary considerably. Preliminary measurements on pure dense specimens prepared at Oak Ridge National Laboratory³¹ indicate higher strengths than those tabulated. Recent data³¹ on the compressive creep of thoria are given in Fig. 2. Thoria is a high-temperature semiconductor, so its thermal and, even more so, electrical conductivity depend on fabrication history and impurity content. The latter property shows quite wide variations.

Although a thorium-base fuel will necessarily contain uranium, little information exists on the properties of thoria-urania solid solutions. Physical and mechanical properties are probably in most cases intermediate between those of the pure components fabricated similarly. However, this is not true of transport properties such as thermal conductivity. Kingery⁴² found lower conductivity for mixed

Table 6. Important Properties of Thorium and Uranium Dioxides

Property	Value for ThO ₂	Reference	Value for UO ₂ (Ref. 24)
Crystal Structure	Face-centered cubic (CaF ₂ type)		Face-centered cubic (CaF ₂ type)
Space group	O _h ⁵ Fm3m		O _h ⁵ Fm3m
Lattice parameter, A	5.5974 at 26°C 5.6448 at 942°C	32	5.4704 at 20°C 5.5246 at 946°C
Theoretical density, g/cm ³	10.00		10.96
Interatomic distances, A			
M-M	3.958		3.868
O-O	2.799		2.735
M-O	2.424		2.368
Thermal Properties			
Melting point, °C	3300 ± 100	30	2760 ± 30
Spectral emissivity, λ = 0.65μ	0.53 at 300°C to 0.21 at 800°C 0.2 to 0.65 at 1300°C depending on sample history	33 34	0.416 ± 0.026 (near m.p.) 0.850 at 727°C 0.370 at 1947°C
Thermal conductivity, w cm ⁻¹ °C ⁻¹	0.103 at 100°C 0.034 at 800°C 0.086 at 200°C 0.031 at 1000°C 0.060 at 400°C 0.025 at 1200°C 0.044 at 600°C	35	0.105 at 100°C 0.0815 at 200°C 0.0590 at 400°C 0.0452 at 600°C 0.0376 at 800°C 0.0351 at 1000°C
Heat capacity, cal mole ⁻¹ °C ⁻¹ (298 to 1200°C)	17.060 + 18.06(10 ⁻⁴)T - 2.5166(10 ⁻⁵)/T ²	26	18.45 + 2.431(10 ⁻³)T - 2.272(10 ⁻⁵)/T ²
Debye temperature, °K	200		870°K < 600°K (300-600°K)
Coefficient of linear expansion, °C ⁻¹	6216(10 ⁻⁶) + 3.541(10 ⁻⁹)T - 0.1125/T ²	36	10.8 × 10 ⁻⁶ (20-926°C) 9.9 × 10 ⁻⁶ (25-800°C) 10.0 × 10 ⁻⁶ (400-900°C) 10.52 × 10 ⁻⁶ (26-1000°C)
Vapor pressure, atm	7.64 - 3.440(10 ⁻⁴)/T (2200-2900°K) 9.02 - 3.78 (10 ⁻⁴)/T (2170-2400°K)	37 38	log P = -33, 115/T - 4.026 log T + 23.111
Thermodynamic Properties			
Heat of formation, ΔH ₂₉₈ , kcal/mole	293.2 ± 0.4	29	259.2 ± 0.6
Free energy of formation, -ΔF ₂₉₈ , kcal/mole	279.2 ± 0.65	29	246.6 ± 0.6
Entropy, 298°K, cal mole ⁻¹ °C ⁻¹	15.593 ± 0.02	25	18.6 ± 0.1
Heat of sublimation, kcal/mole	158.7 ± 2.5, in range 2000-3000°K	37	137.1 ± 1.7 at 1800°K
Entropy of sublimation, cal mole ⁻¹ °C ⁻¹	35.3 ± 1.0 in range 2000-3000°K	37	36.4 at 1800°K
Mechanical Properties			
Elastic Properties			
Young's modulus, kilobars, psi	1370, 19.8(10 ⁶)	23	1930, 28.0(10 ⁶) at room temperature 1827, 26.5(10 ⁶) at room temperature 1655, 24.0(10 ⁶) at 800°C
Shear modulus, kilobars, psi	990, 14.3(10 ⁶) at 30°C 390, 5.6(10 ⁶) at 1300°C	39	745, 10.8(10 ⁶) at room temperature
Poisson's ratio	0.17		0.302 at room temperature
Modulus of rupture, kilobars, psi	0.83, 12,000	40	0.965-1.10, 14,000-16,000
Compressive strength, kilobars, psi	15, 214,000	23	4.14-9.65, 60,000-140,000
Fracture strength, kilobars, psi	1, 14,000	23	0.16-0.37, 2300-5400
Hardness, Knoop	640 (500-g load)	41	666 ± 14 625
Electrical, Magnetic, and Optical Properties			
Electrical resistivity, ohm cm	1 to > 10 ⁴ at 1600°C, depending on sample and treatment	23	3 × 10 ⁻⁴ - 10 ⁻⁶ at room temperature about 10 ⁻¹ at 500°C about 10 ⁻¹ at 1000°C
Index of refraction	2.09		2.35 1.2-2.7

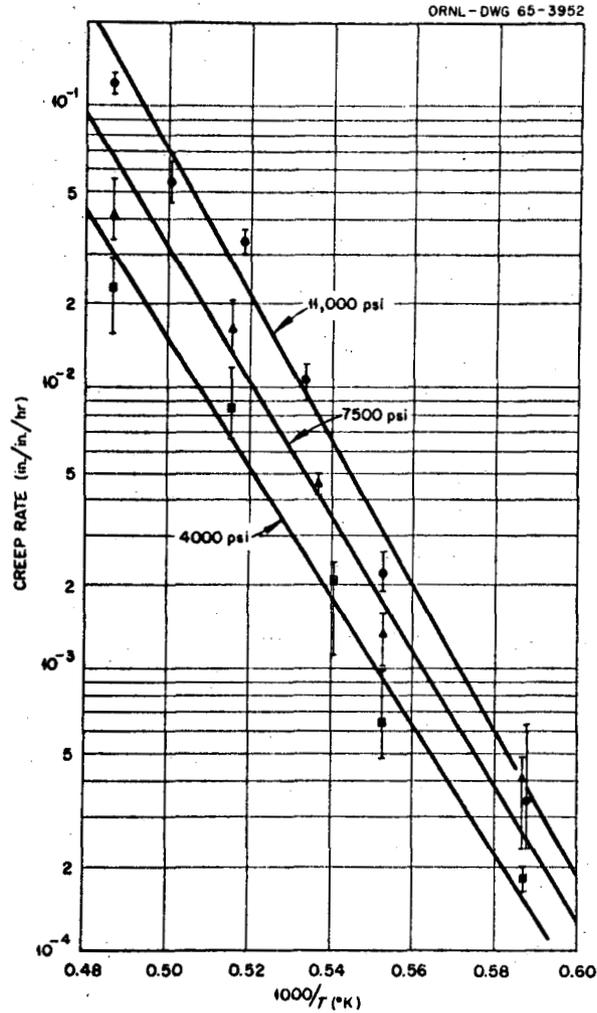


Fig. 2 Compressive Creep Behavior of Thoria. Specimens were isostatically pressed at 35,000 psi and fired in air for 2 hr at 1800°C. Density 97.5% of theoretical, average grain diameter about 10 μ .

Thorium Carbides

Next to thoria, the most prominent thorium ceramic compounds for nuclear application are the carbides. The monocarbide ThC and the dicarbide ThC₂ can both be prepared by a variety of techniques. Arc-melting of the elements, pressing and sintering of the powdered elements, and carbon reduction of the oxide are among the frequently encountered methods. The monocarbide phase apparently is capable of appreciably less but not greater than the stoichiometric carbon content. The same is true of the dicarbide, and preparations of it frequently are hypostoichiometric.

Information on properties of thorium carbides is very scant compared with that on the oxide. Properties of the two carbides are summarized in Table 8. Comparison with Table 6 points up the many gaps in our knowledge. Many of these gaps are probably already being filled, because of the growing interest in these materials. For example, several new publications⁴⁶⁻⁵¹ relate to the thermodynamic properties.

Information on the compatibility of thorium carbides with other materials is scarce. This survey has turned up nothing on compatibility of pure carbides with metals, although likely the thorium carbides resemble the uranium carbides in this. The carbides are reactive toward air and water. Powders have to be handled in inert atmospheres. Various hydrocarbons result from the reaction of the carbides with water.^{52, 57}

In contrast with the oxide, the carbides have been studied principally mixed with uranium. Complete isomorphous solid solutions exist between uranium and thorium monocarbides. The binary dicarbide system is more complex; an equilibrium diagram has been proposed by Hill and Cavin.⁵³ The thorium compound is monoclinic but very nearly orthorhombic. On sufficient heating or substitution with uranium it gradually transforms to body-centered tetragonal. This phase is not completely miscible with the body-centered tetragonal uranium compound, but the two-phase region is narrow.

Several thorium compounds, including both carbides, were prepared by arc-melting with 10 and 20% substitution of uranium for the thorium.⁵⁸

Table 8. Important Properties of Thorium Carbides

Property	ThC ₂		ThC	
	Value	Reference	Value	Reference
Crystal Structure	Pseudo-orthorhombic		Face-centered cubic (NaCl)	
Lattice parameter, Å	10.555 8.233 4.201	52	5.346	52
Theoretical density, g/cm ³	9.6		10.61	52
Interatomic distance, Å				
Th-Th			3.780	
C-C			3.780	52
Th-C			2.673	
Thermal Properties				
Melting point, °C	2655 2640	52 53	2625	52
Specific heat, cal mole ⁻¹ °C ⁻¹	13.55	46		
Coefficient of linear expansion, °C ⁻¹	7.29(10 ⁻⁶) for 40-400°C 8.80(10 ⁻⁶) for 40-1000°C	54		
Thermodynamic Properties				
Heat of formation, -ΔH ₂₉₈ , kcal/mole	44.8 29.6 ± 4.8	56 46		
Free energy of formation, -ΔF, kcal/mole, 298-2300°K	45 - 2.6(10 ⁻³)T	56		
Entropy, 298°K, cal mole ⁻¹ °C ⁻¹	19.3 16.38	56 46		
Mechanical Properties				
Hardness, diamond pyramid, 200-g load	600 kg/mm ²	52	850 kg/mm ²	52
Electrical Properties				
Electrical resistivity, ohm cm, 25°C	30(10 ⁻⁶)	52	25(10 ⁻⁶)	52

Table 9 lists several properties measured and Table 10 gives results of compatibility tests with various materials. The same authors also reported weight gains in air graphically for the carbides with 10% substitution. Cook *et al.*⁵⁴ measured the thermal expansion over the range 40 to 1000°C for ThC₂ and several mixed dicarbides containing up to 75% U substituted. Increasing the uranium content or the temperature increased the thermal expansion; the increase with uranium content was monotonic except at the two-phase region.

Table 9. Properties of Arc-Melted Thorium-Uranium Carbides

Property	Temperature (°C)	Value for	
		Th _{0.9} U _{0.1} C	Th _{0.9} U _{0.1} C ₂
Thermal conductivity, w cm ⁻¹ °C ⁻¹	200	0.13	0.20
	800	0.17	0.25
Thermal expansion, %	0-400	0.24	0.31
	0-900	0.64	0.69
Hardness, kg/mm ² (1 kg load)	25	900	1100
	1000	260	160
Compressive fracture stress, psi	25	1.3 × 10 ⁵	1.5 × 10 ⁵
	975	6.8 × 10 ⁴	2.3 × 10 ⁴
Thermal expansion coefficient, ^a °C ⁻¹	40-400		7.84 × 10 ⁻⁶
	40-1000		9.31 × 10 ⁻⁶

^aValues for 12.3% UC₂ taken from Reference 54.

Table 10. Compatibility Properties of Thorium-Uranium Carbides

Contact Materials	Behavior of (a) $\text{Th}_{0.9}\text{U}_{0.1}\text{C}$, (b) $\text{Th}_{0.8}\text{U}_{0.2}\text{C}$, (c) $\text{Th}_{0.9}\text{U}_{0.1}\text{C}_2$, (d) $\text{Th}_{0.8}\text{U}_{0.2}\text{C}_2$
Al, Zr	all four, no reaction in 1000 hr at 540°C
Inconel, type 321 stainless steel	all four, no reaction in 1000 hr at 815°C
Ta, W	all four, no reaction in 1000 hr at 1095°C
Nb, Mo	all four, no reaction in 1000 hr at 815°C all four, no reaction in 100 hr at 1095°C all four, < 0.002 in. penetration in 1000 hr at 1095°C
H ₂ O at 90°C	(a), (c), ThC, ThC ₂ all very poor
Santowax R at 350°C	Weight loss in $\text{mg cm}^{-2} \text{ day}^{-1}$ (a), 2.0; (c), 0.3; ThC, 0.35; ThC ₂ , 0.5
NaK at 650°C	Weight loss in $\text{mg cm}^{-2} \text{ day}^{-1}$ (a), 0.045; (c), 0.015

Thorium Nitrides

The thorium nitrides reported are ThN, Th₂N₃, and Th₃N₄. The latter two may not both exist, and their use at high temperatures, if possible at all, would require high nitrogen pressures. The mononitride is stable to its melting point of 2790°C in the presence of sufficient nitrogen; less than 1 atm is needed.⁵⁹ It has a face-centered cubic crystal structure (NaCl type) with a lattice parameter of 5.1584 Å and a narrow composition range. The decomposition pressure in atmospheres is given by

$$\log P = 8.086 - 33,244/T + 0.958(10^{-17})T^5$$

It can be prepared by induction melting of thorium under 2 atm N₂ or by hot pressing powder from decomposition of Th₂N₃. With ThC it forms a complete series of solid solutions.⁶⁰ Thorium mononitride is more reactive toward water than UN.⁶¹ Otherwise, thorium nitride information is lacking except for some guessed thermodynamic properties.

Miscellaneous Thorium Compounds

Thorium forms two borides, ThB_4 and ThB_6 . Some available physical and thermal data^{62,63} indicate that the tetraboride is a promising ceramic material, and a few properties have been reported⁵⁸ for material with incorporated uranium. However, the nuclear usefulness of this material requires separated ^{11}B . With silicon and beryllium thorium forms the potentially useful Th_3Si_2 , ThSi , and ThBe_{13} . The data on these compounds is quite scanty, but a few properties have been studied for uranium substitution products.⁵⁸ Thorium forms several high-melting sulfides and an oxysulfide ThOS . Several of these show promising ceramic properties, but we have not yet completed a survey of them. The compounds ThS and Th_2S_3 have received the most attention. There is no information to indicate that phosphides of thorium would have nuclear application.

Summary

The data on physical properties of thorium oxide are quite extensive. The extensive data on mechanical properties is misleading, however, since these properties depend greatly on the fabrication of the specimen. Data on diffusion in thoria is completely lacking. Compatibility with many materials is excellent, either known from experiment or safely predictable. However, information on thoria containing uranium is very meager. Despite the regular isomorphous replacement, the disorder introduced precludes prediction of properties by interpolation between those of the components; the scant data on thermal conductivity prove this.

In contrast is the situation on the carbides, which are of interest only for application in nuclear energy. Information on these compounds is quite scanty, but the rate at which new knowledge is being reported is quite high. Reflecting the nuclear interest, the information on mixed carbides with uranium is keeping pace with or even exceeding that on pure thorium carbides.

The compounds ThBe_{13} , ThB_4 , ThN , ThSi , Th_3Si_2 , ThS , and Th_2S_3 have been partially characterized for ceramic nuclear use.

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