

Research reactor core conversion guidebook

Volume 4: Fuels (Appendices I-K)



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APPENDIX I-1.1

SELECTED THERMAL PROPERTIES AND URANIUM DENSITY RELATIONS FOR ALLOY, ALUMINIDE, OXIDE, AND SILICIDE FUELS

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ABSTRACT

This appendix presents data on the specific heat, thermal conductivity, and other properties of fuel meat materials commonly used, or considered for use, in research and test reactors. Also included are formulae relating the density of uranium in the fuel meat with the weight fraction of uranium and the volume fraction of the dispersed phase.

1.0 URANIUM DENSITY RELATIONS

In general, the density and weight fraction of uranium in fuel meat composed of aluminum, a dispersed phase, and voids can be written as:

$$\rho_U = \frac{(1 - P) W_U}{\frac{1}{\rho_{Al}} - a W_U} \qquad W_U = \frac{\rho_U / \rho_{Al}}{(1 - P) + a \rho_U} \qquad (1)$$

where

$$a = \frac{1}{W_U^D} \left(\frac{1}{\rho_{Al}} - \frac{1}{\rho_D} \right) \qquad (2)$$

$$P = \text{Porosity} = \frac{\text{Volume of Voids}}{\text{Volume of Solids} + \text{Volume of Voids}}$$

W_U = Weight Fraction of Uranium in the Fuel Meat

W_U^D = Weight Fraction of Uranium in the Dispersed Phase

ρ_{Al} = Density of Aluminum = 2.7 g/cm³

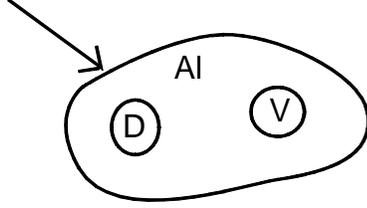
ρ_D = Density of the Dispersed Phase

A useful formula in relating the terminology used by physicists and the terminology used by fuel fabricators is the relationship between the uranium density in the fuel meat and the volume fraction (V_f^D) of the dispersed phase:

$$\rho_U = W_U^D \rho_D V_f^D \qquad (3)$$

The derivation of these formulae is given below.

Fuel Meat



D = Dispersed Phase

v = Void

Al = Aluminum Matrix

$$\text{Volume Balance for Fuel Meat: } V_m = V_D + V_{Al} + V_V$$

$$\text{Divide by } M_m \text{ (mass of meat): } \frac{V_m}{M_m} = \frac{V_D}{M_m} + \frac{V_{Al}}{M_m} + \frac{V_V}{M_m}$$

$$\text{Substitute: } \frac{V_m}{M_m} = \frac{1}{\rho_m}, \quad \frac{V_D}{M_m} = \frac{V_D}{M_D} \frac{M_D}{M_m} = \frac{W_D}{\rho_D},$$

$$\frac{V_{Al}}{M_m} = \frac{V_{Al}}{M_{Al}} \frac{M_{Al}}{M_m} = \frac{W_{Al}}{\rho_{Al}} = \frac{1 - W_D}{\rho_{Al}} \quad \text{and} \quad \frac{V_V}{M_m} = \frac{V_V}{V_m} \frac{V_m}{M_m} = \frac{P}{\rho_m}$$

to obtain:

$$\frac{1 - P}{\rho_m} = \frac{W_D}{\rho_D} + \frac{1 - W_D}{\rho_{Al}} = \frac{1}{\rho_{Al}} - W_D \left(\frac{1}{\rho_{Al}} - \frac{1}{\rho_D} \right)$$

$$\text{Substitute: } \rho_U = \frac{M_U}{M_m} \frac{M_m}{V_m} = W_U \rho_m \quad \text{and} \quad W_U = W_U^D W_D$$

to obtain:

$$\frac{(1 - P) W_U}{\rho_U} = \frac{1}{\rho_{Al}} - W_U \left[\frac{1}{W_U^D} \left(\frac{1}{\rho_{Al}} - \frac{1}{\rho_D} \right) \right] = \frac{1}{\rho_{Al}} - a W_U$$

= a

Solve for ρ_U :

$$\rho_U = \frac{(1 - P) W_U}{\frac{1}{\rho_{Al}} - a W_U} \quad \text{which is Eq. (1)}$$

Also:

$$\rho_U = \frac{M_U}{V_m} = \frac{M_U}{M_D} \frac{M_D}{V_D} \frac{V_D}{V_m} = W_U^D \rho_D V_f^D \quad \text{which is Eq.(3).}$$

2.0 THE URANIUM-ALUMINUM SYSTEM

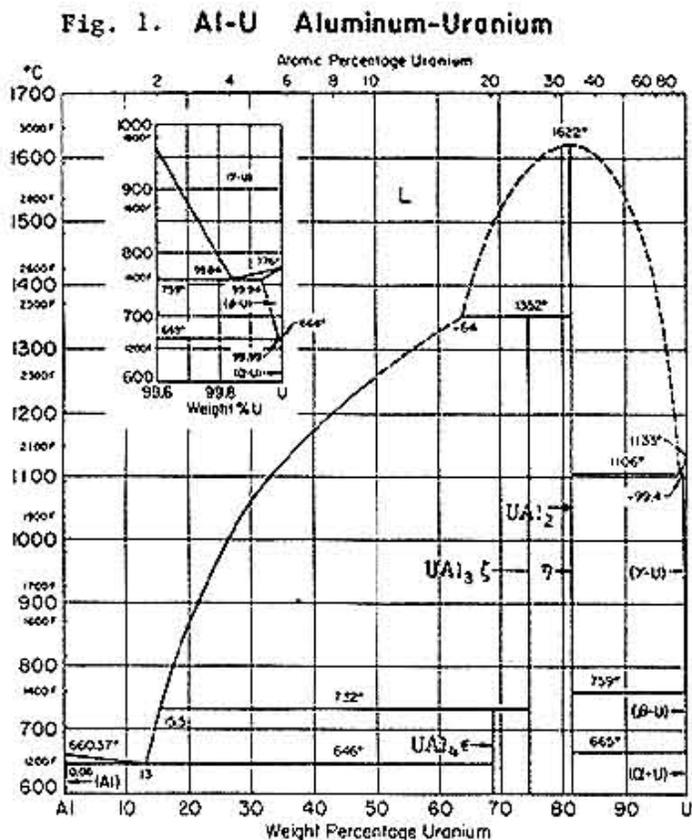
The uranium-aluminum system¹ (Fig. 1) contains three compounds - UAl_2 , UAl_3 , and UAl_4 - which are formed during cooling down from the molten state. Some properties² of these compounds are listed in Table 1:

Table 1. Some Properties of Uranium-Aluminum Compounds

Compound	Density, g/cm ³	W_U^D	Melting Point, °C
UAl_2	8.1	0.813	1590
UAl_3	6.8	0.744	1350
UAl_4^*	6.1 (Theoretical) 5.7 ± 0.3 (Measured ³)	0.685 0.640	730

*The compound UAl_4 has a defect structure³ in which some of the uranium sites are unoccupied. The compound corresponds stoichiometrically to $UAl_{4.9}$ (also referred to as $U_{0.9}Al_4$).

Since UAl_2 and UAl_3 react with an excess of aluminum at moderate temperatures to form UAl_4 , the relative amounts of these compounds that are present in the fuel meat of a finished plate or tube is a function of the wt-% of the uranium and the fabrication processes and heat treatments that are utilized.



3.0 URANIUM-ALUMINUM ALLOY FUEL

For uranium-aluminum alloy fuel with less than ~25 wt-% U, the alloy is mostly aluminum and UAl₄. Above ~25 wt-% U, a considerable amount of metastable UAl₃ may be present. The amount of retained metastable UAl₃ increases with increasing uranium content and with increasing impurity content. If it is advantageous, the brittle UAl₄ phase can be suppressed⁴ in favor of the more ductile UAl₃ phase through the use of ternary additions such as silicon.

3.1 Uranium Density Relations for U-Al Alloy Fuel

The densities of UAl₃ and UAl₄, the corresponding weight fractions of uranium in each compound, and the value of the parameter a in Eq. (2) are:

Uranium Compound	Density, g/cm ³	W _U ^D	a
UAl ₃	6.8	0.744	0.300
UAl ₄	5.7 ± 0.3	0.640	0.305

The relationships between the uranium density and the weight fraction of uranium in the fuel meat, and between the uranium density and the volume fraction of the uranium compound are:

$$\rho_U = \frac{(1 - P) W_U}{0.370 - a W_U} \quad W_U = \frac{0.370 \rho_U}{(1 - P) + a \rho_U}$$

$$\rho_U = 5.1 v_f^{UAl_3} \quad \rho_U = 3.7 v_f^{UAl_4}$$

3.2 Specific Heat of U-Al Alloy Fuel

The specific heat of U-Al alloy fuel meat depends on the relative amounts of its constituents and their respective specific heats. The specific heat of "pure" aluminum is given by:⁵

$$C_{p,Al} = 0.892 + 0.00046 T \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \quad (4)$$

Measured specific heats for pure uranium-aluminum compounds such as UAl₃ and UAl₄ are not available. The best data available are calculated⁶ from specific heat data⁷ for uranium and aluminum employing Kopp's law⁸ and values of excess heat capacity.⁹ The data presented in Ref. 6 yield the following specific heats for UAl₃ and UAl₄:

$$C_{p,UAl_3} = 0.329 + 0.00021 T \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \quad (20-600 \text{ } ^\circ\text{C}) \quad (5)$$

$$C_{p,UAl_4} = 0.473 + 0.00024 T \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \quad (20-600 \text{ } ^\circ\text{C}) \quad (6)$$

Using $W_U^D = 0.744$ for fully-enriched UAl_3 and $W_U^D = 0.640$ for fully-enriched UAl_4 :

$$\begin{aligned} C_{p,U-Al} \text{ alloy} &= (1.0 - W_U/0.744) C_{p,Al} + (W_U/0.744) C_{p,UAl_3} \\ &\text{(100\% } UAl_3\text{)} \\ &= 0.892 + 0.00046 T - W_U (0.757 + 0.00034 T) \text{ J/g K, } T \text{ in } ^\circ\text{C} \end{aligned}$$

$$\begin{aligned} C_{p,U-Al} \text{ alloy} &= (1.0 - W_U/0.640) C_{p,Al} + (W_U/0.640) C_{p,UAl_4} \\ &\text{(100\% } UAl_4\text{)} \\ &= 0.892 + 0.00046 T - W_U (0.655 + 0.00034 T) \text{ J/g K, } T \text{ in } ^\circ\text{C} \end{aligned}$$

Since most plate-type research reactor fuels contain < 25 wt-% U, the uranium compound in the fuel meat is mostly UAl_4 . At 25 wt-% U and 40°C, for example, specific heats for U-Al alloy calculated assuming 100% UAl_4 and 100% UAl_3 differ by less than 4%. In practice, only a small fraction of the uranium compound is likely to be UAl_3 .

3.3 Thermal Conductivity of U-Al Alloy Fuel

The thermal conductivity of U-Al alloy fuel meat decreases with increasing weight fraction of uranium¹⁰ as shown in Fig. 2. A linear regression of the data points for the as-cast material yields the relation:

$$K = 2.17 - 2.76 W_U$$

K = thermal conductivity of fuel meat, W/cm K

W_U = weight fraction of uranium in the fuel meat.

Data presented in Fig. 3 for various uranium weight loadings in uranium-aluminum alloy fuel indicate only a small decrease in thermal conductivity with increasing temperature. Over the temperature ranges expected in research and test reactors, the thermal conductivity of U-Al alloy fuel meat can be assumed to be constant.

The thermal conductivity of a fuel plate can be calculated using:

$$\frac{t_{\text{plate}}}{K_{\text{plate}}} = \frac{t_{\text{meat}}}{K_{\text{meat}}} + \frac{2 t_{\text{clad}}}{K_{\text{clad}}} \quad (7)$$

where t_{plate} , t_{meat} , and t_{clad} are the thicknesses of the plate, fuel meat, and cladding, respectively.

The thermal conductivity of 1100 Al cladding, for example, is 2.22 W/cm K. For U-Al alloy fuel meat containing 21 wt-% U with a thickness of 0.51 mm and 1100 Al cladding with a thickness of 0.38 mm, the thermal conductivity of the fuel plate would be 1.92 W/cm K.

4.0 UAl_x-Al DISPERSION FUEL

The information presented in Section 2.0 on the uranium-aluminum system also applies to UAl_x-Al dispersion fuel. The three broad steps in the manufacture of UAl_x-Al dispersion fuel are production of the UAl_x powder, fabrication of the UAl_x-Al core compacts, and fabrication of the fuel plates.

Specified and typical properties¹¹ of the UAl_x powder and UAl_x-Al core compacts that are used to manufacture finished fuel plates with uranium densities up to 1.7 g/cm³ for the Advanced Test Reactor (ATR) are shown in Table 2. Typical UAl_x powder consists of about 6 wt-% UAl₂, 61 wt-% UAl₃, and 31 wt-% UAl₄. During the hot rolling and annealing steps in fabricating fuel plates, almost all of the UAl₂ reacts with aluminum from the matrix to form UAl₃ and some of the UAl₃ reacts with aluminum to form UAl₄. Thus, the core (fuel meat) of a finished plate contains UAl₃ and UAl₄ as the fuel compounds. The actual fractions of UAl₃ and UAl₄ in a finished plate will vary from manufacturer to manufacturer depending on the processes and heat treatments that are utilized in fabricating the powder, core compacts, and fuel plates.

In the following discussions, it is assumed that the UAl_x in the fuel meat of finished fuel plates consists of 60 wt-% UAl₃ and 40 wt-% UAl₄.

4.1 Uranium Density Relations for Aluminide Fuel

For UAl_x in the meat of finished fuel plates that consists of 60 wt-% UAl₃ and 40 wt-% UAl₄, the density of the UAl_x is:

$$\begin{aligned}\rho_{\text{UAl}_x} &= W_{\text{UAl}_3} \rho_{\text{UAl}_3} + W_{\text{UAl}_4} \rho_{\text{UAl}_4} \\ &= 0.6 (6.8) + 0.4 (5.7) = 6.4 \text{ g/cm}^3\end{aligned}$$

using the measured densities of UAl₃ and UAl₄ from Table 1. Additionally, the weighted value of x in UAl_x (taking 4.9 aluminum atoms per uranium atom in UAl₄) is about 3.8 and the uranium weight fraction (W_U^D) in the UAl_x is about 0.70.

Substituting these values into Eq.(2), a value of "a" = 0.306 is obtained. The relationships between the uranium density and the weight fraction of uranium in the fuel meat, and between the uranium density and the volume fraction of the dispersed phase are then:

$$\rho_U = \frac{(1 - P) W_U}{0.370 - 0.306 W_U} \quad W_U = \frac{0.370 \rho_U}{(1 - P) + 0.306 \rho_U}$$

$$\rho_U = 4.5 V_f^{\text{UAl}_x}$$

A plot¹¹ of the fuel meat (core) density and porosity of uranium aluminide fuel for different uranium densities is shown in Fig. 4. The figure indicates that at constant core density, the porosity increases with increasing uranium loading. For calculational purposes, an average porosity of 7 vol-% is commonly used.

Table 2. Properties of Uranium Aluminide (UAl_x) Powder and Core Compacts Used to Manufacture Fuel for the ATR Reactor (From Ref. 11).

Power	Specified	Typical
Isotopic Composition:		
^{235}U content	93.0 ± 1.0 wt%	93.19
^{238}U content	6.0 ± 1.0 wt%	5.37
^{236}U content	0.3 ± 0.2 wt%	0.44
^{234}U content	1.2 maximum wt%	1.00
Chemical Composition:		
Uranium	69.0 ± 3.0 wt%	71.28
Oxygen	0.60 wt% maximum	0.25
Carbon	0.18 wt% maximum	0.05
Nitrogen	0.045 wt% maximum	0.032
Hydrogen	0.020 wt% maximum	0.005
Nonvolatile matter	99.0 wt% minimum	99.9
Easily extracted fatty and oily matter	0.2 wt% maximum	0.09
EBC ^a	30 ppm maximum	<6
Physical Properties:		
Particle size		
U.S. standard mesh	-100 +325 mesh	76.0
	= 75% minimum	
	-325 mesh	24.0
	= 25% maximum	
Crystalline constituents - by x-ray diffraction	50% UAl_3 minimum no unalloyed U	6% UAl_2 ^b 63% UAl_3 31% UAl_4
Core Compact		
For ATR zone loaded		
core fuel loading, g $^{235}\text{U}/\text{cm}^3$ core	1.00, 1.30, 1.60	
(maximum) wt% UAl_3 in core	1.00 g $^{235}\text{U}/\text{cm}^3$ 1.30 g $^{235}\text{U}/\text{cm}^3$ 1.60 g $^{235}\text{U}/\text{cm}^3$	46.4 54.4 62.8
Uranium concentration, U atom/ cm^3 of core (maximum)	1.00 g $^{235}\text{U}/\text{cm}^3$ 1.30 g $^{235}\text{U}/\text{cm}^3$ 1.60 g $^{235}\text{U}/\text{cm}^3$	2.76×10^{21} 3.58×10^{21} 4.41×10^{21}

^aEBC = equivalent boron content

^bEither UAl_2 or UAl_3 reacts with an excess of aluminum at moderate temperatures to form UAl_4 . Thus, the finished fuel plate cores, ready for reactor use, contain UAl_3 and UAl_4 as the fuel compound.¹¹

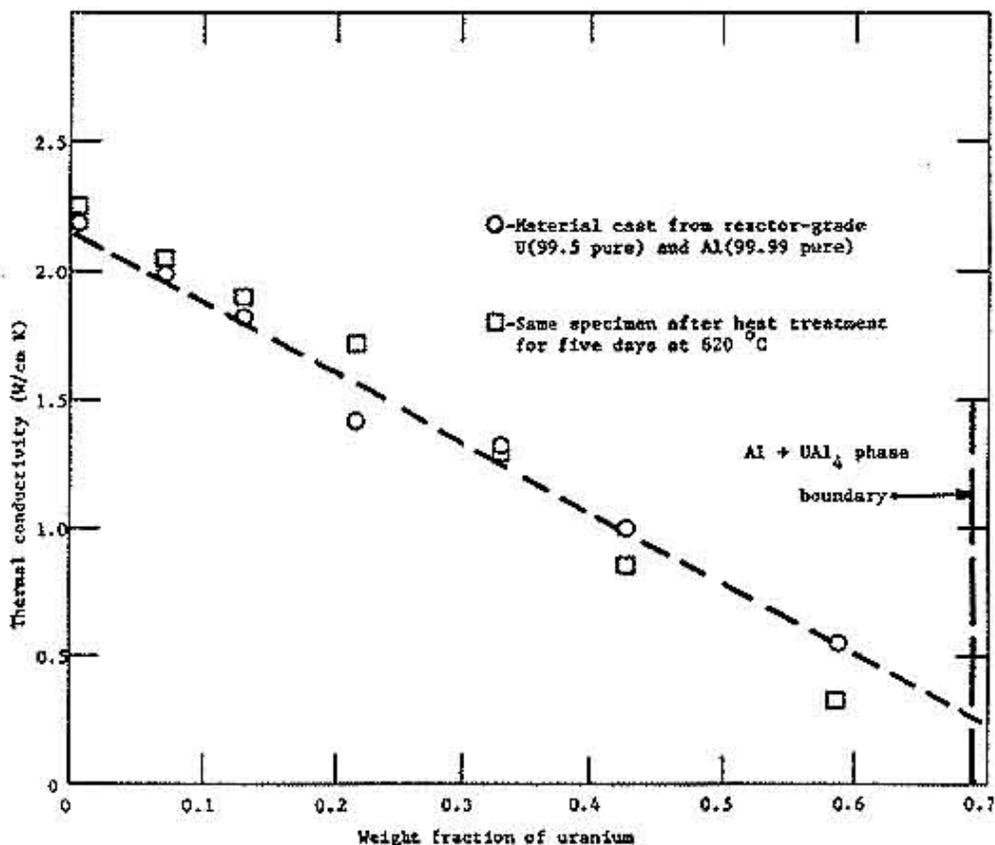
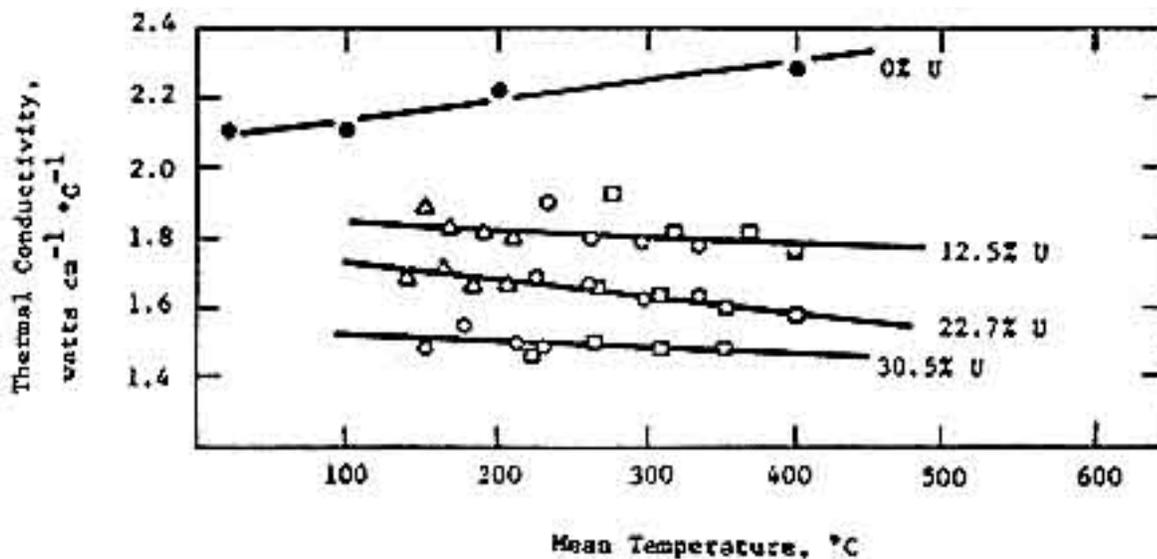


Fig. 2, Thermal Conductivity of U-Al Alloy at 65°C (Ref. 10).



(Δ, ○, and □ Denote Results Obtained on Different Runs)

Fig. 3, Thermal Conductivity versus Temperature for Various Loadings (in Wt%U) of U-Al Alloy Fuel (Ref. 10).

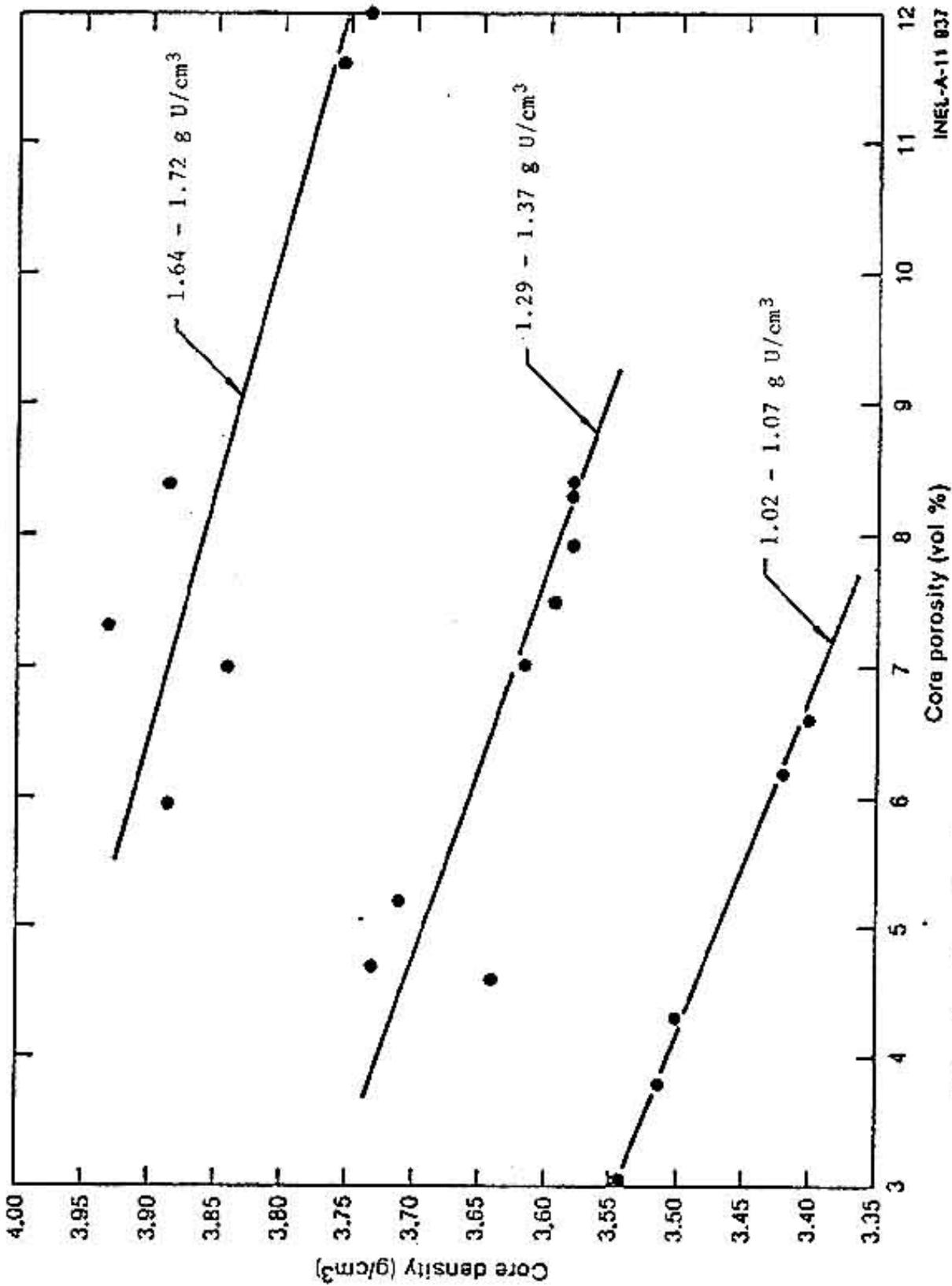


Fig. 4. Core Density and Porosity of Uranium Aluminate Fuel Plates with Different Fuel Loadings (Ref. 11).

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4.2 Specific Heat of Aluminide Fuel

Using Eqs.(5) and (6), the specific heat of UAl_x that consists of 60 wt-% UAl_3 and 40 wt-% UAl_4 is given by:

$$\begin{aligned} C_p \text{ } UAl_x &= W_{UAl_3} C_p \text{ } UAl_3 + W_{UAl_4} C_p \text{ } UAl_4 \\ &= 0.387 + 0.00022 T \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \end{aligned}$$

The specific heat of UAl_x -Al fuel meat is obtained by summing the specific heats of the UAl_x and aluminum phases, weighted by their respective fractions:

$$\begin{aligned} C_{p,UAl_x-Al} &= (1.0 - W_U/0.7) C_{p,Al} + (W_U/0.7) C_{p,UAl_x} \\ &= C_{p,Al} + 1.43 W_U (C_{p,UAl_x} - C_{p,Al}) \\ &= 0.892 + 0.00046 T - W_U (0.722 + 0.00034 T) \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \end{aligned}$$

4.3 Thermal Conductivity of Aluminide Fuel

Available data on the thermal conductivity of aluminide fuels (that are typical of those in reactor use) are limited to three data points (Ref. 6) calculated for ATR sample fuel plates using thermal diffusivity measurements (Ref. 10) at Battelle Northwest Laboratories on MTR-ETR type fuel plates. The data from Table II of Ref. 6 are reproduced in Table 3 below.

Table 3. Thermal Conductivity of ATR Sample Fuel Plates Calculated (Ref. 6) from Thermal Diffusivity Measurements (Battelle Northwest, Ref.10).

Plate	Fuel	Thermal Diffusivity, cm^2/s		Fuel Plate Density, g/cm^3		Heat Capacity, $\text{J}/\text{g } ^\circ\text{C}$		Thermal Conductivity, $\text{W}/\text{cm K}$	
		25 $^\circ\text{C}$	600 $^\circ\text{C}$	25 $^\circ\text{C}$	600 $^\circ\text{C}$	25 $^\circ\text{C}$	600 $^\circ\text{C}$	25 $^\circ\text{C}$	600 $^\circ\text{C}$
P-1-1047	UAl_x	0.32	0.25	2.953	2.830	0.766	0.996	0.724	0.703
P-1-1048	UAl_x	0.38	0.33	2.980	2.855	0.758	0.984	0.858	0.925
P-5-576	UAl_x	0.33	0.24	3.00	2.872	0.737	0.963	0.728	0.661

Thermal conductivities in Table 3 were obtained using the relation:

$$K = \alpha \rho C_p$$

where K is the thermal conductivity, α is the thermal diffusivity, ρ is the density of the plate, and C_p is the heat capacity.

The sample fuel plates in Table 3 had a thickness of 1.296 mm, with a 6061 Al cladding thickness of about 0.394 mm and an assumed fuel meat thickness of 0.508 mm. The fuel meat contained about 35.4 vol-% UAl_x (57.7 wt-% UAl_x) and had a porosity of about 6 vol-%. The matrix material was X8001 aluminum alloy and the uranium density in the fuel meat was about 1.6 g/cm³.

The thermal conductivity of the UAl_x-Al fuel meat can be calculated from the fuel plate data in Table 3 using the relation:

$$\frac{t_{\text{plate}}}{K_{\text{plate}}} = \frac{t_{\text{meat}}}{K_{\text{meat}}} + \frac{2 t_{\text{clad}}}{K_{\text{clad}}}$$

where t_{plate} , t_{meat} , and t_{clad} are the thicknesses of the plate, fuel meat, and cladding, respectively. 6061 Al cladding has a thermal conductivity of 1.80 W/cm K and is essentially constant over the temperature range considered. The thermal conductivity of the UAl_x-Al fuel meat for the three fuel plates listed in Table 3 is then:

Table 4. Calculated Thermal Conductivities of the UAl_x-Al Fuel Meat in the Three Sample Fuel Plates in Table 3.

Plate	Fuel Meat	Vol-% UAl _x	Percent Porosity	Thermal Conductivity of Fuel Meat, W/cm K	
				25°C	600°C
P-1-1047	UAl _x -Al	35.4	6	0.376	0.361
P-1-1048	UAl _x -Al	35.4	6	0.474	0.527
P-5-576	UAl _x -Al	35.4	6	0.378	0.334

The thermal conductivity data for UAl_x-Al fuel at 25°C are plotted in Fig. 8 (Section 6), which compares the thermal conductivities of U₃O₈-Al, U₃Si₂-Al, and U₃Si-Al fuel meats as a function of the volume percent of fuel dispersant plus voids. From the data in Fig. 7, we conclude that all four of these dispersion fuels have approximately the same thermal conductivity. Since the thermal conductivities of UAl_x and U₃Si₂ and the metallurgical properties of UAl_x-Al fuel and U₃Si₂-Al fuel are very similar, we suggest that the measured thermal conductivity data for U₃Si₂-Al fuel be used for UAl_x-Al fuel as well.

5.0 U₃O₈-Al DISPERSION FUEL

5.1 Uranium Density Relations for Oxide Fuel

The density of the high-fired U₃O₈ used by the High Flux Isotope Reactor (HFIR) at ORNL is 8.22 g/cm³ and $W_U^D = 0.846$. Substituting these data into Eq. (2), one obtains $a = 0.294$. The relationships between the uranium density and the weight fraction of uranium in the fuel meat, and between the uranium density and the volume fraction of the dispersed phase are then:

$$\rho_U = \frac{(1 - P) W_U}{0.370 - 0.294 W_U} \qquad W_U = \frac{0.370 \rho_U}{(1 - P) + 0.294 \rho_U}$$

$$\rho_U = 7.0 V_f^{U_3O_8}$$

The void content of U₃O₈-Al fuel meat depends on the concentration of U₃O₈ and to a lesser extent on the fuel meat thickness. The void content as a function of U₃O₈ concentration for plates with two fuel meat thicknesses fabricated by ORNL¹² are shown in Fig. 5. Appropriate values of P for use in the uranium density relations should be obtained from this figure or from similar data supplied by the fuel manufacturer.

5.2 Specific Heat of Oxide Fuel

Specific heat data¹³ for U₃O₈ in the temperature range from 0-300°C is represented approximately by the linear relationship:

$$C_{p,U_3O_8} = 0.27 + 0.00030 T \quad \text{J/g K, } T \text{ in } ^\circ\text{C}$$

If W_U is the uranium weight fraction and $C_{p,Al}$ is the specific heat of aluminum [Eq.(4)], the specific heat of the U₃O₈-Al fuel meat is given by:

$$\begin{aligned} C_{p, U_3O_8-Al} &= (1 - W_U/0.848) C_{p,Al} + (W_U/0.848) C_{p,U_3O_8} \\ &= 0.892 + 0.00046 T - W_U (0.734 + 0.00019 T) \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \end{aligned}$$

5.3 Thermal Conductivity of Oxide Fuel

Figure 6 shows a curve of measured¹⁴ thermal conductivity versus uranium loading for U₃O₈-Al dispersion material. For the lightly loaded dispersions, the decrease in conductivity with increasing volume fraction of U₃O₈ is linear, primarily due to the substitution of the low conductivity oxide ($k \sim 0.3-0.5$ W/m K)¹⁵ for aluminum. As more U₃O₈ is added, however, the thermal conductivity drops more dramatically. In the range of uranium loadings between 2.5 and 3.1 g/cm³, the thermal conductivity ranges from 30 to 12 W/m K.

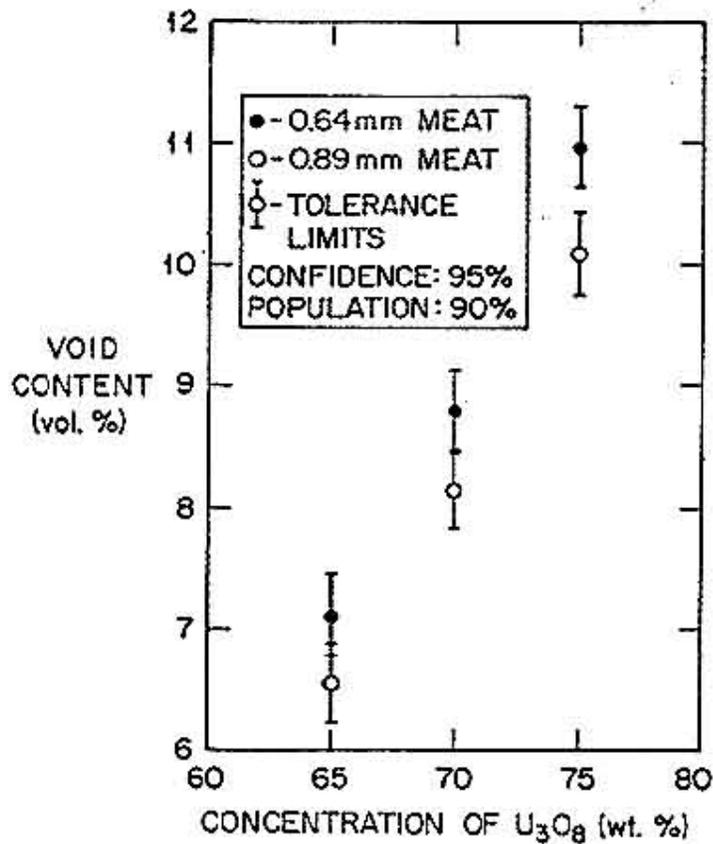


Fig. 5. Void Content of U_3O_8 -Al Fuel Meat versus Concentration of U_3O_8 and Fuel Meat Thickness

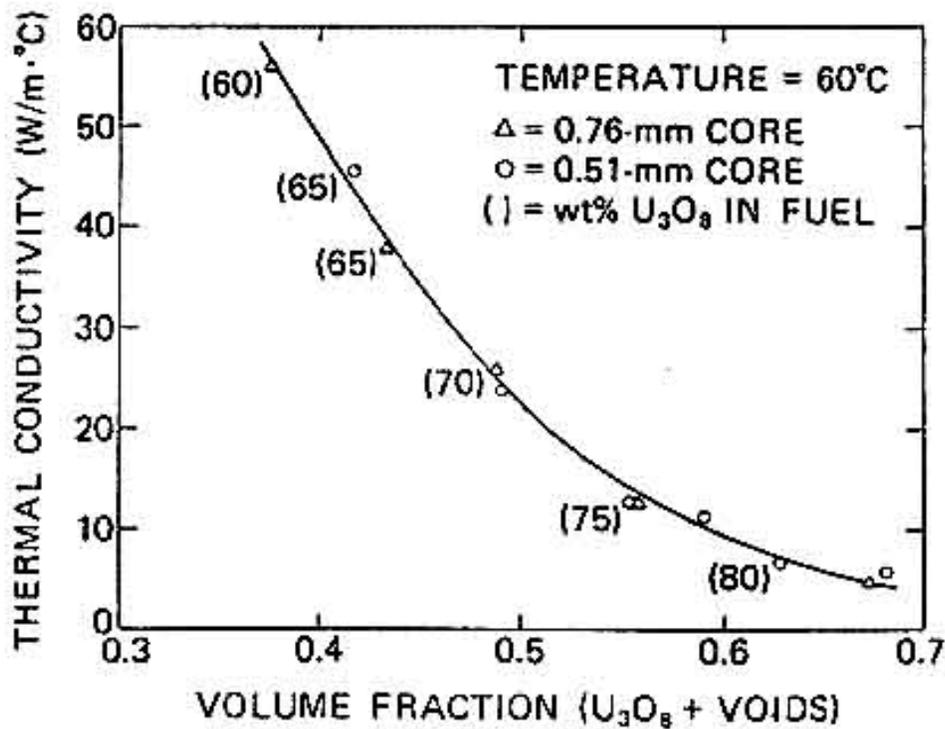


Fig. 6. Thermal conductivity of U_3O_8 -Al core region depends significantly on the volume fraction of U_3O_8 + voids.

6.0 U₃Si₂-Al AND U₃Si-Al DISPERSION FUELS

The development and testing of uranium silicide fuels has been an international effort, involving national reduced enrichment programs, several commercial fuel fabricators, and several test reactor operators. Numerous results of this effort have been published previously. Some of the results are summarized in this section.

As with the uranium-aluminum system, the uranium-silicon system normally consists of a mixture of intermetallic compounds, or phases. The quantity of each phase present depends upon the composition and homogeneity of the alloy and on its heat treatment. Since the different phases behave differently under irradiation, knowledge of the phases to be expected in the fuel is necessary to correctly interpret test results and to prepare specifications. For this reason, a detailed discussion of the phases in the uranium-silicon system is presented in Appendix I-1.2 (Ref. 16). Further properties of uranium silicide fuels can be found in Refs. 17-19.

6.1 Uranium Density Relations for Silicide Fuels

The densities of the U₃Si₂ and U₃Si dispersants that were measured at ANL, the corresponding weight fractions of uranium in each dispersant, and the value of the parameter "a" in Eq.(2) are given below.

Silicide Dispersant	wt-% Si	Measured Density g/cm ³	$\frac{D}{W_U}$	a
U ₃ Si ₂	7.5	12.2 ^a	0.925	0.312
U ₃ Si	4.0	15.2 ^b	0.960	0.317

^aAs-arc-cast.

^bAfter heat treatment of 72 h at 800°C.

The relationships between the uranium density and the weight fraction of uranium in the fuel meat, and between the uranium density and the volume fraction of the dispersed phase are then:

$$\rho_U = \frac{(1 - P) W_U}{0.370 - a W_U} \quad W_U = \frac{0.370 \rho_U}{(1 - P) + a \rho_U}$$

with appropriate values of the parameter "a" for each dispersant.

$$\rho_U = 11.3 V_f^{U_3Si_2} \quad \rho_U = 14.6 V_f^{U_3Si}$$

Porosity remaining after fabrication of dispersion fuel meat provides space to accommodate the initial swelling of the fuel particles under irradiation. Data obtained at ANL from measurements²⁰ on U₃Si₂ miniplates are plotted in Fig. 7. These data are well fit¹⁷ by the cubic function:

$$V_P = 0.072 V_F - 0.275 V_F^2 + 1.32 V_F^3$$

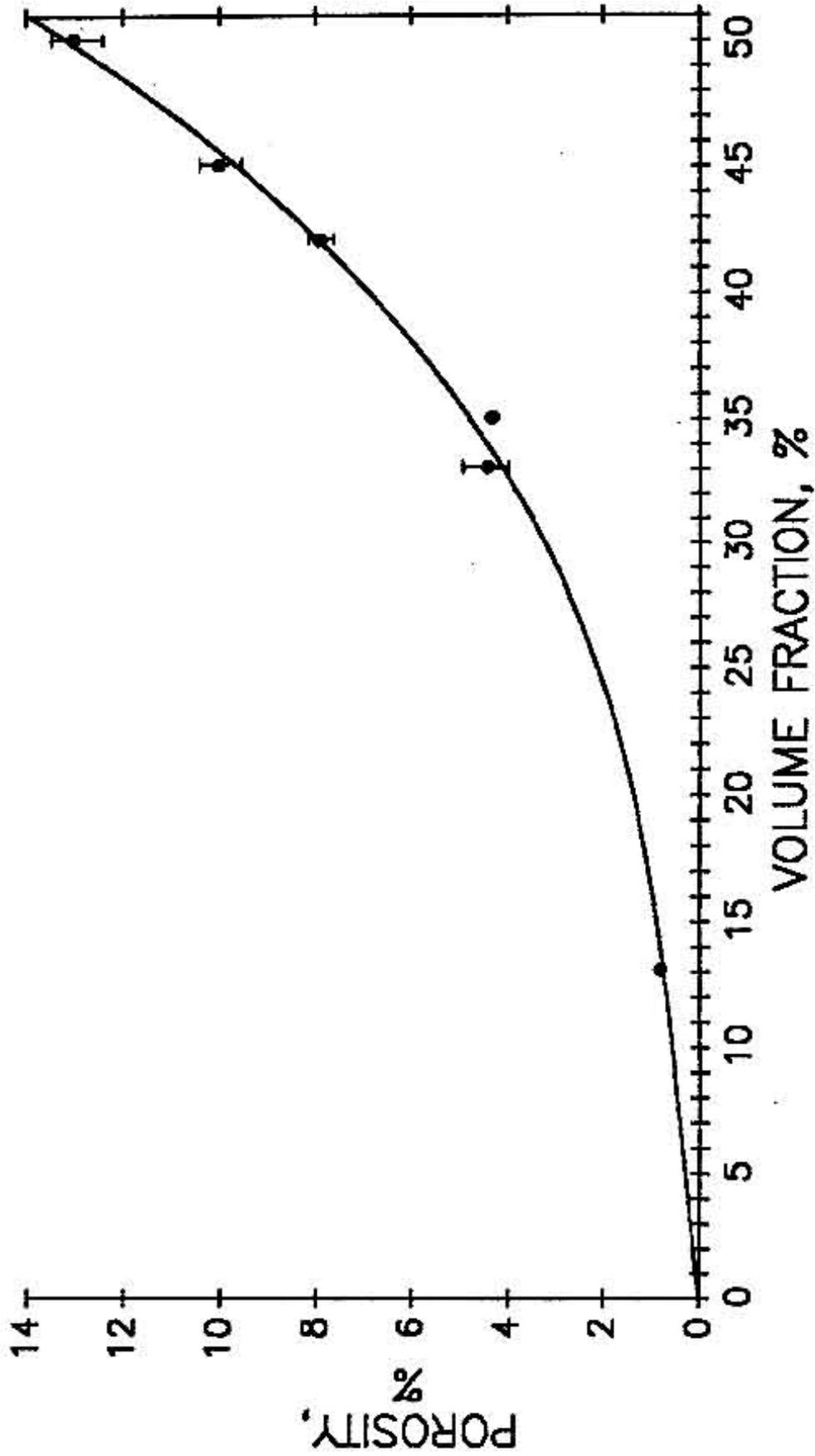


Fig. 7. Percent Porosity as a Function of the Volume Percent of U_3Si_2 in U_3Si_2 -Al Fuel Meat.

where V_p and V_f are the volume fractions of porosity and fuel dispersant in the meat, respectively. The amount of as-fabricated porosity increases significantly as the volume loading of fuel dispersant increases because it becomes more difficult for the matrix aluminum matrix to flow completely around all fuel particles, especially those in contact with one another²⁰.

It is important to note that the porosity in the fuel meat of fabricated fuel plates varies from fabricator to fabricator due to differences in manufacturing techniques in the aluminum alloys of the cladding. For example, consider the nominally identical U_3Si_2 fuel elements fabricated by B&W, CERCA, and NUKEM for irradiation testing in the Oak Ridge Research Reactor. The porosity content of the fuel cores produced by a given fabricator remained virtually constant, but there was a variation from fabricator to fabricator: 4 vol-% for CERCA, 7-8 vol-% for NUKEM, and 9-10 vol-% for B&W. Differences in material or fabrication parameters which might have contributed to the different amount of porosity include: (1) strength of the aluminum alloy used for frames and covers -- the CERCA alloy was by far the strongest while the B&W alloy was the weakest; (2) the rolling temperature -- 425°C for CERCA and NUKEM and ~500°C for B&W; (3) the amount of fines in the U_3Si_2 powder -- 40 wt-% for CERCA and 17-18 wt-% for NUKEM and B&W; (4) the rolling schedule, especially the amount of cold reduction; and (5) the relationship between the size of the compact and the size of the cavity in the frame.

6.2 Specific Heats of Silicide Fuels

The specific heats of U_3Si_2 and U_3Si as a function of temperature have been derived¹⁷ from plots of specific heat data²¹ for stoichiometric U_3Si and for a U-Si alloy at 6.1 wt-% Si:

$$C_{p,U_3Si_2} = 0.199 + 0.000104 T \quad \text{J/g K, } T \text{ in } ^\circ\text{C}$$

$$C_{p,U_3Si} = 0.171 + 0.000019 T \quad \text{J/g K, } T \text{ in } ^\circ\text{C}$$

If W_U is the uranium weight fraction and $C_{p,Al}$ is the specific heat of aluminum [Eq.(4)], the specific heats of U_3Si_2 -Al and U_3Si -Al fuel meat are given by:

$$\begin{aligned} C_{p,U_3Si_2-Al} &= (1 - W_U/0.925) C_{p,Al} + (W_U/0.925) C_{p,U_3Si_2} \\ &= 0.892 + 0.00046 T - W_U (0.749 + 0.00038 T) \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \end{aligned}$$

$$\begin{aligned} C_{p,U_3Si-Al} &= (1 - W_U/0.960) C_{p,Al} + (W_U/0.960) C_{p,U_3Si} \\ &= 0.892 + 0.00046 T - W_U (0.751 + 0.00046 T) \quad \text{J/g K, } T \text{ in } ^\circ\text{C} \end{aligned}$$

6.3 Thermal Conductivity of Silicide Fuels¹⁷

Both U_3Si_2 and U_3Si have a thermal conductivity of ~15 W/m K²². Values of the thermal conductivities of the fuel meat in unirradiated U_3Si_2 -Al dispersion fuel plates, measured at 60°C, are listed in Table 5 and are plotted in Fig. 8.²³ Most of the samples were cut from miniature fuel plates produced at ANL for use in out-of-pile studies. Two samples came from a full-sized plate from a lot of plates fabricated by CERCA for the ORR test elements. The porosities of these miniplates follows the trend discussed in Section 6.1 but are somewhat larger, owing, presumably, to the different shape of the fuel zone than in the miniplates fabricated for irradiation testing (cylindrical rather than rectangular compacts were used).

Table 5. Thermal Conductivities of U₃Si₂-Aluminum Dispersions

Sample Identif- ication	Fraction of Fuel -325 Mesh, wt-%	U ₃ Si ₂ Volume ¹ Fraction, %	Porosity, ² vol-%	Thermal Conduct.of Dispersion at 60°C, W/m K	Temperature Coefficient, W/m K ²
CS148	15	13.7	1.9	181	0.148
CS106	15	32.3	6.0	78	0.029
CS140	0	39.4	9.2	40	0.014
CS141	15	37.0	9.3	48	5 x 10 ⁻⁴
CS142	25	39.1	9.5	40	0.017
CERCA #1	41.5	46.4	4.0	59	0.161
CERCA #2	41.5	46.4	4.0	59	0.076
CS143	15	46.4	15.4	13.9	0.010

¹Determined on the thermal conductivity specimens using a radiographic technique.

²Average value for roll-bonded fuel plate.

In Fig. 8, the thermal conductivity decreased rapidly as the volume fraction of fuel plus porosity increases (and the volume fraction of aluminum matrix decreases), owing to the ~14 times larger thermal conductivity of aluminum than U₃Si₂. For very low volume loading of U₃Si₂, it would be expected that the thermal conductivity of the dispersion would be proportional to the amount of aluminum present, since the aluminum matrix should provide a continuous thermal path. Indeed, this is the case for sample CS148. At higher volume fractions of U₃Si₂ plus void, however, the aluminum ceases to be the continuous phase, and the thermal conductivity decreases more rapidly than the volume fraction of aluminum. At very high loadings the aluminum ceases to play a significant role, and the thermal conductivity approaches that of the fuel. It may even become lower than that of the fuel alone because of poor thermal contact between fuel particles. The microstructure of the meat, specifically the distribution of the voids, can significantly affect the thermal conductivity. It appears that thin planar regions in which voids are associated with fractured fuel particles are responsible for the large difference in thermal conductivity exhibited by the CERCA samples and sample CS143. The larger void content of the CS samples than measured in the miniplates fabricated for irradiation testing or in full-sized plates most likely indicated the presence of more of such planar void regions. Therefore, it is believed that the thermal conductivity curve in Fig. 8 for U₃Si₂-Al fuel meat represents essentially a lower limit for the thermal conductivities of full-sized fuel plates.

The data for U₃Si₂-Al dispersions are virtually indistinguishable from those obtained in the same series of measurements for U₃Si-Al dispersions. They are also quite similar to data obtained in other measurements of thermal conductivities of UAl_x-Al dispersions⁶ and U₃O₈-Al dispersions.¹⁴ The U₃O₈-Al data fall somewhat below the U₃Si₂-Al data, possibly because the friable nature of U₃O₈ leads to the formation of more planar void regions than are present in U₃Si₂-Al fuel.

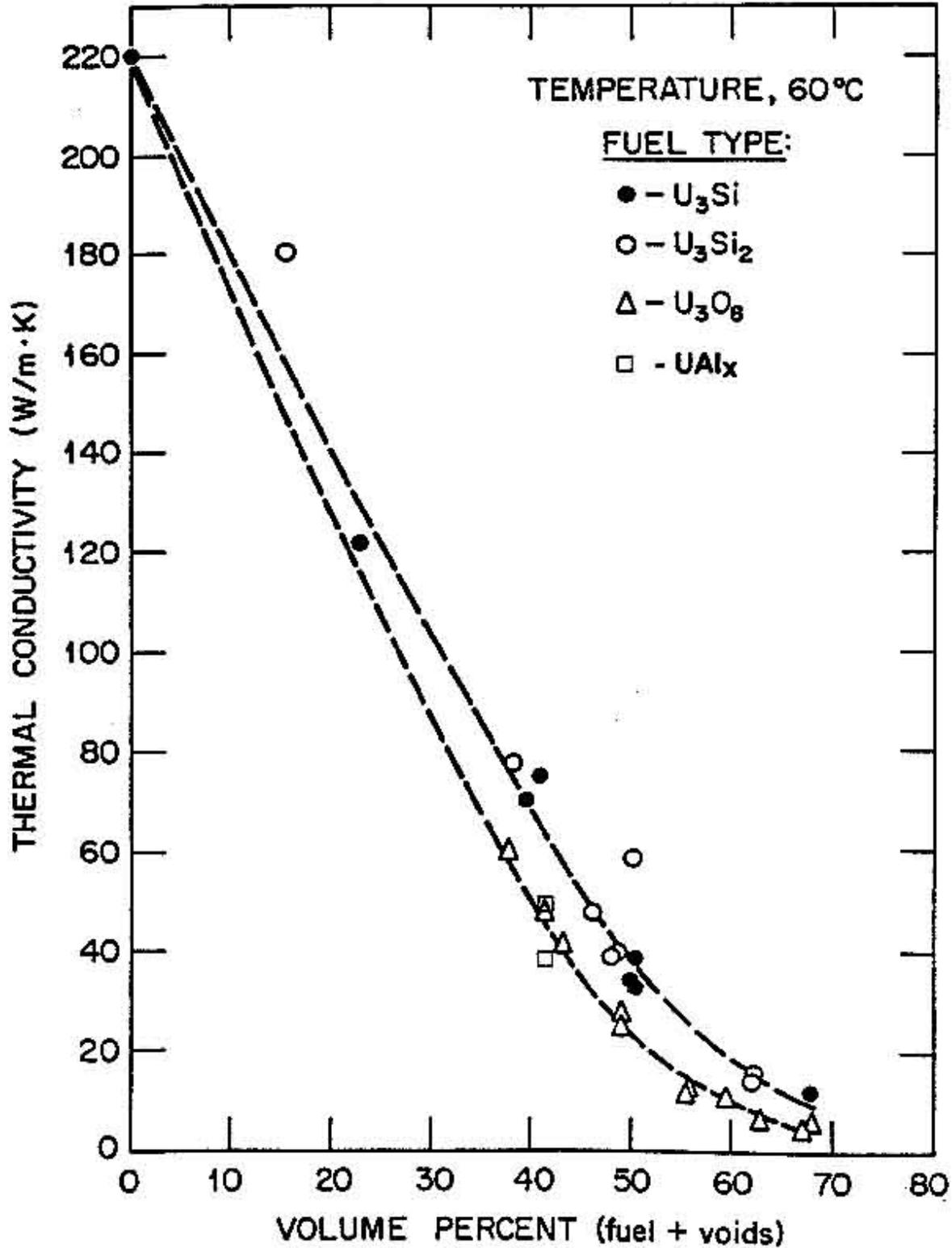


Fig. 8. Thermal Conductivities of Uranium Silicide-, U_3O_8 -, and UAl_x -Aluminum Dispersion Fuels as a Function of Volume Fraction of Fuel Particles Plus Voids (Porosity).

REFERENCES

1. Metals Handbook, Eighth Edition, Volume 8, "Metallography, Structures, and Phase Diagrams" (1973).
2. H.S. Kalish et al., "Uranium Alloys" in Reactor Handbook, Vol. I, Materials, p. 174, C.R. Tipton, Ed., Interscience Publishers, Inc., New York (1960).
3. B.S. Borie, "Crystal Structure of UAl_4 ", Journal of Metals, Vol. 3, September 1951, p. 800.
4. D. Stahl, "Fuels for Research and Test Reactors, Status Review, July 1982", ANL-83-5, December 1982.
5. CRC Handbook of Chemistry and Physics, 58th Edition (1977).
6. R.R. Hobbins, "The Thermal Conductivity and Heat Capacity of UAl_3 and UAl_4 ", Aerojet Nuclear Company Interoffice Correspondence, January 4, 1973.
7. Handbook of Chemistry and Physics, 40th Ed., Chemical Rubber Publishing Co. (1958).
8. L.S. Darken and R.W. Gurry, Physical Chemistry of Metals, McGraw-Hill (1953) p. 158.
9. P. Chiotti and J.A. Kateley, "Thermodynamic Properties of Uranium-Aluminum Alloys", J. Nucl. Mat. 32, 135 (1969).
10. J.L. Bates, "Thermal Diffusivity of MTR-ETR Type Fuel Plates, Battelle Pacific Northwest Laboratories Report BNWL-CC-456 (Jan. 10, 1966).
11. J.M. Beeston, R.R. Hobbins, G.W. Gibson, and W.C. Francis, "Development and Irradiation Performance of Uranium Aluminide Fuels in Test Reactors", Nuclear Technology 49, 136 (1980).
12. G.L. Copeland and M.M. Martin, "Fabrication of High-Uranium-Loaded U_{308} -Al Developmental Fuel Plates," Proc. International Meeting on Development, Fabrication, and Application of Reduced-Enrichment Fuels for Research and Test Reactors, Argonne National Laboratory, Argonne, Illinois, November 12-14, 1980.
13. Y.S. Youloukian and E.H. Buyco, Thermophysical Properties of Matter, Vol. V, "Specific Heat - Nonmetallic Substances" (1970).
14. G.L. Copeland and M.M. Martin, "Development of High-Uranium-Loaded U_{308} -Al Fuel Plates," Nucl. Tech. 56, 547 (1982).
15. Y.S. Youloukian and E.H. Buyco, Thermophysical Properties of Matter, Vol. I, "Thermal Conductivity" (1970).
16. R.F. Domagala, "Phases in U-Si Alloys", Proc. 1986 Int. Mtg. on Reduced Enrichment for Research and Test Reactors, Gatlinburg, Tennessee, November 3-6, 1986, Argonne National Laboratory Report ANL/RERTR/TM-9, CONF-861185 (May 1988) p. 45.

17. J.L. Snelgrove, R.F. Domagala, G.L. Hofman, T.C. Weincek, G.L. Copeland, R.W. Hobbs, and R.L. Senn "The Use of U_3Si_2 Dispersed in Aluminum in Plate-Type Fuel Elements for Research and Test Reactors", ANL/RERTR/TM-11, October 1987.
18. R.F. Domagala, T.C. Weincek, and H.R. Thresh, "Some Properties of U-Si Alloys in the Composition Range from U_3Si to U_3Si_2 ", Proc. 1984 Int. Mtg. on Reduced Enrichment for Research and Test Reactors, Argonne, Illinois, October 15-18, 1984, Argonne National Laboratory Report ANL/RERTR/TM-6, CONF-8410173 (July 1985) p. 47.
19. T.C. Weincek, R.F. Domagala, and H.R. Thresh, " Thermal compatibility Studies of Unirradiated Uranium Silicide Dispersed in Aluminum", Proc. 1984 Int. Mtg. on Reduced Enrichment for Research and Test Reactors, Argonne, Illinois, October 15-18, 1984, Argonne National Laboratory Report ANL/RERTR/TM-6, CONF-8410173 (July 1985) p. 61.
20. T.C. Weincek, "A Study of the Effect of Fabrication Variables on the Quality of Fuel Plates," Proc. 1986 Int. Mtg. on Reduced Enrichment for Research and Test Reactors, Gatlinburg, Tennessee, November 3-6, 1986, Argonne National Laboratory Report ANL/RERTR/TM-9, CONF-861185 (May 1988) p. 54.
21. H. Shimizu, "The Properties and Irradiation Behavior of U_3Si_2 ", Atomics International Report NAA-SR-10621, p.14 (July 25, 1965).
22. A.G. Samoilov, A.I. Kashtanov, and V.S. Volkov, Dispersion-Fuel Nuclear Reactor Elements, (1965), translated from the Russian by A. Aladjem, Israel Program for Scientific Translations Ltd.. Jerusalem, pp. 54-57 (1968).
23. R.K. Williams, R.S. Graves, R.F. Domagala, and T.C. Weincek, "Thermal Conductivities of U_3Si and U_3Si_2 -Al Dispersion Fuels", Proc. 19th Int. Conf. on Thermal Conductivity, Cookville, Tennessee, October 21-23, 1985, in press.