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PROPERTIES OF CERAMIC AND CERMET FUELS FOR SODIUM GRAPHITE REACTORS

AEC Research and Development Report

ATOMICS INTERNATIONAL
A DIVISION OF NORTH AMERICAN AVIATION, INC.

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Department of Commerce
Washington 25, D. C.

NAA-SR-3880
METALLURGY AND CERAMICS
50 PAGES

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By:
N. R. KOENIG
B. A. WEBB

ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN AVIATION, INC.
P.O. BOX 309 CANOGA PARK, CALIFORNIA

CONTRACT: AT(11-1)-GEN-8
ISSUED: JUN 30 1960

DISTRIBUTION

This report has been distributed according to the category "Metallurgy and Ceramics" as given in "Standard Distribution Lists for Unclassified Scientific and Technical Reports" TID-4500 (14th Ed.), October 1, 1958. A total of 605 copies was printed.

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ABSTRACT

Nuclear materials of interest as potential reactor fuels for sodium graphite reactors are reviewed to select those which appear most feasible for high-temperature, long-burnup application.

Fuel properties such as melting point, thermal neutron absorption cross section, uranium content, chemical and physical properties, and fabrication details are presented. Other factors such as expected burnup capabilities, required enrichments, and conversion ratios have been compared.

A program is currently in progress to evaluate promising nuclear fuels. Fuel materials under consideration include the uranium compounds, uranium dioxide, uranium monocarbide, borides, sulfides, aluminide, nitrides, silicides, and phosphides.

Various cermets are also under consideration. These include dispersions of uranium compounds in matrices of uranium, uranium alloys, thorium, and thorium alloys. Included among the uranium alloy matrices are binary and ternary combinations of uranium with niobium, molybdenum, and zirconium.

I. INTRODUCTION

The growing trend toward higher operating temperatures in nuclear reactors indicates the necessity to develop new fuels which will withstand long burnups at high temperatures. Several factors must be considered in the selection of materials for use as fuels. It is desirable to have a material with a high uranium density, low neutron absorption cross section, and a high melting temperature with no phase changes to the melting point. Other desirable characteristics include a high thermal conductivity, compatibility with cladding material and coolant, good mechanical strength and thermal shock resistance, and most important, a high degree of irradiation stability. The ability of a fuel material to retain a high percentage of gaseous fission products is another characteristic which is desirable.

The use of unalloyed uranium metal as a fuel material in nuclear reactors has encountered many difficulties, the most significant of which is the lack of dimensional stability under irradiation above approximately 450°C. Depending on the history of the fuel, permanent macroscopic deformation, in the form of warping and dimensional changes, results. This gross distortion is associated with the generation and accumulation of gaseous fission products. These gases diffuse to nucleation sites and form small bubbles which expand, causing the uranium metal to increase in volume. This phenomenon has a serious effect on solid unalloyed uranium fuels at operating temperatures above 450°C.

Efforts to minimize or eliminate these effects have followed several approaches. Strengthening through alloying offers a method of reducing the amount of deformation observed for a given stress. An alternative approach is the use of high-melting compounds of uranium in which fission gases either escape readily, or are retained in a dispersed atomic state within the lattice so that they do not accumulate to form bubbles. A third approach is to use combinations of alloys and compounds as cermets. In cermets, the stable, refractory compound is dispersed in a high-strength alloy matrix, which effectively takes advantage of the desirable properties of each material.

A program is now in progress to evaluate various uranium-bearing materials as nuclear fuels. The study consists of two phases. The first is the study of uranium compounds with regard to fabrication problems, physical and chemical properties, and irradiation stability. The second phase consists of studying the properties and irradiation characteristics of cermets composed of uranium compounds dispersed in fissile alloy matrices. Too few data are now available to predict which materials or combinations of materials may be suitable as high-temperature fuels, but the characteristics of these materials and their possible performance as nuclear fuels will be reviewed in this paper.

II. URANIUM COMPOUNDS

There are several compounds of uranium which may be useful as nuclear fuel materials. After a preliminary survey, certain binary uranium compounds were selected for further study on the basis of melting point, thermal neutron absorption cross section, and uranium inventory.

Binary uranium compounds, formed with elements having a thermal neutron absorption cross section of less than 20 barns, are listed with approximate melting points in Table I. The compounds are listed in Table II in the order of their decreasing uranium content. In Table VII, those compounds with greater than 80 wt % uranium content are tabulated with their density, weight percent of uranium, weight of uranium per cubic centimeter of compound, crystal structure, and thermal neutron absorption cross section of the second element.

The compounds in Table VII exhibit basic properties, such as high uranium density, high melting point, and low thermal neutron absorption cross section, which are desirable in a high-temperature nuclear fuel. U_3Si, U_6Fe, and U_2Ti have relatively low decomposition points, but were included because of their high uranium inventories. In systems containing more than one compound, interest is generally in the phase with the highest uranium content.

The binary uranium compounds of greatest interest in this program are those of aluminum, boron, carbon, iron, nitrogen, phosphorus, silicon, sulfur, and titanium.

A. URANIUM-ALUMINUM

Uranium combines with aluminum to form three intermetallic compounds: UAl_2, UAl_3, and UAl_4. UAl_2 has the highest uranium density and highest melting point, and therefore is of primary interest as a fuel material. Data on crystallography and physical properties of UAl_2 are included in Table VII. The phase diagram for the U-Al system is reproduced in Figure 1.

UAl_2 has been synthesized by the fusion of stoichiometric mixtures of U + Al in an electric arc furnace. The resultant product has an extremely bright, silvery, metallic luster, and is relatively brittle. The material is easily cut with a silicon carbide wheel under a water coolant.[66] The reaction of UAl_2

TABLE I

MELTING POINT AND ABSORPTION CROSS SECTION OF URANIUM COMPOUNDS*

Compound	Melting Point (°C)	Absorption Cross Section of Alloying Element (σ_a) (barns)	Ref.	Compound	Melting Point (°C)	Absorption Cross Section of Alloying Element (σ_a) (barns)	Ref.	Compound	Melting Point (°C)	Absorption Cross Section of Alloying Element (σ_a) (barns)	Ref.
UH_3	435†	0.33	11	UF_3	1000†	0.009	11	UNi_2	810†	4.6	14
UBe_{13}	2000	0.010	14	UF_4	960	0.009	11	UNi_5	1300	4.6	14
UB^{11}_2	1500	0.050	14	UF_5	400†	0.009	11	UCu_5	1050†	3.7	14
UB^{11}_4	1500	0.050	14	UF_6	69†	0.009	11	UGa_2	-	2.9	14
UB^{11}_{12}	1000†	0.050	11,14	UAl_2	1590	0.230	14	UGa_3	1300	2.9	14
UC	2400	0.0032	11,14	UAl_3	1350†	0.230	14	UGe_3	-	2.3	14
U_2C_3	1800†	0.0032	11,14	UAl_4	730†	0.230	14	UBi	1400	0.033	14
UC_2	2450	0.0032	11,14	UAs	-	4.3	14	U_3Bi_4	1150	0.033	14
UN	3630	1.88	11,14	UAs_2	-	4.3	14	UBi_2	1010	0.033	14
U_2N_3	1300†	1.88	11,14	USe_2	-	13.0	11	UPd_5	-	8.0	14
UN_2	Unstable	1.88	11,14	UBr_3	730†	6.6	16	UPd_3	1650	8.0	14
U_3Si	930†	0.13	11,14	UBr_4	519†	6.6	16	U_5Sn_4	1500	0.6	14
U_3Si_2	1650	0.13	11,14	UBr_5	600†	6.6	16	U_3Sn_5	1350†	0.6	14
USi	1600	0.13	11,14	U_2Ru	947†	2.46	15	USn_3	1350	0.6	14
USi_2	1600	0.13	11,14	URu	1175	2.46	15	USb	-	5.5	14
USi_3	1500	0.13	11,14	U_3Ru_4	1200	2.46	15	U_3Sb_4	-	5.5	14
UP	-	0.19	14	U_2Ru_3	1100	2.46	15	USb_2	-	5.5	14
U_3P_4	-	0.19	14	URu_3	-	2.46	15	UTe	-	4.6	11
UP_2	-	0.19	14	U_2Ti	900†	5.6	14	U_2Te_3	-	4.6	11
US	2000	0.49	11,14	U_6Mn	716†	13.3	14	UI_3	680	6.3	16
U_2S_3	1850 ± 100	0.49	11,14	UMn_2	1100	13.3	14	UI_4	506†	6.3	16
US_2	1850 ± 200	0.49	11,14	U_6Fe	815†	2.53	14	UOs_2	-	15.0	14
UO_2	2850	0.002	11	UFe_2	1235	2.53	14	UPt_2	-	10.0	14
U_3O_8	900†	0.0002	11	U_6Ni	754†	4.6	14	UTl_3	-	3.3	14
U_4O_9	900†	0.0002	11	U_7Ni_9	790†	4.6	14	UPb	1280	0.17	14
UO_3	470†	0.0002	11	U_5Ni_7	820†	4.6	14	UPb_3	1210	0.17	14

*All compounds listed have a thermal neutron (0.025 ev) absorption cross section less than 20 barns.

†Decomposition temperature

TABLE II

URANIUM CONTENT OF SOME URANIUM COMPOUNDS

A		B		C			
90 - 100 wt % U		80 - 90 wt %		Below 80 wt % U			
U_3Si	96.21	USi	89.44	UP_2*	79.4	U_2Ru_3	61.30
U_6Fe	96.20	UP*	88.5	US_2	78.90	U_2Te_3*	55.5
UC	95.19	US	88.12	UAs*	76.1	U_3Sn_5	54.60
UN	94.44	UO_2	88.10	UF_4	75.90	UPb	53.40
U_3Si_2	92.70	U_3P_4*	85.2	UAl_3	74.60	UBi	53.20
U_2N_3	92.00	UB_4	84.61	USi_3	73.86	UGa_3	53.20
UB_2	91.66	U_2S_3	83.40	U_5Sn_4	71.5	UPd_2*	52.8
U_2Ti	90.80	U_2Ru	82.50	URu	70.30	UGe_3*	52.4
		UAl_2	81.52	UAl_4	68.80	USb_2*	49.5
		USi_2	80.91	UFe_2	68.00	U_3Bi_4	46.00
		UF_3	80.80	UBe_{13}	67.00	URu_3*	44.0
				USb*	66.2	UCu_5	42.80
				UTe*	65.0	UPd_3	42.80
				U_3Sb_4*	64.6	USn_3	40.15
				U_3Ru_4	63.80	UPt_2*	37.9
				UGa_2*	63.1	UBi_2	35.70
				UAs_2*	61.5	UTl_3*	27.9
						UPb_3	27.7

*Compounds with few or no available data

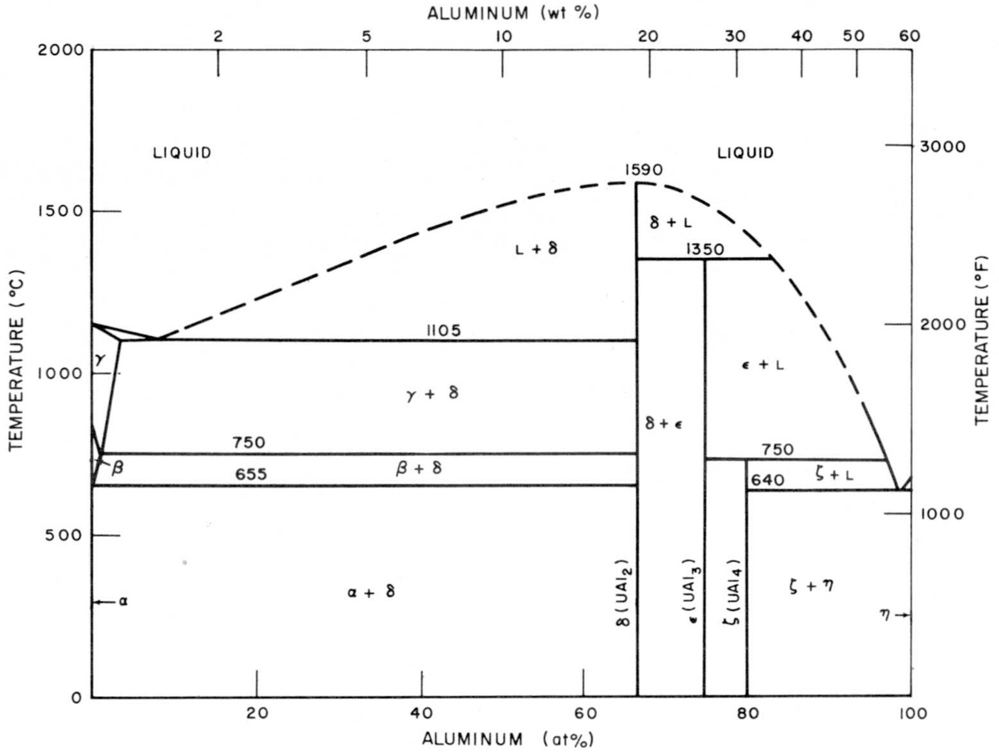

Figure 1. Uranium-Aluminum Phase Diagram

with graphite crucibles during casting is appreciable only at high temperatures. This effect is minimized with a relatively cool mold.[66]

UAl_2 forms a very thin, highly protective film upon exposure to air or water and has excellent corrosion resistance at room temperature.[66] No data on the thermal conductivity of this material have been found, but it is expected that it will be relatively high.

B. URANIUM-BORON

Uranium forms the refractory borides, UB_2 and UB_4, and a third boride, UB_{12}, which is of little interest because of its low uranium density and relative instability at high temperatures. The diboride, although not the most refractory, is of primary interest because of its high uranium density. Table VII shows the crystal structure and density for UB_2.

The phase diagram for the uranium-boron system has not been completed, but the U-UB_2 eutectic is reported to be very close to pure U, and the temperature very close to the U melting point (1132°C). The UB_2-UB_4 eutectic is thought to be considerably higher than 1570°C.[12,14]

There are no data available on thermal conductivity values for UB_2, but diborides of titanium, zirconium, and niobium all have higher reported thermal conductivity values than the respective monocarbides.[12]

There are several methods reported for the fabrication of UB_2:

1) Stoichiometric mixtures of uranium metal and amorphous boron are arc melted.[12]
2) Mixtures of uranium and boron powder are pressed and sintered in molybdenum crucibles under inert atmosphere at 1500°C for 1 hr. Compacts thus formed may be subsequently arc melted.[10]
3) Mixtures of the respective oxides are reduced by Al, Mg, Si, or C.[12]
4) Uranium or UO_2 is reacted with B_4C, with or without B_2O_3.[12]
5) Uranium powder and amorphous boron are hot pressed at 200 atmospheres and 1000°C to make electrodes for consumable arc melting.[11]

The silver gray product resultant from all these processes is resistant to aqueous alkalies and to acids, with the exception of nitric and hydrofluoric acids.[12]

Natural boron, containing 18.8% of the high cross section B^{10} (4010 barns) has too high a cross section for the reaction B^{10} (η, α) Li^7 to permit its use as a nuclear fuel. It is probable that the low (50 millibarns) cross section isotope, B^{11}, would be required. The high cost of the separated isotope may seriously influence any development of uranium diboride as a nuclear fuel.

C. URANIUM-CARBON

Uranium monocarbide is being extensively studied as a high temperature nuclear fuel. Physical and chemical properties, as well as recent irradiation data, make UC a promising fuel material. A comparison of crystallographic data and physical properties of UC with other uranium compounds is given in Table VII. Various reported properties of UC are compiled in Table III. Thermal conductivity is given in Table IV. The uranium-carbon phase diagram is reproduced in Figure 2.

TABLE III

SOME PROPERTIES OF URANIUM MONOCARBIDE

		Remarks	Ref.
Melting Point (°C)	2250		11
	2390		7
	2375		9
	2350 to 2400		10
	2590 ± 50		54
	2450 to 2500		12
Density	13.63 gm/cm^3	X-ray	62
Hardness	750 to 800 VPN	Sintered at 2100°C to 88% of Theoretical Density	13
	635 Knoop		17
Electrical Resistivity (ohm-cm)	50×10^{-6} at 20°C		68
	99×10^{-6} at 20°C		64
	33 to 44 $\times 10^{-6}$ at 20 °C		64
Thermal Expansion (/°C)	10 to 11 $\times 10^{-6}$	20 to 1000°C. Sintered at 2100°C to 88% of Theoretical Density	13
	10.5×10^{-6}	20 to 1000°C.	68
Thermal Conductivity (cal/sec-cm-°C)	.078	44°C	10
	.082	Temperature not specified	10
	.057	600°C. Arc melted and cast	64
	.032	60°C. Density 10.4 gm/cm^3	68
Modulus of Rupture (tsi)	5	20°C. Sintered at 2100°C	13
	20 to 25	20°C. 4.5 wt %	13
	10 to 15	1000°C	13
	20 to 30		17

TABLE IV

THERMAL CONDUCTIVITY OF URANIUM MONOCARBIDE*

Temperature (°C)	Cal/(sec-cm-°C)
100	0.0603
150	0.0583
200	0.0563
250	0.0550
300	0.0542
350	0.0536
400	0.0534
450	0.0535
500	0.0543
550	0.0555
600	0.0570
650	0.0586
700	0.0601
735	0.0615

*Density > 98% of theoretical. Arc-cast material. BMI data for UC - 5.2 wt % C specimen.

At the present time, techniques for the preparation of UC and the fabrication of UC shapes are being evaluated. There are several methods reported for the preparation of UC and the fabrication of shapes:

1) High density UC can be readily prepared in the electric arc furnace with tungsten, graphite, or consumable electrodes. The monocarbide thus formed can be cast into shapes in graphite molds.[11,12,13,17]

2) Uranium monocarbide is also prepared by carbon reduction of the oxides, UO_2 or U_3O_8, at temperatures greater than 1600°C. The UC thus formed can subsequently be pressed and sintered or hot pressed.[11,13]

3) UC can be prepared and fabricated into shapes in one operation, by mixing U and C powders and hot pressing at 900 to 1000°C, or cold pressing and sintering at temperatures in excess of 1200°C.[11,13,69]

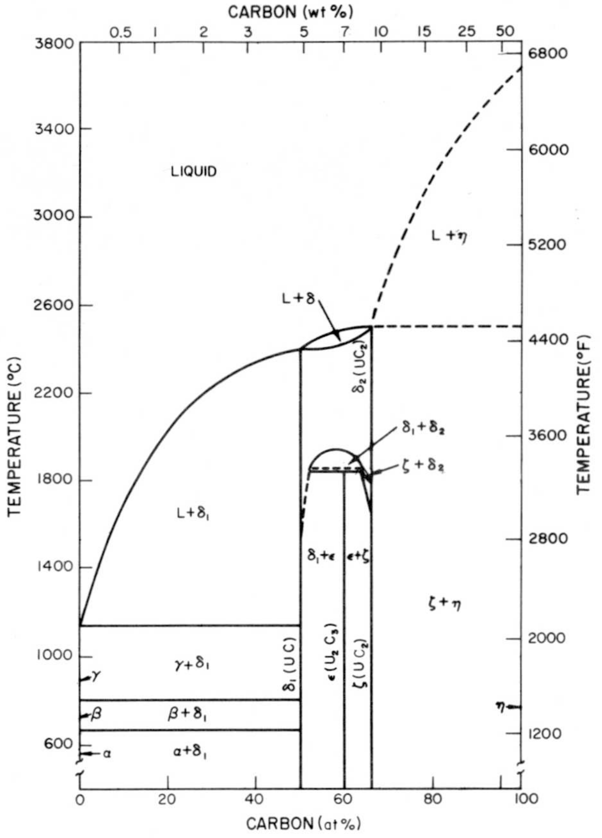

Figure 2. Uranium-Carbon Phase Diagram

4) Methane carburization of uranium is reported to produce high purity UC powder, at a temperature of 650 to 700°C.[10,11]

Finely divided UC is a black, pyrophoric powder; but in the cast condition it has a metallic luster and is quite brittle. UC can be wet ground with diamond wheels using a water-base coolant and can be drilled with carbide tools or by ultrasonics.[66]

Samples of arc melted UC reacted slowly with water at room temperature, although the threshold reaction temperature with water has also been reported to be 83°C.[17,66] UC oxidizes in CO_2 at reactor operating temperatures (500°C at 8 atm. pressure).[68] Compatibility tests showed that UC is relatively stable in isopropyl alcohol, n-butyl alcohol, methyl alcohol, ethyl alcohol, mineral oil, carbon disulfide, vacuum diffusion pump oil, and a commercial water-soluble cutting oil.[30] UC is also reported to be inert in liquid sodium at 600 to 800°C.[68]

UC does not react appreciably with Type 302B stainless steel below 1000°C,[8] and appears to be compatible with other canning materials, such as beryllium and niobium, at reactor temperatures.[68] There is a marked reaction with titanium at 1100°C.[68]

Limited data are available on the irradiation characteristics of massive, arc melted and cast UC. One irradiation has been conducted in the Battelle Research Reactor and two others in the MTR. The carbon content was slightly in excess of stoichiometric in all these specimens (about 5.0 wt % C).

1) UC with 5 wt % C was irradiated in the Battelle Research Reactor to a burnup of 500 Mwd/t at an average axial temperature of 375°C. No change in dimensions or appearance was detectable, and fission gas release was negligible, even on subsequent out-of-pile heating to 1800°F.
2) The second UC irradiation went to 1420 Mwd/t (peak) in the MTR, with an average surface temperature of 450°C and central temperature of 730°C. Some cracking occurred, and a volume increase in the order of 1% was noted.
3) The third UC irradiation in MTR went to a nominal peak burnup of 6000 Mwd/t, with an average central temperature of 870°C and a peak temperature of 982°C. Some cracking occured, and a density decrease of less than 2% was noted.

It is planned to irradiate 4.8 and 4.6 wt % C specimens in the near future. Irradiation of 5 wt % UC to a higher burnup is now in progress.

D. URANIUM-IRON

Iron and uranium form the two intermetallic compounds, U_6Fe and UFe_2. The compound U_6Fe is of interest because of the exceptionally high uranium density, but the decomposition temperature is low. U_6Fe transforms to uranium and liquid in a peritectic reaction at 815°C, as can be seen in Figure 3.[1] This compound is reported to be extremely brittle. U_6Fe is inert to nitric acid, and therefore any excess uranium can be leached out with dilute or concentrated HNO_3.[11] This material does not appear promising as a high temperature nuclear fuel, in itself, but may be useful as a dispersion in a cermet fuel.

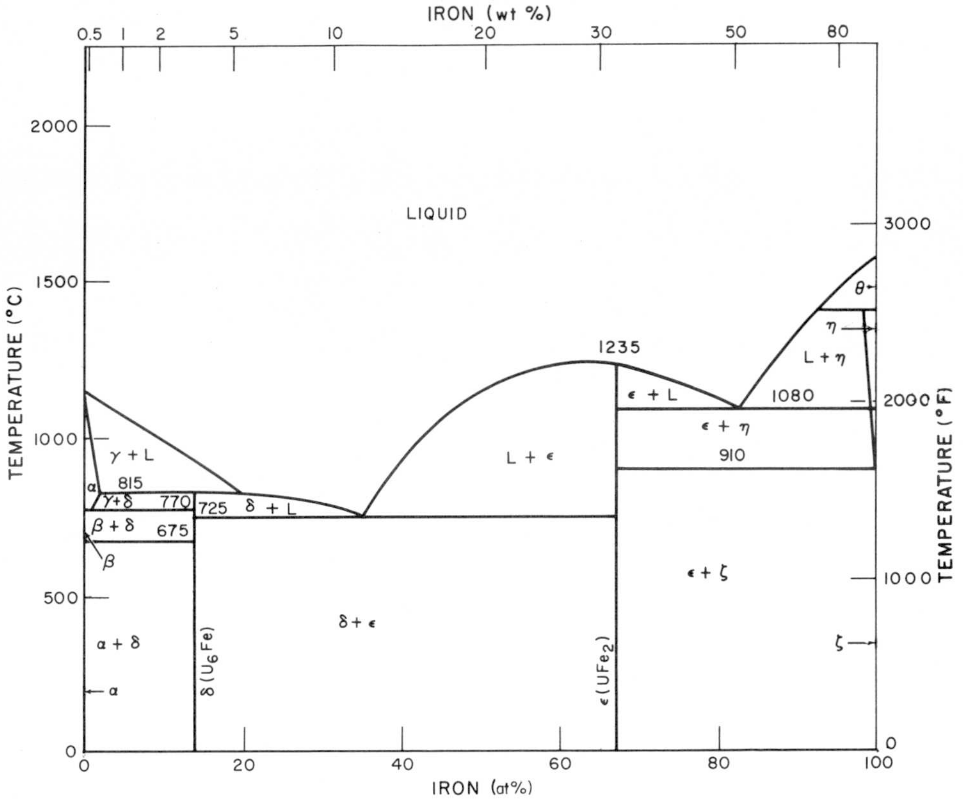

Figure 3. Uranium-Iron Phase Diagram

E. URANIUM-NITROGEN

The mononitride of uranium is under consideration as a nuclear fuel material because of its refractory nature and high uranium inventory. Very little is known of the properties and irradiation stability of this material.

In solid form, UN has a metallic luster; and, finely divided, it is a gray-black pyrophoric powder. UN is inert to hot or cold HCl, H_2SO_4, and NaOH.[11]

Uranium nitrides are generally prepared by the reaction of finely divided uranium or uranium hydride with nitrogen or ammonia. This reaction, at temperatures up to 1300°C, produces a mixture of the higher nitrides which must be subsequently reduced to UN, at a temperature greater than 1300°C.[11] Reduction can also be accomplished by reacting U_2N_3 or UN_2 with uranium metal. If the reaction temperature is increased to 1300°C, UN is produced directly; since, in this temperature region, all higher nitrides are less stable than UN[11]. Uranium nitride has been produced by arc melting uranium in a nitrogen atmosphere. The resultant product contained about 12% free uranium.[66]

F. URANIUM-PHOSPHORUS

Uranium metal reacts with phosphorus to form three compounds: UP, U_3P_4, and UP_2.[11] The monophosphide is under consideration as a nuclear fuel because it has a density comparable to that of uranium dioxide. Relatively little is known about its properties and irradiation behavior. Data on its crystallography and uranium content are given in Table VII.

Uranium phosphides have been prepared by reacting uranium powder with an excess of red phosphorus at 600 to 1000°C in an argon atmosphere. The resultant product was a sintered mass, containing a mixture of higher phosphides and uncombined phosphorus. The free phosphorus was leached with NaOH. Subsequent arc melting resulted in a button containing UP and free uranium. Decomposition of UP apparently occurs in arc melting. The sample was remelted several times, and each time there was a significant density increase.[66]

Uranium monophosphide is presently being studied, with particular regard to fabrication, physical and chemical properties, and irradiation behavior.

G. URANIUM-SILICON

The uranium-silicon system has been found to be quite complex. It is generally agreed that the compounds: U_3Si, U_3Si_2, USi, $\alpha\,USi_2$, $\beta\,USi_2$, and USi_3 exist in this system.[10,11,62] The phase diagram for the uranium-silicon system is reproduced in Figure 4. Of these compounds, U_3Si and U_3Si_2 are of interest because of the high uranium content per unit volume of compound.

As-cast U - 3.8 wt % Si is composed of the solid phases, $U + U_3Si_2$. To produce a homogeneous $\epsilon(U_3Si)$ phase, it is necessary to heat treat this product at about 800°C for several days. The density of as-cast U - 3.8 wt % Si is reported as 14.65 gm/cm^3,[1] versus 15.58 gm/cm^3 for the ϵ phase, indicating a considerable volume decrease in the transformation from the as-cast condition to the ϵ phase.

It is reported that annealing as-cast U_3Si_2 in vacuum for 8 hr at 1400°C produced a single-phase compound, identified as U_3Si_2 by X-ray diffraction.[56]

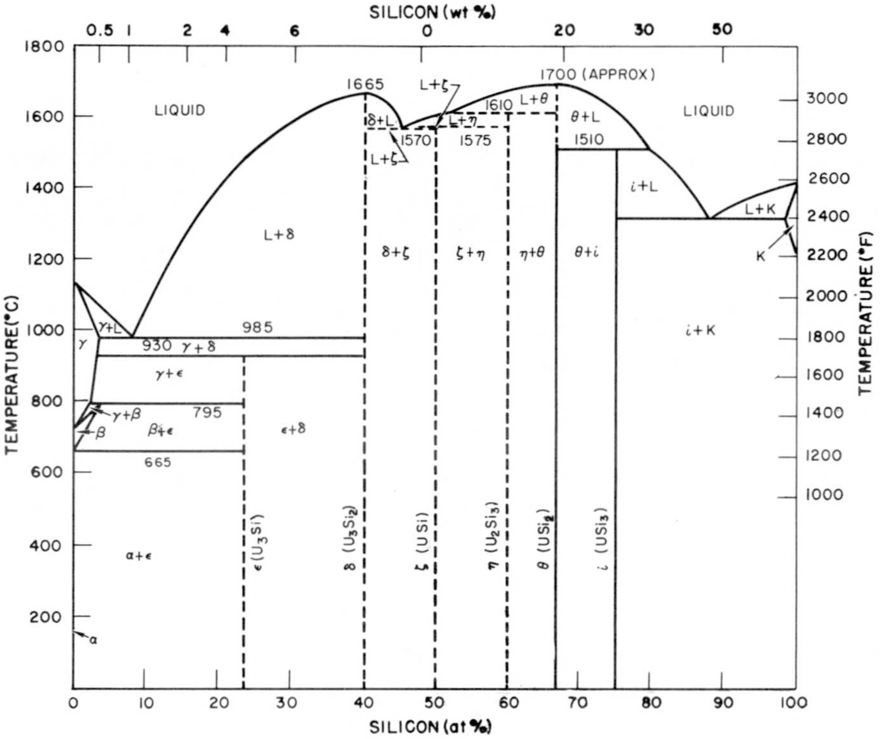

Figure 4. Uranium-Silicon Phase Diagram

It has been shown that hardness, density, and electrical resistivity are directly dependent on the amount of ϵ phase present in U - 3.8 wt % Si alloys. U_3Si has a Vickers hardness of approximately 240, while as-cast samples had a hardness of 530 VHN. The density increases and the hardness decreases with increasing ϵ phase content. Electrical resistivity of U - 3.8 wt % Si increases with temperature, regardless of ϵ phase content. However, the resistivity at a given temperature decreases with increasing ϵ phase content,[1] as shown in Table V. The coefficient of thermal expansion is given in Table VI. The average thermal expansion of U_3Si increases with temperature, while the thermal expansion of U_3Si_2 decreases with increasing temperature.

The uranium silicides have been prepared by arc melting and by vacuum induction melting in BeO crucibles and chill casting in water-cooled copper molds.[44] U_3Si is the most ductile of the silicides, and has been successfully coextruded with Zircaloy 2.[2] U_3Si_2 has been sintered to 12.0 gm/cm^3, or 98% of the theoretical density, at 1550°C.[53]

TABLE V

ELECTRICAL RESISTIVITY OF U - 3.8 wt % Si AS A FUNCTION OF EPSILON PHASE CONTENT AND TEMPERATURE

Treatment	ϵ Phase (%)	Resistivity (ohm-cm) Temperature (°C) -200	25	200	400
Heated at 800°C for several days in vacuum	95	25.97	57.67	65.36	70.58
As cast	0	42.54	63.88	75.42	84.14

TABLE VI

LINEAR EXPANSION OF U_3Si and U_3Si_2

Temperature Range (°C)	Mean Coefficient of Linear Thermal Expansion (in./in.-°C x 10^6) U_3Si*	U_3Si_2†
20 - 200	13.0	15.5
20 - 300	13.4	15.3
20 - 400	14.2	15.2
20 - 500	14.9	15.3
20 - 600	15.8	15.2
20 - 700	16.8	15.1
20 - 750	17.5	-
20 - 800	-	15.0
20 - 900	-	14.7
20 - 950		

*Heated in vacuum at 800°C for 168 hr

†U_3Si_2 powder, sintered at 1400°C in argon for 15 min[57]

Efforts to reduce the residual U and $\delta(U_3Si_2)$ phases in as-cast U - 3.8 wt % Si have shown that additives, such as antimony and bismuth, reduce the excess uranium content by forming the compounds, UBi or USb. Corrosion resistance is also improved.[22] It has been suggested that a solid solution of U_3Si and UAl_2 may stabilize U_3Si at high temperature. Other elements which do not form silicides are beryllium, titanium, and tin.[25]

Low burnup, low-temperature irradiation of U_3Si indicates that the initial damage is a decomposition of the ϵ phase or a disordering effect, resulting in a volume increase and a corresponding density decrease. Pre- and postirradiation density measurements show a maximum decrease of 4.1% at 0.104 at. % burnup with a maximum calculated central temperature of 249°C and a surface temperature of 174°C. The irradiated U_3Si samples exhibited large increases in hardness and became very brittle. There were also large increases in electrical resistivity, and the normal positive temperature coefficient of electrical resistivity reverted to an abnormal negative coefficient. The corrosion resistance to 650°F water appeared to be unchanged. The microstructure also appeared unchanged, but the irradiated material evidenced no X-ray diffraction lines. These results were tentatively explained, on the basis of a disordering reaction induced by neutron bombardment.[2] More irradiation data for U_3Si, as well as U_3Si_2, are essential to establish confidence in the irradiation behavior of these materials.

H. URANIUM-SULFUR

The uranium sulfides: US_3, U_3S_5, U_2S_3, US, and the mixed oxide-sulfide, UOS, have been identified in this system. The monosulfide is the most refractory, has the highest uranium density, and therefore is most suitable for study as a nuclear fuel.

US can be prepared by the reaction of finely divided uranium (obtained from the hydride) and the calculated amount of H_2S at 400 to 500°F. The product is then heated to 2000°C for homogenization. Another method is the interaction of UH_3 and US_2. The mixture is heated to 300°C to decompose the hydride, then to 400 to 600°C to reduce the disulfide, and finally homogenized at 2000°C.[11]

U_2S_3 is prepared by the reaction of UH_3 and H_2S, UH_3 and US_2, or the thermal decomposition of US_2 at 1600°C in vacuum.[11]

US_2 can be prepared by the reaction of UCl_4 and H_2S, or $2NaCl \cdot UCl_4$ and hydrogen charged with sulfur vapors at 500°C and above. The reaction of $2NaCl \cdot UCl_4$ with the sulfides of Na, Mg, Al, Sb, or Sn at high temperatures is reported to produce slightly impure US_2. The reduction of UO_2 or U_3O_8 with carbon, in the presence of H_2S at 1200 to 1300°C, forms UOS. This is reduced to US_2, at higher temperatures, by the action of CS_2, formed from H_2S and carbon.[11]

Uranium reacts slowly with molten sulfur at 250 to 300°C, and burns in sulfur vapor at 599°C. Depending on exact conditions, U_2S_3, US_2, or mixtures of the two are obtained.[11]

Little is known of the properties of uranium monosulfide. It is reported to be stable in boiling water, if well sintered, but slowly soluble in dilute acids. US will oxidize to UOS in air at 160°C.[11]

I. URANIUM-TITANIUM

The compound U_2Ti is the only one reported in the uranium-titanium system. This material is of interest because of the high uranium density, but the

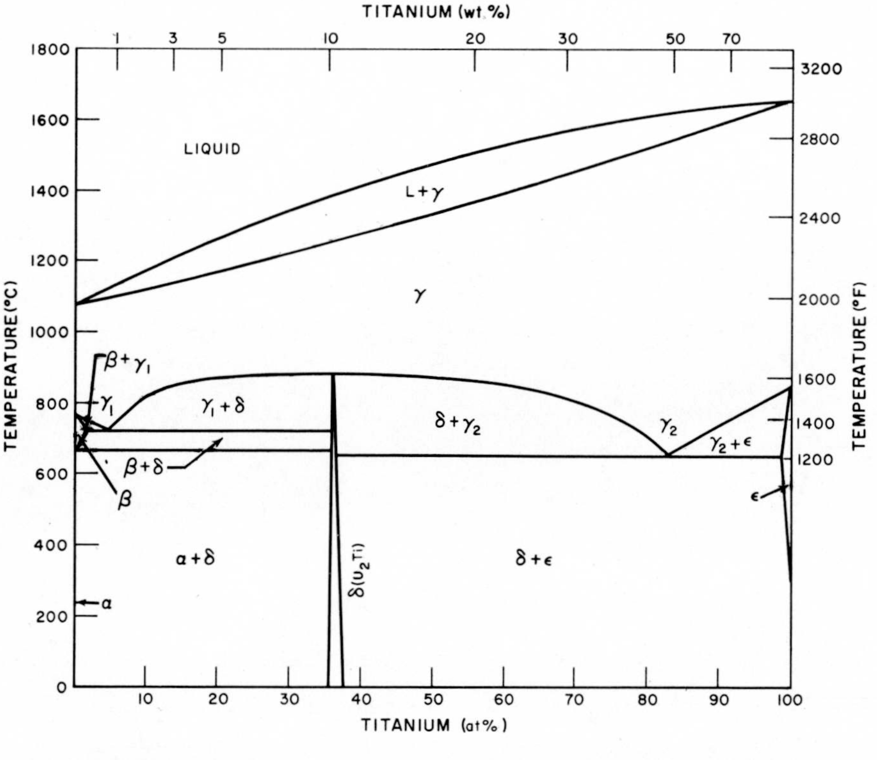

Figure 5. Uranium-Titanium Phase Diagram

high cross section of titanium, combined with a low decomposition temperature, makes U_2Ti less promising as a nuclear fuel. There is the possibility, however, that this material may be useful as a dispersion in a cermet fuel.

U_2Ti transforms to γ phase at 890°C, as shown in Figure 5. U_2Ti is reported to be quite brittle at room temperature, but relatively ductile at 700°C.[8] U_2Ti has been prepared by induction melting, followed by casting in a graphite or copper mold.[47] The compound can also be prepared by arc-melting techniques. Annealing for 24 hr at 800°C produced an improved structure.[47]

III. CERMETS

Development in the general direction of high temperature reactors brings with it much interest in ceramic materials, either alone or as cermets, in combination with metals, for use in fuel elements. Solid metallic fuels offer advantages, such as good thermal conductivity, good neutron economy, and a high resistance to thermal shock. However, coupled with these advantages are such detrimental features as phase inversions and low strength at elevated temperatures, which limit their use in reactors operating at high specific power.

Compounds of uranium, on the other hand, usually have no phase inversions, and generally exhibit good strength properties at elevated temperatures. The disadvantages of ceramic fuels, such as lower thermal conductivity and less resistance to thermal and mechanical shock, may be remedied by combining them with metals. It is for these reasons that uranium compounds, dispersed in uranium or uranium-bearing alloys, become attractive as reactor fuels requiring a high uranium inventory. Such combinations should offer a compromise of both the advantages and disadvantages of compounds and alloys of uranium.

Matrix materials under consideration are unalloyed uranium, an α phase uranium alloy, and several γ phase alloys of uranium. Also under consideration are Th-U alloys. Table VIII lists some of the nuclear and physical properties of typical matrix alloys.

Compound dispersions to be considered are those which have been discussed in preceding paragraphs. Table VII lists some of the nuclear and physical properties of these compounds.

A. DISPERSION FUELS

1. Uranium Compounds Dispersed in Unalloyed Uranium

Unalloyed uranium has the disadvantages of extensive anisotropic dimensional changes and roughening under irradiation. This anisotropy in polycrystalline uranium is minimized in material with random grain orientation. One method of producing unalloyed uranium fuel bodies with random orientation is by powder metallurgical methods. Some of the properties of unalloyed uranium are presented in Table VIII. Another problem inherent in the irradiation of unalloyed

TABLE VII

PROPERTIES AND CROSS SECTION OF URANIUM COMPOUNDS AT SEVERAL ENRICHMENTS

Compound $(UX)_n$ and Crystal Structure	UX Density (gm/cm^3)	U in UX		Enrichment (wt % U^{235})	Density			Thermal Neutron Absorption Cross Section Per Cubic Centimeter Fuel (cm^{-1})				$\frac{U^{238}}{U^{235}}$ Ratio	$\frac{Moles(U^{235})}{\Sigma_c(UX)}$ Ratio	Thermal Conductivity (cgs)	Melting Point or Transformation Temperature (°C)	Coefficient of Linear Expansion (in./in.-°C) (x 10^6)	Hardness	$\sigma_a(X)$
		$\frac{gm(U)}{cm^3(UX)}$	wt %		$\frac{gm(U^{235})}{cm^3(UX)}$	$\frac{gm(U^{238})}{cm^3(UX)}$	$\frac{gm(X)}{cm^3(UX)}$	$\Sigma_c(X)$	$\Sigma_c(U^{235})$	$\Sigma_c(U^{238})$	$\Sigma_c(UX)$							
U_3Si	15.58	14.99	96.21	1	0.1499	14.840	0.59	0.0016	0.0384	0.1036	0.1437	97.8	45	0.036	930†	17.5	-	0.13
BCT				2	0.2998	14.690			0.0769	0.1022	0.1811	48.4	70	(25°C)		(20 - 750°C)		
a = 6.029				4	0.5996	14.390			0.1537	0.1006	0.2559	23.7	100					
c = 8.697				8	1.1992	13.791			0.3074	0.0956	0.4047	11.4	126	0.041				
				3.58*	0.5366	14.453			0.1376	0.1009	0.2401	26.6	95	(50°C)				
U_6Fe	17.7	17.02	96.20	1	0.1702	16.850	0.68	0.0186	0.0436	0.1179	0.1801	97.8	40	-	815†	-	-	2.53
BCT				2	0.3404	16.680			0.0872	0.1165	0.2224	48.4	65					
a = 10.36				4	0.6808	16.339			0.1744	0.1142	0.3073	23.7	95					
c = 5.21				8	1.3616	15.658			0.3489	0.1094	0.4769	11.4	121					
				3.14*	0.5344	16.468			0.1369	0.1152	0.2707	30.5	85					
UC	13.63	12.97	95.19	1	0.1297	12.840	0.66	0.0001	0.0332	0.0896	0.1230	97.8	45	0.057	2400	10 - 11	750 - 800	0.0032
FCC				2	0.2594	12.711			0.0665	0.0888	0.1554	48.4	71	(600°C)		(20 - 1000°C)	DPH	
a = 4.961				4	0.5188	12.451			0.1330	0.0869	0.2200	23.7	100					
				8	1.0376	11.932			0.2660	0.0833	0.3494	11.4	126	0.060				
				4.13*	0.5357	12.434			0.1373	0.0867	0.2242	22.9	101	(700°C)				
UN	14.32	13.52	94.44	1	0.1352	13.385	0.80	0.0647	0.0346	0.0937	0.1931	97.8	30	-	2630	-	435	1.88
FCC				2	0.2704	13.250			0.0693	0.0926	0.2266	48.4	51				Knoop	
a = 4.88				4	0.5408	12.979			0.1385	0.0906	0.2939	23.7	78					
				8	1.0816	12.438			0.2771	0.0869	0.4288	11.4	108					
				3.96*	0.5354	12.985			0.1371	0.0909	0.2929	24.0	78					
U_3Si_2	12.20	11.31	92.70	1	0.1131	11.197	0.89	0.0025	0.0290	0.0789	0.1104	97.8	43	0.035	1650	14.6	912	0.13
TETR				2	0.2262	11.084			0.0579	0.0781	0.1385	48.4	70	(25°C)		(20 - 950°C)	DPH	
a = 7.3298				4	0.4524	10.858			0.1159	0.0758	0.1942	23.7	100					
c = 3.9003				8	0.9048	10.405			0.2318	0.0726	0.3069	11.4	126					
				4.73*	0.5350	10.775			0.1370	0.0753	0.2148	19.9	106					
U_2N_3	11.24	10.34	92.00	1	0.1034	10.237	0.90	0.0729	0.0265	0.0723	0.1717	97.8	25	-	1300†	-	-	1.88
BCC				2	0.2068	10.133			0.0530	0.0708	0.1967	48.4	45					
a = 10.678				4	0.4136	9.926			0.1060	0.0693	0.2482	23.7	71					
				8	0.8272	9.513			0.2120	0.0665	0.3514	11.4	100					
				5.18*	0.5356	9.804			0.1373	0.0685	0.2787	18.1	81					
UB_2^{11}	12.82	11.75	91.66	1	0.1175	11.632	1.07	0.0030	0.0301	0.0821	0.1152	97.8	43	-	2440	-	-	0.050
HEX				2	0.2350	11.515			0.0602	0.0799	0.1431	48.4	70					
a = 3.13				4	0.4700	11.280			0.1204	0.0788	0.2023	23.7	100					
c = 3.98				8	0.9400	10.810			0.2408	0.0755	0.3194	11.4	125					
				4.56*	0.5358	11.214			0.1373	0.0783	0.2187	20.7	105					

TABLE VII (Continued)

PROPERTIES AND CROSS SECTION OF URANIUM COMPOUNDS AT SEVERAL ENRICHMENTS

Compound $(UX)_n$ and Crystal Structure	UX Density (gm/cm^3)	U in UX		Enrichment (wt % U^{235})	Density			Thermal Neutron Absorption Cross Section Per Cubic Centimeter Fuel (cm^{-1})				$\frac{U^{238}}{U^{235}}$ Ratio	$\frac{Moles(U^{235})}{\Sigma_c(UX)}$ Ratio	Thermal Conductivity (cgs)	Melting Point or Transformation Temperature (°C)	Coefficient of Linear Expansion (in./in.-°C) (x 10^6)	Hardness	$\sigma_a(X)$
		$\frac{gm(U)}{cm^3(UX)}$	wt %		$\frac{gm(U^{235})}{cm^3(UX)}$	$\frac{gm(U^{238})}{cm^3(UX)}$	$\frac{gm(X)}{cm^3(UX)}$	$\Sigma_c(X)$	$\Sigma_c(U^{235})$	$\Sigma_c(U^{238})$	$\Sigma_c(UX)$							
U_2Ti	15.22	13.80	90.80	1	0.1380	13.662	1.42	0.0996	0.0354	0.0953	0.2304	97.8	25	-	890†	-	-	5.6
HEX				2	0.2760	13.524			0.0707	0.0944	0.2649	48.4	45					
a = 4.828				4	0.5520	13.248			0.1414	0.0926	0.3338	23.7	70					
c = 2.847				8	1.1040	12.696			0.2828	0.0886	0.4712	11.4	100					
				3.88*	0.5354	13.265			0.1372	0.0928	0.3297	24.5	70					
USi	10.40	9.30	89.44	1	0.0930	9.207	1.00	0.0028	0.0239	0.0644	0.0910	97.7	43	-	1600	-	-	0.13
ORTHO				2	0.1860	9.114			0.0477	0.0637	0.1141	48.4	68					
a = 5.66				4	0.3720	8.928			0.0954	0.0623	0.1605	23.7	98					
b = 7.67				8	0.7440	8.556			0.1908	0.0597	0.2533	11.4	125					
c = 3.91				5.77*	0.5366	8.763			0.1376	0.0613	0.2017	16.1	113					
US	10.87	9.58	88.12	1	0.0958	9.484	1.29	0.0119	0.0245	0.0663	0.1026	97.8	40	-	>2000	-	-	0.490
BCC				2	0.1916	9.388			0.490	0.0653	0.1263	48.4	65					
a = 5.484				4	0.3832	9.197			0.0981	0.0641	0.1741	23.7	93					
				8	0.7680	8.814			0.1961	0.0615	0.2695	11.4	121					
				5.6*	0.5365	9.044			0.1373	0.0632	0.2124	16.7	108					
UO_2	10.96	9.60	88.10	1	0.0960	9.504	1.36	0.0000	0.0246	0.0663	0.0909	97.8	45	0.006	2850	7.7	-	<0.0002
CUBIC				2	0.1920	9.408			0.0491	0.0655	0.1147	48.4	71	(600°C)		(600°C)		
a = 5.4691				4	0.3840	9.216			0.0983	0.0643	0.1627	23.7	100					
				8	0.7680	8.832			0.1966	0.0617	0.2583	11.4	126					
				5.59*	0.5366	9.063			0.1374	0.0634	0.2008	16.7	113					
UBe_{13}	4.37	3.06	67.00	1	0.0306	3.029	1.31	0.0009	0.0078	0.0211	0.0299	97.8	43	0.028	2025	-	-	0.01
FCC				2	0.0612	2.999			0.157	0.0210	0.0375	48.4	70					
a = 10.2568				4	0.1224	2.938			0.0313	0.0204	0.0526	23.7	100					
				8	0.2448	2.815			0.0626	0.0196	0.0832	11.4	125					
				17.5*	0.5358	2.524			0.1370	0.0176	0.1556	4.7	146					
UAl_2	8.14	6.63	81.52	1	0.0662	6.550	1.52	0.0078	0.0169	0.0457	0.0704	97.8	40	-	1590	16.1	-	0.230
FCC				2	0.1324	6.488			0.0338	0.0454	0.0870	48.4	65			on heating		
a = 7.811				4	0.2648	6.355			0.0677	0.0444	0.1199	23.7	95			(20 - 900°C)		
				8	0.5296	6.090			0.1354	0.0426	0.1858	11.4	121			17.1		
				8.1*	0.5360	6.084			0.1371	0.0426	0.1875	11.2	121			on cooling		
UP	10.23	9.06	88.50											-	-			0.190
FCC																		
a = 5.589																		

BCT - Body centered tetragonal
FCC - Face centered cubic
TETR - Tetragonal
BCC - Body centered cubic

HEX - Hexagonal
ORTHO - Orthorhombic
DPH - Diamond Pyramid Hardness
* - Enrichment required to match unalloyed uranium loading
† - Transformation temperature

TABLE VIII

SOME PROPERTIES OF URANIUM AND URANIUM-BASE ALLOYS

Alloy (UX) and Crystal Structure	UX Density (gm/cm^3)	U in X		Enrichment (wt % U^{235})	Density			Thermal Neutron Absorption Cross Section Per Cubic Centimeter Fuel (cm^{-1})				$\frac{U^{238}}{U^{235}}$ Ratio	$\frac{\text{Moles}(U^{235})}{\Sigma_c(UX)}$ Ratio	Thermal Conductivity (cgs)	Transformation and Melting Point (°C)
		$\frac{gm(U)}{cm^3(UX)}$	wt %		$\frac{gm(U^{235})}{cm^3(UX)}$	$\frac{gm(U^{238})}{cm^3(UX)}$	$\frac{gm(X)}{cm^3(UX)}$	$\Sigma_c(X)$	$\Sigma_c(U^{235})$	$\Sigma_c(U^{238})$	$\Sigma_c(UX)$				
U	19.13	19.13	100.00	1	0.1913	18.939	-	-	0.0490	0.1323	0.1814	97.8	45	0.071	660
BCC				2	0.3826	18.747	-	-	0.0981	0.1310	0.2291	48.4	71	(70°C)	762
a = 3.524				4	0.7652	18.365	-	-	0.1961	0.1283	0.3245	23.7	100	0.078	1133
				8	1.5304	17.600	-	-	0.3922	0.1229	0.5151	11.4	126	(400°C)	
				2.8*	0.5356	18.594	-	-	0.1373	0.1301	0.2674	34.3	85		
U - 10 wt % Mo	17.20	15.48	90.00	1	0.155	15.325	1.72	0.0270	0.0398	0.1070	0.1738	97.8	38	0.059	575
BCC				2	0.310	15.170	1.72		0.0795	0.1059	0.2123	48.4	61	(500°C)	600
				4	0.620	14.861	1.72		0.1589	0.1037	0.2896	23.7	91		1160
				8	1.240	14.242	1.72		0.3178	0.0994	0.4942	11.4	118		
				3.45*	0.535	14.945	1.72		0.1371	0.1044	0.2685	27.6	85		metastable
U - 60 wt % Nb	12.69	5.07	39.95	1	0.0507	5.02	7.62	0.0494	0.0130	0.0351	0.0975	97.8	22	-	
BCC				2	0.1014	4.97	7.62		0.0260	0.0348	0.1102	48.4	40		1800
				4	0.2028	4.87	7.62		0.0520	0.0341	0.1355	23.7	63		
				8	0.4056	4.66	7.62		0.1041	0.0328	0.1862	11.4	93		
				10.55*	0.5349	4.54	7.62		0.1373	0.0317	0.2184	8.4	105		stable
U - 20 wt % Nb - 10 wt % Zr	15.71	11.00	70.02	1	0.1100	10.8900	Nb-3.14	Nb-0.0204	0.0282	0.0761	0.1266	97.8	37	-	
				2	0.2200	10.7800	Zr-1.57	Zr-0.0019	0.0564	0.0753	0.1539	48.4	61		
				4	0.4400	10.5600			0.1128	0.0738	0.2088	23.7	90		
				8	0.8800	10.1200			0.2255	0.0706	0.3184	11.4	118		
				4.87*	0.5356	10.4644			0.1373	0.0731	0.2326	19.3	95		metastable
U - 20 wt % Nb - 5 wt % Zr	16.03	12.02	74.98	1	0.1202	11.900	Nb-3.21	Nb-0.0208	0.0308	0.0831	0.1356	97.8	38	-	
				2	0.2404	11.780	Zr-0.80	Zr-0.0010	0.0615	0.0823	0.1655	48.4	61		
				4	0.4808	11.540			0.1230	0.0806	0.2255	23.7	91		
				8	0.9616	11.060			0.2460	0.0773	0.3452	11.4	118		
				4.46*	0.5356	11.484			0.1373	0.0803	0.2393	21.2	95		metastable

BCC - Body centered cubic
* - Enrichment required to match unalloyed uranium loading for SRE

uranium is swelling and distortion, due to the production of fission gases. This swelling and distortion can be alleviated to a degree, and additional high temperature strength added, through the incorporation of a second fissile phase which is capable of retaining the fission gases produced during irradiation.

The dispersed fissile phase will constitute from 20 to 40 vol % of the cermet, and the balance will consist of unalloyed uranium. It is important that the dispersed particles are not too fine, with respect to the metallic matrix. If so, they may form a continuous phase which might impair the mechanical properties of the mixture and have undesirable effects on the distribution of fission gases produced during irradiation.

A uranium-uranium compound cermet has the added advantage, in that two different enrichments can be utilized within the same fuel body. This provides a means for extending the irradiation life of a fuel body containing unalloyed uranium. Longer burnups can be attained if the compound dispersion contains enriched uranium and the matrix consists of depleted uranium. When the unalloyed uranium matrix of such a cermet has reached 1000 Mwd/t average burnup, the dispersed fissile phase will have reached a much higher burnup, giving an average burnup of the cermet considerably in excess of the limiting value of 1000 Mwd/t currently expected with unalloyed uranium.

Table IX lists some of the salient properties of a 25 vol % dispersion of UO_2 in unalloyed uranium. By comparing the uranium density of such a dispersion with some of the alloys listed in Table VIII, it can be seen that the dispersion has a greater uranium inventory and a higher ratio of available U^{235} to parasitic absorption cross section. The ratio of U^{238} to U^{235} is also considerably higher, indicating a higher possible conversion ratio.

Equivalent properties for a 25 vol % dispersions of UO_2 and UC in unalloyed uranium, and for 30 vol % dispersions of UO_2 and UC in unalloyed uranium are presented in Tables IX, X, XI, and XII. Similar advantages can be noted for these dispersion materials.

2. Uranium Compounds Dispersed in Alpha Phase Uranium Alloys

U - 5 wt % Zr - 1.5 wt % Nb is an α phase alloy with low alloying content, and consequently with a high uranium inventory, which should be ideally suited as a cermet matrix. Powder metallurgy combined with suitable heat treatment

TABLE IX

SALIENT PROPERTIES OF CERMETS CONTAINING A 25 vol % DISPERSION OF UO_2 IN A 75 vol % URANIUM ALLOY MATRIX

(U^{235} CONSTANT AT 0.5356 gm/cm^3)

Matrix	Density		U in C (wt %) UO_2 in C (wt %)	Thermal Neutron Absorption Cross Section $\Sigma_c(C)$ (cm^{-1})	$\frac{U^{238}}{U^{235}}$ Ratio	$\frac{\text{Moles } U^{235}}{\Sigma_c(C)}$ Ratio	Matrix Melting Point (°C)
	Cermet (C) (gm/cm^3)	Uranium (U) (gm/cm^3)					
U	17.08	16.74	98.01 16.0	0.2505	29.9	91	1133
U - 10 wt % Mo	15.64	14.00	89.51 17.5	0.2515	24.8	91	~1160
U - 60 wt % Nb	12.25	6.20	50.61 22.4	0.2139	10.4	107	~1800
U - 20 wt % Nb - 10 wt % Zr	14.53	10.65	73.30 18.8	0.2246	18.6	101	-
U - 20 wt % Nb - 5 wt % Zr	14.77	11.40	77.18 18.5	0.2294	20.0	99	-

TABLE X

SALIENT PROPERTIES OF CERMETS CONTAINING A 25 vol % DISPERSION OF UC IN A 75 vol % URANIUM ALLOY MATRIX

(U^{235} CONSTANT AT 0.5356 gm/cm^3)

Matrix	Density		U in C (wt %) UC in C (wt %)	Thermal Neutron Absorption Cross Section $\Sigma_c(C)$ (cm^{-1})	$\frac{U^{238}}{U^{235}}$ Ratio	$\frac{\text{Moles } U^{235}}{\Sigma_c(C)}$ Ratio	Matrix Melting Point (°C)
	Cermet (C) (gm/cm^3)	Uranium (U) (gm/cm^3)					
U	17.75	17.58	99.04 19.2	0.2563	31.4	89	1133
U - 10 wt % Mo	16.31	14.85	91.05 20.9	0.2574	26.4	85	~1160
U - 60 wt % Nb	12.92	7.04	54.49 26.4	0.2197	12.0	104	~1800
U - 20 wt % Nb - 10 wt % Zr	15.20	11.49	75.59 22.4	0.2305	20.2	99	-
U - 20 wt % Nb - 5 wt % Zr	15.44	12.24	79.27 22.1	0.2354	21.6	97	-

TABLE XI

SALIENT PROPERTIES OF CERMETS CONTAINING A 30 vol % DISPERSION OF UO_2 IN A 70 vol % URANIUM ALLOY MATRIX (U^{235} CONSTANT AT 0.5356 gm/cm^3)

Matrix	Density		U in C (wt %) UO_2 in C (wt %)	Thermal Neutron Absorption Cross Section $\Sigma_c(C)$ (cm^{-1})	$\frac{U^{238}}{U^{235}}$ Ratio	$\frac{\text{Moles } U^{235}}{\Sigma_c(C)}$ Ratio	Matrix Melting Point (°C)
	Cermet (C) (gm/cm^3)	Uranium (U) (gm/cm^3)					
U	16.68	16.27	97.54 19.7	0.2471	29.0	92	1133
U - 10 wt % Mo	15.33	13.72	89.50	0.2483	24.3	92	~1160
U - 60 wt % Nb	12.17	6.43	52.83 27.0	0.2131	10.9	107	~1800
U - 20 wt % Nb - 10 wt % Zr	14.29	10.58	74.04 23.0	0.2231	18.5	102	-
U - 20 wt % Nb - 5 wt % Zr	14.51	11.59	77.80 22.6	0.2276	19.8	100	-

TABLE XII

SALIENT PROPERTIES OF CERMETS CONTAINING A 30 vol % DISPERSION OF UC IN A 70 vol % URANIUM ALLOY MATRIX

(U^{235} CONSTANT AT 0.5356 gm/cm^3)

Matrix	Density		U in C (wt %) UC in C (wt %)	Thermal Neutron Absorption Cross Section Σ_c (C) (cm^{-1})	$\frac{U^{238}}{U^{235}}$ Ratio	$\frac{\text{Moles } U^{235}}{\Sigma_c(C)}$ Ratio	Matrix Melting Point (°C)
	Cermet (C) (gm/cm^3)	Uranium (U) (gm/cm^3)					
U	17.48	17.28	98.86 23.4	0.2544	30.9	90	1133
U - 10 wt % Mo	16.13	14.73	91.32 25.3	0.2554	26.2	89	~1160
U - 60 wt % Nb	12.97	7.44	57.36 31.5	0.2201	12.7	104	~1800
U - 20 wt % Nb - 10 wt % Zr	15.09	11.59	76.81 27.1	0.2300	20.4	99	-
U - 20 wt % Nb - 5 wt % Zr	15.31	12.30	80.34 26.6	0.2346	21.7	97	-

and/or fabricating techniques should produce a fine-grained, randomly oriented structure. The alloy exhibits better irradiation stability than unalloyed uranium.[70]

3. Uranium Compounds Dispersed in Gamma Phase Uranium Alloys

An alloy of uranium containing 60 wt % niobium is a γ solid solution, up to approximately 1900°C (Figure 6). Such an alloy exhibits no allotropic transformations; and, were it not for its low uranium density, should present itself as a likely fuel for high-temperature reactor operations. As a cermet matrix, however, the uranium density limitation should not prove of cardinal importance. A 30 vol % dispersion of UC in a U - 60 wt % Nb alloy matrix exhibits a considerably higher $\left[\text{Moles } (U^{235})/\Sigma_c(C)\right]$ ratio than the $\left[\text{Moles}(U^{235})/\Sigma_c(UX)\right]$ ratio for a U -10 wt % Mo alloy (comparison of Table XII with Table VIII). This is assuming than equivalent U^{235} inventory is required in both cases. Comparison of the ratios of U^{238} to U^{235}, however, indicates a lower conversion ratio.

In U-Nb alloys containing more than 58 wt % Nb, the γ phase is stable to room temperature. There are other alloys, such as U - 10 wt % Mo (see Figures 7 and 8), U - 20 wt % Nb - 10 wt % Zr, U - 20 wt % Nb - 5 wt % Zr, U - 10 wt % Nb - 6 wt % Zr, and many others, that are metastable γ solid solutions.[71] The transformation kinetics of these systems indicate stability of the γ phase at lower temperatures, and therefore they become possible fuels for high-temperature reactors.

As alloy matrices for uranium compound dispersions, the metastable γ alloys appear promising. Distortion and growth due to allotropic transformations are greatly reduced; and swelling due to fission gas accumulation should also be reduced, due to the ability of the uranium compound dispersions to retain fission gas without appreciable swelling. The γ phase (body centered cubic structure) can be retained at room temperature, and persists during irradiation as body centered cubic metal, thereby permitting higher central metal temperatures without swelling than with orthorhombic uranium.

4. Uranium Compounds Dispersed in Thorium or Thorium-Uranium Alloys

Dimensional changes in alloys ranging up to 11 wt % U^{235} in Th are very slight, even at total atom burnup as high as 1.1% at 1100°F central temperature.[72] These results are not surprising, since thorium and high thorium alloys are cubic.

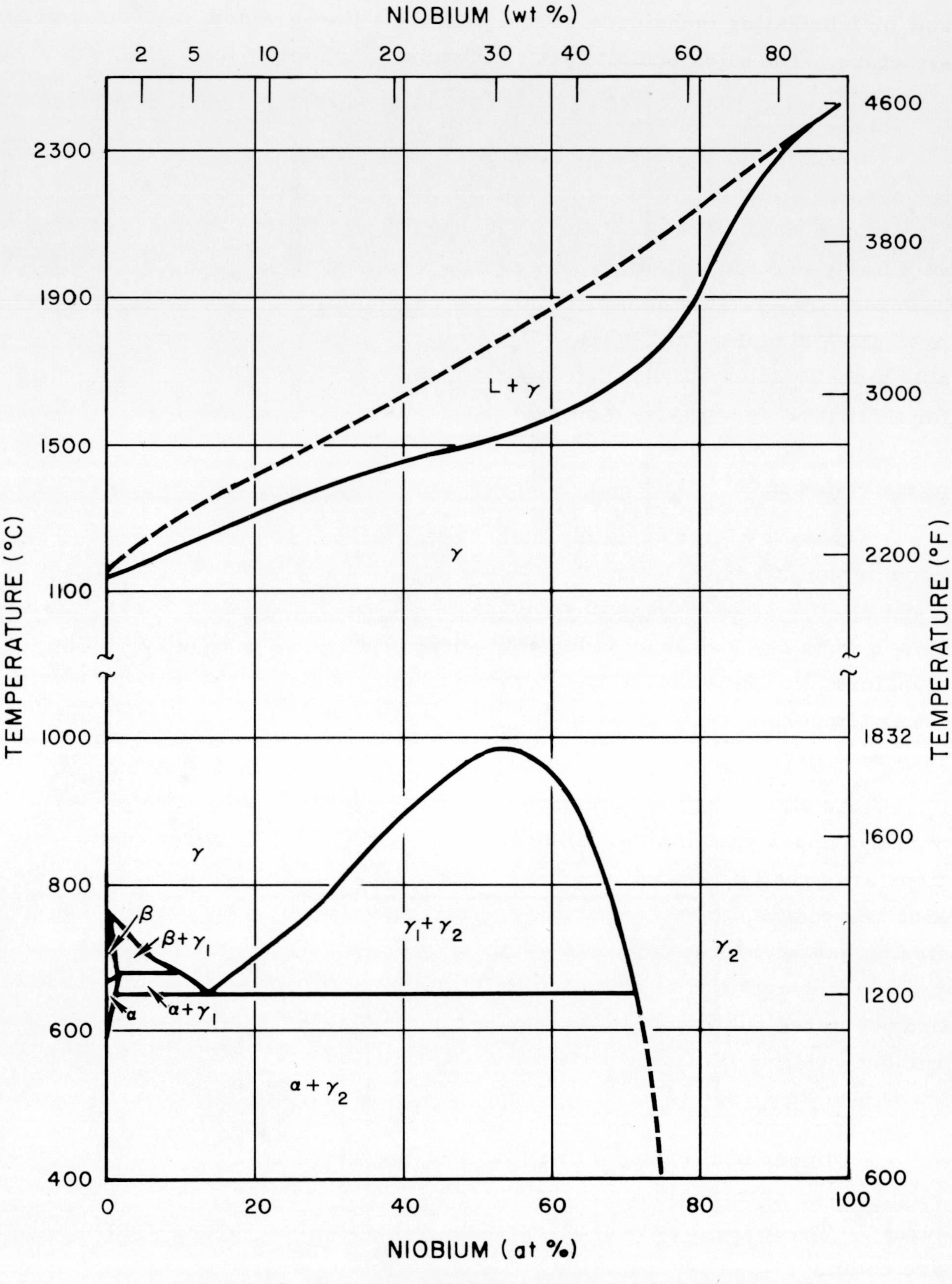

Figure 6. Uranium-Niobium Phase Diagram

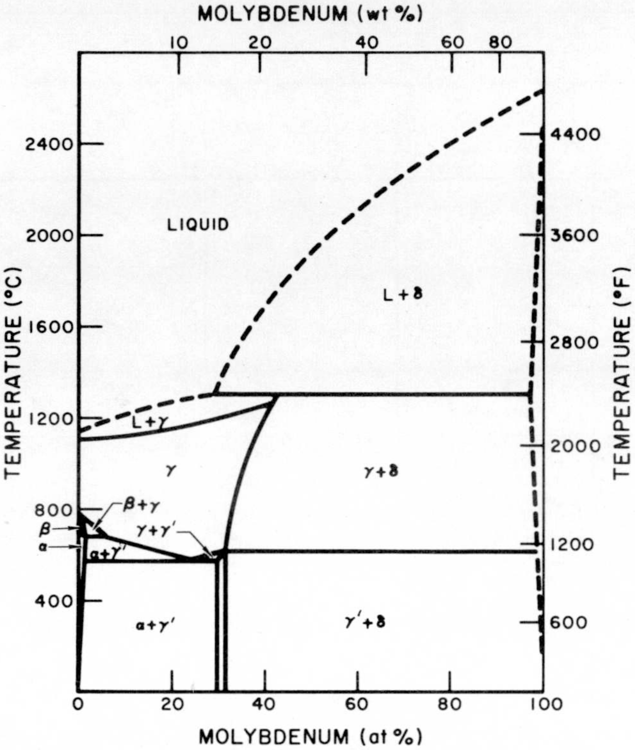

Figure 7. Uranium-Molybdenum Phase Diagram

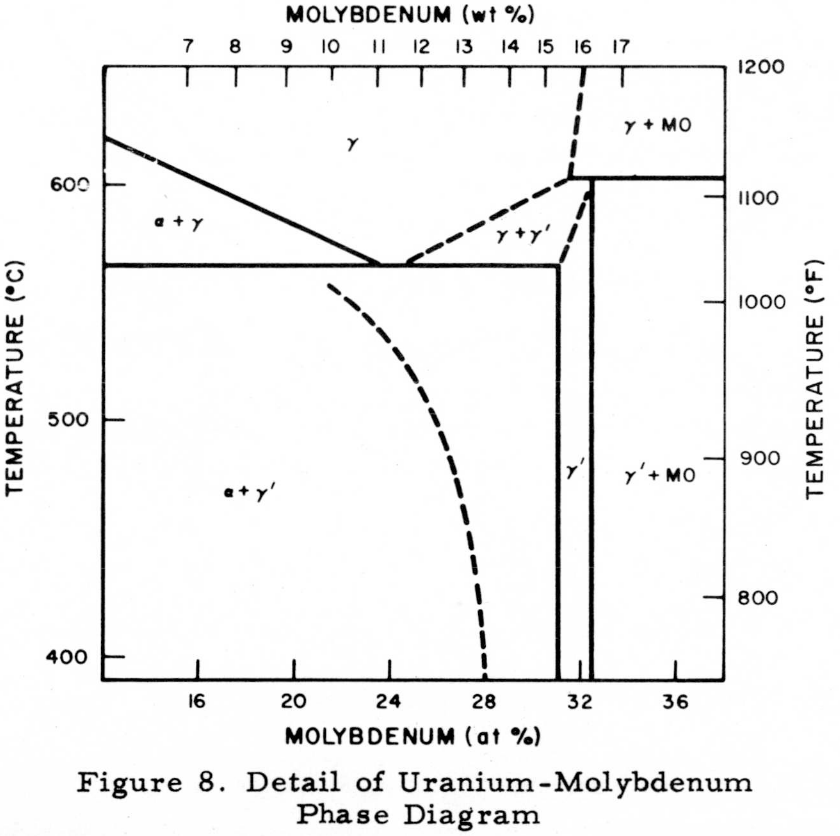

Figure 8. Detail of Uranium-Molybdenum Phase Diagram

The fabrication of thorium by cold compacting and sintering is possible. Thorium-uranium alloys produced by powder metallurgy techniques, and containing dispersions of uranium compounds, should produce fuels with high irradiation stability and high conversion ratios, and need not require enrichments in excess of 20%.

5. Thoria-Urania Solid Solutions Dispersed in Fissile Matrices

Solid solutions of UO_2 and ThO_2 in powdered form, dispersed in a matrix of a stable uranium alloy, such as U - 60 wt % Nb or a metastable uranium alloy (U - 10 wt % Nb), should prove interesting. Another possible matrix is BeO, which would combine fuel and moderator in a single unit.

B. METHODS OF CALCULATION

The thermal neutron parasitic absorption cross section and the burnup of a UC dispersion in U were calculated by the methods described below.

The content of U^{235} was, in all cases, predicated on the uranium loading of the SRE, which had a U^{235} content of approximately 0.5356 gm/cm^3 of fuel. It should be noted that maintaining the same total fission cross section (Σ_f) is a rough approximation for maintaining reactivity, but the U^{238} content and matrix cross section will have some influence on reactivity.

1. Thermal Neutron Parasitic Absorption Cross Section

$$\Sigma_c = N_1\sigma_1 + N_2\sigma_2 + \ldots + N_i\sigma_i + \ldots$$

and

$$N_i = \frac{\rho_i}{A_i}N_a$$

where:

N_i = Nuclei/cm^3,

N_a = Avogadro's Number,

A = Atomic weight,

ρ_i = Density,

σ_i = microscopic thermal neutron parasitic absorption cross section of the ith constituent.

Therefore,

$$\Sigma_c = \frac{\rho_1}{A_1} N_a \sigma_1 + \frac{\rho_2}{A_2} N_a \sigma_2 + \ldots + \frac{\rho_i}{A_i} N_a \sigma_i + \ldots \quad .$$

$$\Sigma_c(\mathrm{UC}) = \left[\frac{\rho_{U^{238}}}{238}\right]\left[N_a\right]\left[\sigma_a^{U^{238}}\right] + \left[\frac{\rho_{U^{235}}}{235}\right]\left[N_a\right]\left[\sigma_a^{U^{235}}\right] + \left[\frac{\rho_C}{12.01}\right]\left[N_a\right]\left[\sigma_a^{C}\right]$$

where:

ρ_{UC} = density of UC (Table VII, Col. 2),

ρ_U = partial density of UC attributable to its uranium content (Table VII, Col. 3),

$\rho_{U^{235}}$ = partial density of UC attributable to its uranium-235 content (Table VII, Col. 6),

$\rho_{U^{238}}$ = partial density of UC attributable to its uranium-238 content (Table VII, Col 7),

$\rho_C = \rho_{UC} - \rho_U$ = partial density of UC attributable to its carbon content (Table VII, Col. 8) .

Therefore, for UC of theoretical density, with an enrichment of 4.13%, which provides the predicated 0.5356 gm/cm^3 of U^{235} in UC, the following calculation applies:

$$\begin{aligned} \Sigma_c(\mathrm{UC}) &= \left[\frac{12.434}{238}\right]\left[6.02 \times 10^{23}\right]\left[2.76 \times 10^{-24}\right] \\ &+ \left[\frac{0.5356}{235}\right]\left[6.02 \times 10^{23}\right]\left[100 \times 10^{-24}\right] \\ &+ \left[\frac{0.66}{12.01}\right]\left[6.02 \times 10^{23}\right]\left[0.0032 \times 10^{-24}\right] \\ {}^{*} &= 0.0867 + 0.1373 + 0.0001 = 0.2241 \text{ cm}^{-1} \quad . \end{aligned}$$

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*Values of $N_i \sigma_i$ are tabulated in Table VII, columns 9, 10, and 11, for uranium compounds; and in Table VIII, columns 9, 10, and 11, for some uranium alloys.

Column 12 of Tables VII and VIII indicates the thermal neutron parasitic absorption cross section of some uranium compounds and alloys, as a function of enrichment in U^{235}. Tables IX, X, XI, and XII have similar listings. This parameter is one of the factors which might be used as a figure of merit, in determining the superiority of one fuel over another.

2. Estimated Burnup of Fissile-Matrix, Fissile-Dispersion Cermet

Burnup of a fissile-matrix, fissile-dispersion cermet consists of the summation of the burnups attained within each constituent phase.

$$\text{Burnup (Cermet)} = \text{Burnup (U matrix)} + \text{Burnup} (U_{ux}, {}_{uy}, {}_{etc}).$$

a. Case 1: U-UC cermet (Matrix normally enriched)

As a first approximation, it can be seen, from Figure 10, that with a U matrix (75 vol %) of normal enrichment (0.7 wt % U^{235}), an enrichment of 13.4 wt % U^{235} in U of the UC (25 vol %) would be required to match the 0.5356 gm/cm^3 of U^{235} in the uranium loading of the SRE.

Since

$$\text{Burnup (Cermet)} = \text{Burnup (U)} + \text{Burnup (UC)}$$

and

$$\text{Burnup} \approx (E)(\text{wt \% U}), \text{ for both the dispersed and matrix phases,}$$

where:

wt % U in the U matrix = 81.6,
wt % U in the UC dispersion = 18.4,
E = enrichment.

Thus

$$\text{Burnup (U)} \approx (0.7)(81.6)$$
$$\approx 57.1,$$

$$\text{Burnup (UC)} \approx (13.4)(18.4)$$
$$\approx 247,$$

$$\text{Burnup(UC)} : \text{Burnup (U)} \approx 4 : 1 .$$

Limiting the burnup in the U matrix to 1000 Mwd/t, the total burnup becomes

$(1000)(0.816) + (4)(1000)(0.184) \approx 1550$ Mwd, per long ton of contained uranium in the U-UC cermet.

b. Case 2: U-UC cermet (matrix consisting of depleted uranium with U^{235} content at 0.4 wt %)

$$\text{Burnup (U)} \approx (0.4)(81.6)$$
$$\approx 32.6,$$
$$\text{Burnup (UC)} \approx (14.7)(18.4)$$
$$\approx 270,$$
$$\text{Burnup (UC)} : \text{Burnup (U)} \approx 8.3:1.$$

Limiting the burnup in the U matrix to 1000 Mwd/t, the total burnup then becomes

$(1000)(0.816) + (4)(1000)(0.184) \approx 1550$ Mwd, per long ton of contained uranium in the U-UC cermet.

The burnup possible for a fuel consisting of UC dispersed in unalloyed uranium is greater than that for conventional unalloyed uranium fuel. The conversion ratio of U^{238} to PU^{239}, and the thermal conductivity of this type of cermet; remains essentially the same as that of unalloyed uranium. The greater the depletion of the matrix uranium, the greater is the burnup attainable in the cermet. In a cermet with 0.4% U^{235}, burnup is limited by radiation damage in the matrix, due to U^{235} fission. With lower U^{235} content in the matrix, burnup would be limited by fission of converted PU^{239}.

3. Comparison of Enrichments of Cermet Constituents

Illustrated in Figure 9 is a plot of U^{235} density vs the required enrichments in U^{235} fuels, such as unalloyed uranium, uranium alloys, and uranium compounds.

Figure 10 compares the U^{235} density of compound and alloy constituents for cermets consisting of 75 vol % uranium alloy and 75 vol % uranium compound, as a function of enrichment. The desired enrichment of the matrix is located on the ordinate and projected to the intersection of the selected matrix. The U^{235} content is obtained from the abscissa. The difference between this value and the required U^{235} inventory of the fuel is located on the abscissa and projected to the

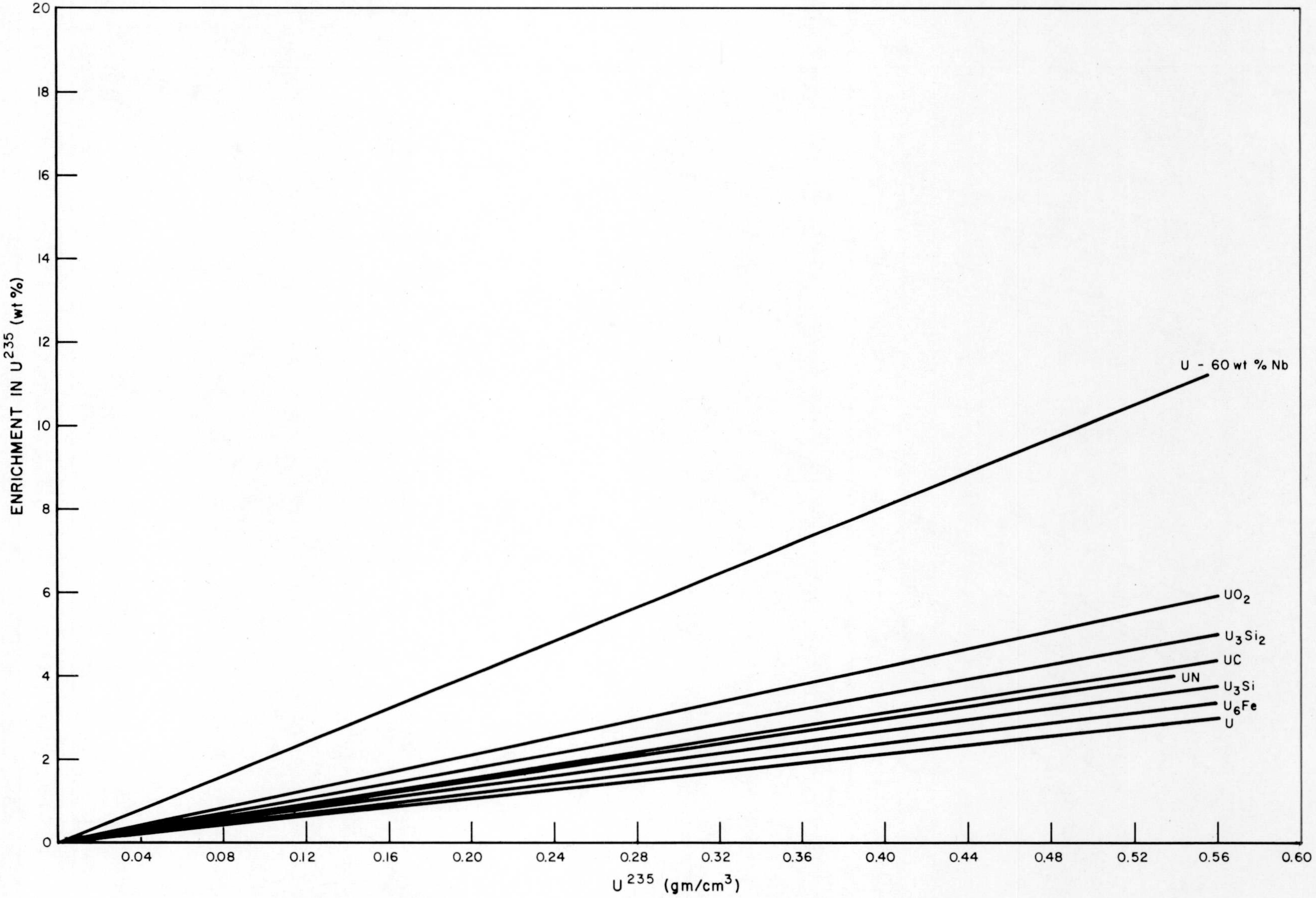

Figure 9. U^{235} Content vs Enrichment of Compounds and Alloys

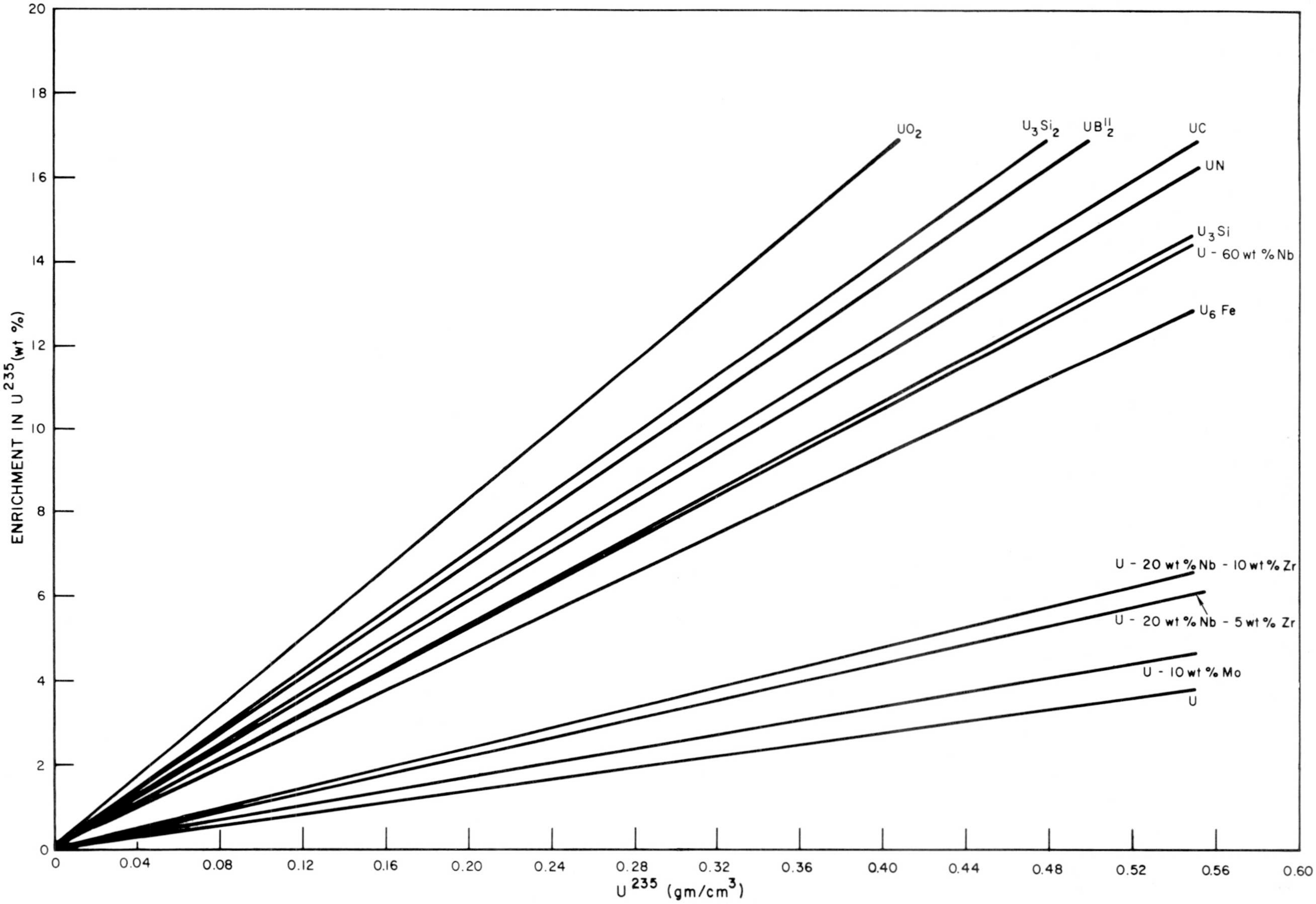

Figure 10. U^{235} Content vs Enrichment of Compound and Alloy Constituents for Cermets ($\overline{75}$ vol % U Alloy, 25 vol % U Compound)

selected compound line. The ordinate value at the intercept yields the required enrichment of the dispersant.

A similar chart for a 70 vol % matrix, 30 vol % dispersion fuel is presented in Figure 11.

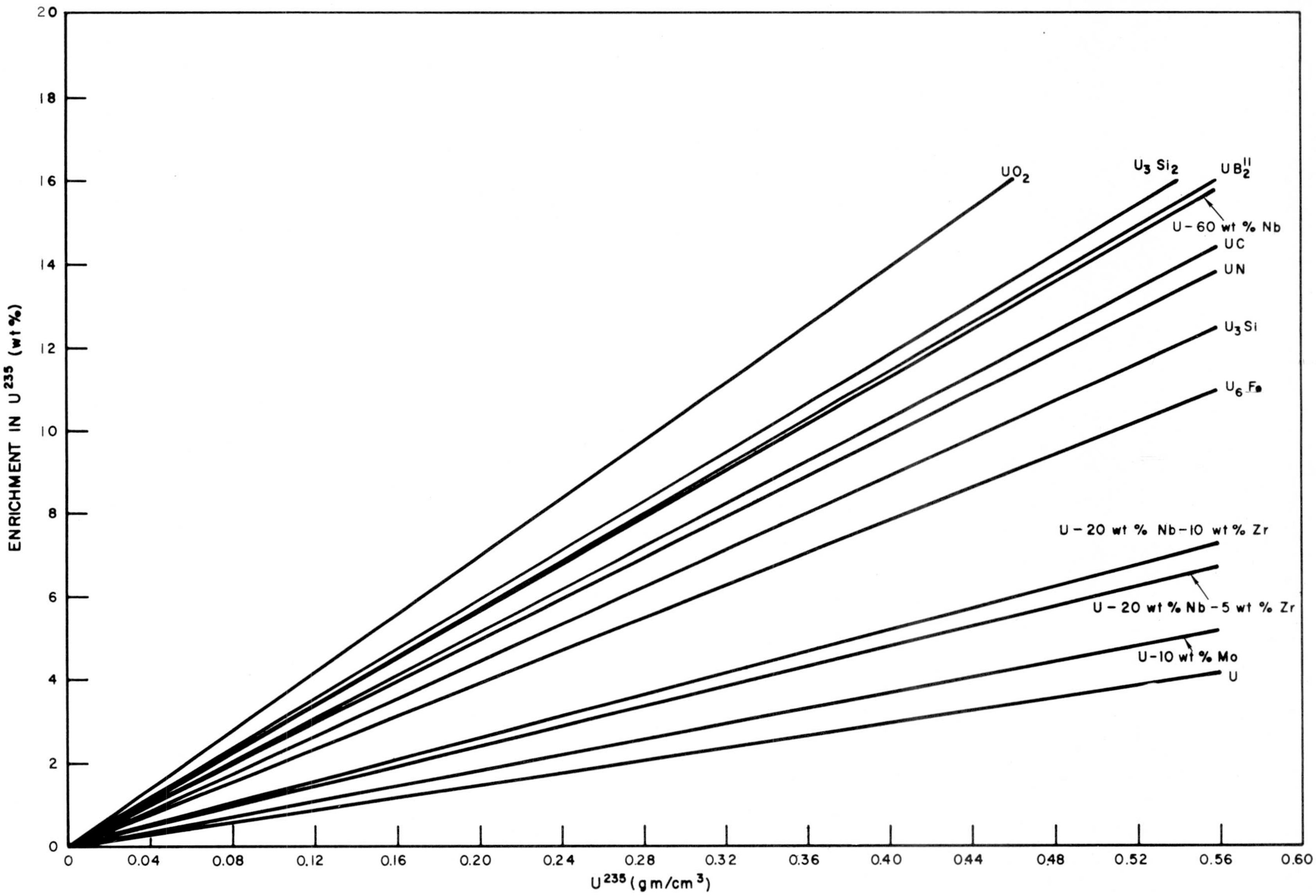

Figure 11. U^{235} Content vs Enrichment of Compound and Alloy Constituents for Cermets ($\overline{70}$ vol % U Alloy, 30 vol % U Compound)

IV. SUMMARY

Further study of binary uranium compounds is necessary, to supplement the limited data currently available. Uranium monocarbide has received appreciable attention, and is being critically evaluated as a fuel material for sodium graphite reactors. MTR irradiation data are encouraging. Triuranium silicide has also been studied extensively, in relation to pressurized water reactors, but not in the light of SGR requirements. Certain other compounds have been the subject of superficial evaluation, and may be considered as potential SGR fuels.

Of the possible fuel materials, the monocarbide, mononitride, silicides, boride, phosphide, sulphide, and aluminide of uranium appear most promising, on the basis of uranium content, melting point, and thermal neutron absorption cross section. A program is now in progress to evaluate these materials as nuclear fuels, with emphasis on chemical and physical properties, as well as irradiation behavior. Economic considerations, such as material costs and availability, fuel cycle costs, and fabrication costs, will be evaluated.

Promising compounds are being looked at as dispersions in gamma-stable and gamma-metastable fissile alloys. Cermets composed of these materials may enable us to minimize the undesirable effects of each, and thereby accomplish what may not be possible with either material alone.

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