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THERMAL CONDUCTIVITY
AND ELECTRICAL RESISTIVITY
OF URANIUM OXYCARBIDE

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THERMAL CONDUCTIVITY AND ELECTRICAL RESISTIVITY
OF URANIUM OXYCARBIDE

By

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THERMAL CONDUCTIVITY AND ELECTRICAL RESISTIVITY
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ABSTRACT

The thermal diffusivity (α) of uranium oxycarbide was measured from 100 and 1500 °C as a function of oxygen concentration from 2 to 17 at.%. Thermal diffusivity decreased as oxygen content increased. Oxycarbides with the lowest oxygen content showed a decrease in α as temperature increased, whereas oxycarbides with the highest oxygen contents exhibited an increase in α as temperature increased. Thermal diffusivities of all specimens approach a common value of 0.05 cm²/sec at the high temperatures. Calculated thermal conductivities near 1500 °C are about 0.18 W/cm-°C, very near the reported values for nearly stoichiometric and slightly hypostoichiometric UC. Electrical resistivity of uranium oxycarbides between room temperature and 1775 °K increased as oxygen concentration increased from 2 to 16 at.%. Electronic thermal conductivity is predominant above about 1200 °C; phonon conductivity is predominant at lower temperatures and decreases with increasing oxygen content.

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THERMAL CONDUCTIVITY AND ELECTRICAL RESISTIVITY OF URANIUM OXYCARBIDE

J. Lambert Bates

INTRODUCTION

Uranium monocarbide has received considerable attention as a high-temperature fuel for nuclear reactors. Unfortunately, it has been difficult to prepare uranium monocarbide entirely free of oxygen, the oxygen often being found in solid solution in the monocarbide structure. In many studies the presence of small concentrations of oxygen has been ignored, although oxygen, either present in solution or as the oxide, can undoubtedly influence the thermal, electrical, and other properties of the monocarbide. A review of the existing thermal conductivity and electrical resistivity data for UC suggests that the wide scatter in reported thermal conductivity data at low temperatures may be due to differences in the oxygen contents of the carbide (Appendix A).

Recent extensive investigations have been made of the phase relations in the uranium monocarbide region of the uranium-carbon-oxygen system.⁽¹⁻⁴⁾ Uranium oxycarbides of desired composition can be prepared as solid solutions with oxygen contents approaching 17 at.%. Although primarily single phase UC_xO_y , the oxycarbide also contains second phases of higher carbides, oxides, and/or metallic uranium.

The purpose of this study was to measure the thermal diffusivity and electrical resistivity of some uranium oxycarbides in the temperature range from 100 to 1500 °C, to calculate thermal conductivity values, and to evaluate the data in terms of electronic and lattice conduction processes.

SUMMARY

The thermal diffusivity of uranium oxycarbide was measured from 100 to 1500 °C as a function of oxygen concentration from 2 to 17 at.%. The thermal diffusivity (α) of the oxycarbides decreased, and the shape of the α -versus-T curve changed as the oxygen content increased. Oxycarbides with the lowest oxygen content showed a decrease in α as the temperature increased, whereas the oxycarbides with the highest oxygen content exhibited a slight increase in α with an increase in temperature. The thermal diffusivities of all specimens approach a common value near $0.05 \text{ cm}^2/\text{sec}$ at the higher temperatures.

The calculated thermal conductivities of the uranium oxycarbides near 1500 °C are approximately $0.18 \text{ W/cm-}^\circ\text{C}$, very near the reported values for nearly stoichiometric and slightly hypostoichiometric UC.

The electrical resistivities of uranium oxycarbides containing 2 at.% and 16 at.% oxygen were measured from room temperature 1775 °K to 1500 °C. The electrical resistivity increases as the oxygen concentration increases.

Thermal conduction occurs by both electron and lattice (phonon) processes. The electronic thermal conductivity is predominant above about 1200 °C. The phonon conductivity is predominant at lower temperature and decreases with increasing oxygen content.

URANIUM OXYCARBIDE SPECIMENS

The uranium oxycarbide samples were prepared by reaction sintering of calculated quantities of uranium, graphite, and UO_2 powders.* The compacts were pulverized and milled to a fine powder in an all-nickel mill under highly purified argon. Pellets were cold pressed and sintered at 1700 °C for 2 hr in an atmosphere of carbon monoxide at or very near the decomposition pressure for each oxycarbide specimen. Details of the fabrication process have been published by Henry et al.⁽¹⁾

Ceramographic specimens were mounted in resin and rough ground on dry 3/0 and 2/0 energy papers. The specimens were first polished for 1 hr on a vibratory polisher with 0.3 μ Al_2O_3 in water on Metcloth. A final polish was obtained using 20 μ MgO powder in water on Gamal cloth for 1/2 to 1 hr. The polished surfaces on some specimens were improved by a 5 sec etching followed by hand polishing with MgO in water. This was repeated if necessary to remove final scratch marks. Specimens were etched in 1:1:1 nitric acid, acetic acid, and water at room temperatures.

The compositions and densities of the uranium oxycarbide specimens are shown in Table 1. The compositional relationships of each specimen to the reported uranium-carbon-oxygen phase diagram at 1700 °C are illustrated in Figure 1.

UCON-283 and UCON-458 ($\text{U}_{0.495}\text{C}_{0.485}\text{O}_{0.02}$) are single phase with minor traces of free uranium and UC_2 (Widmanstätten precipitate)(Figure 2). The relative slow cooling rate during the

* The uranium oxycarbide cylinders for this study were fabricated at the Albany Research Laboratory, United States Bureau of Mines, Albany, Oregon.

TABLE 1. Description of Uranium Oxycarbide Specimens

Designation	Composition, at. %			Density, g/cm ³	Microstructure
	U	C	O		
UCON-283	49.5	48.5	2	12.7	UC _x O _y with traces of free U and UC ₂
UCON-288	49.5	35.5	15	12.5	UC _x O _y and UO ₂ with traces of free U
UCON-289	49.5	33.5	17	12.3	UC _x O _y and UO ₂ with traces of free U
UCON-365	48.0	49.0	3	13.1	Two phase UC _x O _y + UC ₂ with trace of UO ₂ .
UCON-458	49.5	48.5	2	12.75	UC _x O _y with trace of UC ₂ in grain boundaries
UCON-459	49.5	34.5	16	17.5	UC _x O _y and UO ₂ and trace of free U.

thermal diffusivity measurements resulted in a slight increase in UC₂ precipitates. The presence of free uranium indicates inhomogeneity on a microscale. Some grain growth appears to have occurred during the measurements.

UCON-365 (U_{0.48}C_{0.49}O_{0.03}) was deliberately made more hyperstoichiometric, resulting in a higher Widmanstätten precipitate of UC₂ (Figure 3). A precipitate in the grain boundaries in the etched specimen after the thermal diffusivity measurements is probably secondary sesquicarbide (U₂C₃).

UCON-288 (U_{0.495}C_{0.355}O_{0.15}), UCON-459 (U_{0.495}C_{0.345}O_{0.16}), and UCON-289 (U_{0.495}C_{0.355}O_{0.17}) are two-phase UC_xO_y and UO₂ with traces of free uranium (Figures 4 and 5). The UO₂ is the light grey phase, and the free uranium is the white phase in the unetched micrographs. UO₂ appears white in the etched specimens.

FIGURE 1. Phases and Phase Relations in the U-C-O System at 1700 °C, Showing Composition of Uranium Oxycarbide Samples

Neg 0673132-1

300X

FIGURE 2. Microstructure of $U_{0.495}C_{0.485}O_{0.02}$ (UCON 283) Before and After Thermal Diffusivity Measurements to 1500 °C. The oxycarbide is single phase with a trace of free uranium (white phase). UCON 458 exhibited similar structure.

Neg 0673132-2

300X

FIGURE 3. Microstructure of $U_{0.48}C_{0.49}O_{0.03}$ (UCON 365)
Before and After Thermal Diffusivity Measurements
to 1500 °C. The oxycarbide contains a second
phase of U_2C_3 (white phase at grain boundaries)
and a trace of UO_2

Neg 0673132-3

300X

FIGURE 4. Microstructure of $U_{0.495}C_{0.355}O_{0.15}$ (UCON288)

Before and After Thermal Diffusivity Measurements to 1500 °C. The oxycarbide is two phase with minor phase of UO_2 (grey phase in as-polished and white phase in etched surfaces) and a trace of free uranium (a white phase in as-polished surface). Black areas are pores.

$U_{0.495}C_{0.345}O_{0.016}$ (UCON-459) and $U_{0.495}C_{0.335}O_{0.17}$ (UCON 289) exhibited similar structures.

Neg 0673132-4

300X

FIGURE 5. Microstructure of $U_{0.495}C_{0.335}O_{0.17}$ (UCON 289) Before and After Thermal Diffusivity Measurements to 1500 °C. The oxycarbide is two phase with minor phase of UO_2 (grey phase in as-polished surface and white phase in etched surface) and a trace of free uranium (white phase in as-polished surface). Black areas are pores.

The as-fabricated cylinders were cut into thin disks (about 0.100 cm thick and 0.635 cm in diameter) for the thermal diffusivity measurements. Larger specimens (UCON-458 and UCON-459) for electrical resistivity measurements were machined into right cylinders 0.635 cm diameter and 1.91 cm long.

THERMAL DIFFUSIVITY MEASUREMENTS

Thermal diffusivity was measured from 100 to 1500 °C using a laser-pulse technique.^(5,6) The pulse was provided by a 0.5 in. diameter ruby laser with energy output of 3 to 7 Joules. The pulse width was 0.54 msec.

The temperature transients on the back surfaces of the specimens were measured optically using a liquid nitrogen cooled, indium antimonide infrared detector. The signal from the detector was displayed on an oscilloscope and recorded on film. Corrections were made for heat losses using the method proposed by Cowan.^(7,8) No pulse time corrections were required.

The specimens were heated in a vertical tungsten tube furnace which was heated by direct current resistance. The sample was held in a UO₂ holder with small tungsten pins.

Temperatures were measured using a Pt versus Pt-13% Rh thermocouple positioned in the holder. This thermocouple was calibrated using another Pt Pt-13% Rh thermocouple welded to a thin platinum disk set in the sample position. Temperatures were controlled manually.

Measurements were made in purified argon; the inlet argon contained less than 1 ppm oxygen and less than 5 ppm H₂O. The pressure in the furnace was one atmosphere.

Each specimen was heated initially to 1000 °C, and thermal diffusivity measurements made during the increase and decrease

in temperature. Each sample was removed from the furnace to check for possible reaction with the tungsten or UO_2 . The specimen was then reinserted in the furnace, and thermal diffusivity measurements were made to approximately 1500 °C. No reactions between the samples and the UO_2 or tungsten of the holder were observed.

The thermal diffusivity apparatus was calibrated using ARMCO iron (see Appendix B). The thermal diffusivity data for the iron are in satisfactory agreement with the data reported by Cody et al.,⁽⁹⁾ Godfrey et al.,⁽¹⁰⁾ and Shanks et al.⁽¹¹⁾

ELECTRICAL RESISTIVITY MEASUREMENTS

Electrical resistivity was measured using a four-probe technique which has been successfully used to measure the electrical conductivity of oxides.⁽¹²⁾ Measurements were made in purified argon at atmospheric pressure. The inlet argon contained less than 1 ppm oxygen and less than 5 ppm water.

RESULTS AND DISCUSSION

Thermal diffusivity data for uranium oxycarbide from 100 to 1500 °C are shown in Figure 6.* The thermal diffusivity (α) of the oxycarbides decreased with increasing oxygen content. This change was most pronounced at the lower temperatures. The diffusivities of the samples containing 15 to 17 at.% oxygen are approximately 60% of that for the samples with 2 at.% oxygen. This difference decreases as the temperature increases.

* *The thermal diffusivity and conductivity data for UC_xO_y are tabulated in Appendix C.*

FIGURE 6. Thermal Diffusivity of Uranium Oxycarbides

The temperature dependence of the thermal diffusivity varied significantly with oxygen content. The oxycarbides with the lowest oxygen concentration showed a decrease in α as the temperature increased. In contrast, the oxycarbides with the highest oxygen concentration exhibited a slight increase in α with an increase in temperature. The overall result is that the diffusivities of all the specimens approach a common value near $0.05 \text{ cm}^2/\text{sec}$ at the higher temperatures.

Thermal conductivities of the uranium oxycarbides* were calculated using heat capacity data for uranium monocarbide. Since the preliminary heat content data of UC_xO_y do not differ significantly from that of uranium monocarbide,⁽¹³⁾ the heat capacity data reported by Godfrey et al.⁽¹⁴⁾ were used for these calculations. Results are shown in Figure 7.

The thermal conductivities of the uranium oxycarbides are in fair agreement with the values reported by other investigators for stoichiometric UC with comparable oxygen content (see Appendix A). Comparisons of the thermal diffusivity and conductivity for UC_xO_y can best be made using data reported for uranium monocarbides. An analysis must consider not only the oxygen content but also the nitrogen and carbon contents of the monocarbide. It is apparent from this survey that many of the so-called uranium monocarbides actually contain oxygen equivalent to or greater than 2 at.%. Since a few hundred ppm oxygen is equivalent to a significant percent of the total atoms in the carbide (1 at.% is equivalent to approximately 1250 ppm of oxygen), small quantities of oxygen should be expected to affect the thermal conductivity.

* The thermal diffusivity and conductivity data for UC_xO_y are tabulated in Appendix C.

FIGURE 7. Thermal Conductivity of Uranium Oxycarbides Calculated from Thermal Diffusivity Data and Heat Capacity of Uranium Monocarbide

The calculated thermal conductivity values are compared with some selected conductivity values for stoichiometric uranium monocarbides and oxycarbides in Figure 8. Agreement with the values for uranium carbides containing comparable amounts of oxygen is good. The data of Warren and Lacis⁽¹⁹⁾ for an oxycarbide containing 5.3 at.% oxygen falls between the two values for the oxycarbides with 2 at.% and 16 at.%.

The data of Sobon et al.⁽¹⁷⁾ and Hayes and DeCrescente⁽¹⁸⁾ exhibit a slight negative temperature coefficient in the same temperature range, with values lower than those for uranium monocarbide. The variations in thermal conductivity are too large to be explained by density differences.

ELECTRICAL RESISTIVITY

The electrical resistivity for $U_{0.495}C_{0.485}O_{0.02}$ and $U_{0.495}C_{0.345}O_{0.16}$ increases with increases in temperature (Figure 9).^{*} The electrical resistivity for the oxycarbide containing 16 at.% is approximately 35% higher than the resistivity of the oxycarbide containing 2 at.% oxygen. The latter oxycarbide also exhibited a slight decrease in resistivity at the lower temperatures after being heated to 1775 °K. No change after heating was observed for the 16 at.% oxycarbide.

The electrical resistivity of the $U_{0.495}C_{0.485}O_{0.02}$ is very close to the resistivities reported for stoichiometric uranium monocarbide (Figure 10). The electrical resistivity for UC is reviewed in Appendix A. Sobon et al.⁽¹⁷⁾ and Hayes and DeCrescente⁽¹⁸⁾ reported no significant increase in resistivity for UC with an increase in combined oxygen and nitrogen contents

^{*} The electrical resistivity data for UC_xO_y are tabulated in Appendix D.

FIGURE 8. Thermal Conductivity of Uranium Oxycarbide Compared with Some Selected Conductivity Values for Uranium Monocarbide (See Appendix A)

FIGURE 9. Electrical Resistivity of Uranium Oxycarbide

FIGURE 10. *Electrical Resistivity of Uranium Oxycarbide Compared with Some Selected Resistivity Values for Uranium Monocarbide (See Appendix A).*

less than approximately 1.75 at.%. However, when the oxygen plus nitrogen content was increased to approximately 3.1 at.% or above, increases in resistivity were observed. (The oxygen content excluding nitrogen was 1 at.% and 2 at.%, respectively.) The present results support these previous observations since the resistivity of the $U_{0.495}C_{0.485}O_{0.02}$ is very close to the values for UC containing significantly less oxygen.* If, in addition, the resistivities were corrected to a common density, the resistivities would agree more closely with the data of Carniglia,⁽¹⁵⁾ and Mustacchi and Guiliani.⁽¹⁶⁾

The slope of the resistivity versus temperature curve for $U_{0.495}C_{0.485}O_{0.02}$ is slightly less positive than that for UC. However, the slight increase in slope for the resistivity on cooling makes it agree more closely with the UC data. This suggests that some changes occurred in the oxycarbide at the higher temperatures. However, no variations in the microstructures before and after heating were observed.

The results of this study agree qualitatively with some preliminary electrical property measurement⁽¹⁹⁾ of uranium oxycarbides with oxygen contents between 5 and 7.5 at.% (Figure 10).

CONDUCTION PROCESSES

Heat conduction in carbides occurs partially by electrons and partially by lattice vibrations (phonons). Previous investigators^(16,18,23,25,26) have suggested that mixed conduction occurs in uranium monocarbide below 1000 °C; above 1000 °C conduction is essentially 100% electronic.

* See Appendix A.

The total thermal conductivity can be expressed as

$$\lambda_T = \lambda_\ell + \lambda_e \quad (1)$$

where λ_T is the total thermal conductivity, and λ_ℓ and λ_e are the lattice and electronic conductivities, respectively. The phonon conductivity in mixed conductors is difficult to measure directly. However, the electronic conductivity can be estimated by means of the Wiedemann-Frantz relationship if the electrical resistivity is known.

The total conductivity can also be expressed as

$$\lambda_T = \frac{A}{T} + \frac{LT}{\rho} \quad (2)$$

where A is a constant, T is the absolute temperature, L is the Lorenz number, ρ is the electrical resistivity, and LT/ρ represents the electronic conductivity.

To evaluate the data, a special curve-fitting program developed at Oak Ridge National Laboratory was employed to analyze the data.⁽²⁷⁾ A smooth curve for λ versus T was obtained for $U_{0.495}C_{0.335}O_{0.17}$, $U_{0.495}C_{0.355}O_{0.15}$, and $U_{0.495}C_{0.485}O_{0.02}$ using least squares analysis of data. Curves for the 15 and 17 at.% samples were very close and were treated as one curve with a nominal composition of $U_{0.495}C_{0.345}O_{0.16}$.

Preliminary calculations of λ_e using a theoretical Lorenz number (L_0) of $2.443 \times 10^{-8} \text{ (V/}^\circ\text{C)}^2$ ($\lambda_e = \frac{L_0 T}{\rho}$) indicated that λ_e was less than λ_T but approached λ_T at the higher temperatures.

Smooth curves for ρ versus T were obtained for $U_{0.495}C_{0.485}O_{0.02}$ and $U_{0.495}C_{0.345}O_{0.16}$ (on heating) and $U_{0.495}C_{0.345}O_{0.16}$ (On cooling).

If Equations (1) and (2) are valid for the oxycarbides, then

$$\lambda_T T = \frac{LT^2}{\rho} + A \quad (3)$$

A plot of λT versus $\frac{T^2}{\rho}$ would be linear, and L and A can then be determined. The results show that for

$U_{0.495}C_{0.485}O_{0.02}$

$$L = 2.71 \times 10^{-8} \text{ (V/}^\circ\text{C)}^2$$

$$A = 11.4 \text{ (W/}^\circ\text{C)}$$

and for $U_{0.495}C_{0.345}O_{0.16}$

$$L = 1.98 \times 10^{-8} \text{ (V/}^\circ\text{C)}^2$$

$$A = 54.1 \text{ (W/}^\circ\text{C)}$$

The $\lambda_T T$ versus T^2/ρ curve for $U_{0.495}C_{0.485}O_{0.02}$ had a break in the curve near $T^2/\rho = 3000$ and values of T^2/ρ less than 3000 were ignored.

Seven curve-fitting methods (computerized and least squares) were fitted to the smoothed data for $U_{0.495}C_{0.345}O_{0.16}$ and $U_{0.495}C_{0.485}O_{0.02}$ (both heating and cooling). The following equations were considered

$$\lambda = \frac{LT}{\rho} + \frac{C}{\rho} + \frac{A}{T} \quad (4)$$

The first two terms allow for an L that decreases with an increase in temperature and the last term for a lattice conduction that decreases with increasing temperature.

$$\lambda = \frac{LT}{\rho} + \frac{CB}{\rho} + \frac{A}{T} + \frac{B}{T^2} \quad (5)$$

This equation is similar to Equation (4) except an additional term $\frac{B}{T^2}$ is added to the lattice condition contribution.

$$\lambda = \frac{L_{\max} T}{\rho} [1 - \exp(-\frac{T}{\theta}) + 0.2138] + \frac{A}{T} + \frac{B}{T^2} \quad (6)$$

The first term is an approximation to the theoretically expected behavior for the Lorenz function. The term $\frac{B}{T^2}$ is chosen to give the minimum standard deviation

$$\lambda = \frac{L_{\max} T}{\rho} [1 - \exp(-\frac{T}{\theta}) + 0.2138] + \frac{A}{T} \quad (7)$$

This equation is same as Equation (6) with only a single $\frac{A}{T}$ lattice conduction term.

$$\lambda = \frac{L_{\max} T}{\rho} [1 - \exp(-\frac{T}{\theta}) - 0.2138] \quad (8)$$

This equation employs no lattice thermal conduction term.

$$\lambda = \frac{LT}{\rho} + \frac{A}{T} + \frac{B}{T^2} \quad (9)$$

This equation employs a constant Lorenz number, not necessarily equal to L_0 and two lattice conduction terms

$$\lambda = \frac{LT}{\rho} + \frac{A}{T} \quad (10)$$

This equation is similar to Equation (9) but uses only a single lattice conduction term.

The results of this analysis are summarized in Tables 2 and 3.

For each material, the Lorenz number ($L, L_{\max}$) and also the A for all equations are very nearly the same. The increase in the oxygen content increases the value of L , where L for $U_{0.495}C_{0.485}O_{0.02}$ is less than the theoretical Lorenz number (L_0) [$2.443 \times 10^{-8} (V/^{\circ}C)^2$] which is less than the L for $U_{0.495}C_{0.345}O_{0.16}$. The increase in oxygen might be analogous to the increase in an alloying element in pure metals, where a similar increase in L is expected.

TABLE 2. *Standard Deviation of Data of Equations (Percent)*

Equation Number	$U_{0.495}C_{0.485}O_{0.02}$		$U_{0.495}C_{0.345}O_{0.16}$
	(On Heating)	(On Cooling)	
4	Fails: negative lattice term	Fails: negative lattice term	1.1
5	0.68	0.58	Fails: negative electronic term
6	0.65	0.54	0.93
7	4.3	3.1	0.97
8	Fails	Fails	Fails
9	0.74	0.63	0.97
10	4.82	3.6	1.2

TABLE 3. *Analysis of Electronic and Lattice Thermal Conductivity of Uranium Oxycarbide*

	$U_{0.495}C_{0.485}O_{0.02}$	$U_{0.495}C_{0.385}O_{0.02}$	$U_{0.495}C_{0.345}O_{0.16}$
	(On Heating)	(On Cooling)	
<u>Equation (6)</u>	$\lambda = \frac{A}{T} + \frac{B}{T^2} + \frac{L_{\max} T}{\rho} [1 - \exp(-\frac{T}{\theta})] + 0.2138]$		
A	77.54	66.05	15.95
B	-9602.1	-7111.0	-441.8
$L_{\max}$	1.832×10^{-8}	1.918×10^{-8}	2.672×10^{-8}
Standard Deviation %	0.54	0.65	0.93
θ	170 °K	150 °K	80 °K
<u>Equation (9)</u>	$\lambda = \frac{A}{T} + \frac{B}{T^2} + \frac{LT}{\rho}$		
A	75.86	65.51	16.76
B	-10,320.0	-8018.0	-856.4
L(constant)	1.860×10^{-8}	1.934×10^{-8}	2.665×10^{-8}
Standard Deviation %	0.74	0.63	0.97
<u>Equation (10)</u>	$\lambda = \frac{A}{T} + \frac{LT}{\rho}$		
A	54.1		11.4
L	1.98×10^{-8}		2.71×10^{-8}
Standard Deviation %	4.82		1.2

The increase in oxygen content also increases the lattice resistance (as indicated by the decrease in A) which is indicative of increased phonon scattering with increased oxygen additions.

When Equation 9 is used, the electronic conductivity at 200 °C for $U_{0.495}O_{0.485}O_{0.02}$ and $U_{0.495}C_{0.345}O_{0.16}$ represents 50% and 80% of λ_T , respectively; at 1500 °C the electronic conductivity represents 80% and 95% of λ_T , respectively.

Although the analysis of the electrical and thermal conductivities exhibited some quantitative limitations, there is little doubt that mixed conduction does occur in the uranium oxycarbides. It is also apparent from the analysis that the presence of oxygen in the lattice affects the lattice (phonon) conductivity significantly more than it affects the electronic conductivity. The results indicate that although electronic conduction is predominant above 1000 °C, the lattice conduction is still significant, especially in the oxycarbides containing small amounts of oxygen.

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APPENDICES

APPENDIX A

REVIEW OF THERMAL CONDUCTIVITY AND ELECTRICAL
RESISTIVITY OF URANIUM MONOCARBIDE

The thermal conductivity and electrical resistivity data of nearly stoichiometric UC were reviewed to evaluate possible effects of combined oxygen. Tables A-1 and A-2 list the investigators and references, the compositions, the reported oxygen and nitrogen levels, the densities, the temperature ranges of the measurements, and other information concerning the uranium carbides used by these investigators. The results are also plotted as a function of temperature in Figures A-1 and A-2 with no correction being made for density differences. The review is concerned primarily with stoichiometric UC containing 4.7 to 4.9 wt% carbon. No attempt has been made to evaluate the properties of hypo- or hyperstoichiometric carbides.

THERMAL CONDUCTIVITY DATA

The thermal conductivity (λ) of nearly stoichiometric UC appears to decrease rapidly from room temperature to 500 °C, approaching a constant thermal conductivity value above 500 °C. Above 1200 °C, the thermal conductivity is relatively independent of temperature with a value of approximately 0.17 W/cm-°C.

The scatter in the data at temperatures below 1000 °C is approximately ± 0.065 W/cm-°C [excluding data of Mihailovic,⁽²⁾ Howard,⁽¹⁾ and Taylor⁽¹⁹⁾]. The scatter above 1000 °C is significantly less, ± 0.013 W/cm-°C.

The following conclusions have been made concerning the thermal conductivity of nearly stoichiometric UC.

- (1) λ exhibits only a small temperature coefficient above 1000 °C^(3,7,8,12-15,18) with a value at 1000 °C of approximately 0.17 W/cm-°C.

TABLE A-1. Summary of Thermal Conductivity for UC

Investigator	Ref.	Carbon Content, wt%	Oxygen Content, ppm	Nitrogen Content, ppm	Density, g/cm ³	Percent TD	Temperature Range, °C	Measured Thermal Conductivity, ¹ W/cm-°C	Measuring Method	Additional Comments
Howard	1	NR	NR	NR	10.03- 11.07	74-81	60	0.134	Steady State Cut Bar	5 points only
Accary and Caillat	2	NR	NR	NR	13.3	98	119-236	0.180-0.197	NR	4 points only; hot pressed UC
Secrest et al.	3	5.2	NR	NR	13.63	100	100-733	0.255-0.222	Steady State Axial Flow	arc cast UC
Chubb and Dickerson	6	4.8-5.3	NR	NR	12.5- 12.8	98	100-1000	0.230	Steady State	Sintered and arc cast UC
Mustacchi and Guiliani	7	4.8	90	500	13.63	100	1100-1700	0.181	Thermal Diffusivity	arc cast UC; $\alpha = 0.049 \pm$ $0.004 \text{ cm}^2/\text{sec};$ $C_p = 0.065$ $\text{cal/g-}^\circ\text{C}$
Grossman	8	5.3	NR	NR	13.6	100	875-1775	0.239 ± 0.008	Special Technique	λ temperature independent; arc cast UC
Dumas and Mansard	9	4.72- 4.85	600	200	13.46	99	20-600	0.322-0.203	Steady State	
Kubota et al.	10	4.8	NR	NR		95	125-233	0.149-0.205	Steady State Axial Flow	4 points only
Leary et al.	11	4.8	NR	NR	13.6	100	300-460	0.224-0.228	Steady State Axial Flow	
Wheeler	12	4.8- 5.1	NR	NR	13.6	94-100	1187-2007	0.139-0.190	Thermal Diffusivity	Assumed $C_p = 0.05$ $\text{cal/g-}^\circ\text{K}$ at all temperatures
Carniglia	13	4.04- 4.50	200-400	200-400	13.6	100	100-100	~ 0.234	Thermal Diffusivity	$C_p = 0.053 \pm$ $0.003 \text{ cal/cm-}^\circ\text{C};$ arc cast UC

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TABLE A-1. (contd)

Investigator	Ref.	Carbon Content, wt%	Oxygen Content, ppm	Nitrogen Content, ppm	Density, g/cm ³	Percent TD	Temperature Range, °C	Measured Thermal Conductivity, W/cm-°C	Measuring Method	Additional Comments
Sobon et al.	14	4.63- 4.85	1250- 6800	93-1400	11.8- 12.9	86.5- 95	880-1600	0.176-0.159 (4.85 wt% C)	Steady State Radial Flow	
DeCrescente	15	4.85	1250	NR	12.4	91	880-1440	0.189-0.175	Steady State Radial Flow	
Wittenberg and Grove	16	4.78	NR	NR	13.10	96	250-1000	0.303 ± 0.046- 0.163 ± 0.017	Thermal Diffusivity	Cp measured; UC contained 0.1 wt% Ni, U ₂ C ₃ , UO ₂ and UC ₂
Hayes and DeCrescente	18a 18b	4.8	860 860- 2000	520 520- 1400	12.9	95	110-1600 880-1600	0.175 ± 0.005	Steady State Radial Flow	Excellent Review
Taylor et al.	19	NR	NR	NR	12.9	95	30	0.127	Thermal Diffusivity	λ independent of T; Cp = 0.048 cal/ g-°C; UC contained 0.1 wt% Ni
Moser and Kruger	20	4.8	50	NR	13.6	99+	23-600	0.251-0.167	Thermal Diffusivity	Cp measured
Warren and Lacis	21a 21b	5.52 3.58	4060 7500	NR NR	NR NR	NR NR	800-1500 900-1300	0.150-0.176 0.145-0.164	Steady State Steady State	High Oxygen Carbides
Mihalovic ²	23	4.85	600	200	13.46	99	450-1200	0.235-0.088	Steady State	

1. Thermal conductivity calculated from thermal diffusivity data $\lambda = \alpha \cdot C_p \cdot \rho$ using constant $C_p = 0.065$ cal/g-°C, $\rho = 13.6$ g/cm³ and $\alpha = 0.049$ cm²/sec. Mustacchi and Guilian⁽⁷⁾ originally used value of C_p much lower than presently accepted value.

2. Data of Mihalovic reported by Dumas and Mansard.⁽⁹⁾

TABLE A-2. *Summary of Electrical Resistivity of UC*

Investigator	Ref.	Carbon Content, wt%	Oxygen Content, ppm	Nitrogen Content, ppm	Density, g/cm ³	Percent TD	Temperature Range, °C	Measured Electrical Resistivity μohm-cm at Lowest and Highest Temperature	Additional Comments
Accary and Caillat	2	NR	NR	NR	13.3	98	40-920	20-98	
Dayton and Tipton	4	2.2- 9.2	NR	NR	12.5- 12.8	92-94	60	41	ρ as function of car- bon content
Griffiths	5	4.8	Varied	NR	12.4	91	375-1000	100-161	[0] changed during measurement
Mustacchi and Guiliani	7	4.8	90	500	13.6	100	23-1200	60-208	2 ppm H ₂
Grossman	8	5.3	NR	NR	13.6	100	925-1775	160-260	
Leary et al.	11, 22	4.8	NR	NR	13.6	100	60	69	Single measurement
Carniglia	13	4.15- 9.1	180	NR	13.6	100	23-1100	45-185 (4.80 wt% C)	
Sobon et al.	14	4.67- 4.77	1650- 5600	170- 5600	10.7- 12.6	79-93	21-1308	43-225 (4.77 wt% C 93% TD)	
Costa and Lallement	17	4.8	NR	NR	NR	NR	-269-925	47-24 °C	
Hayes and DeCrescente	18	4.8	815- 2100	150- 1200	11.2- 12.4	82-91	23-1300	43-215 (91.4 % TD)	Excellent Review
Warren and Lacis	21	3.58- 5.52	4060- 7500	NR	NR	NR	375-130		

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FIGURE A-1. A Summary of the Thermal Conductivities of Uranium Monocarbide

FIGURE A-2. A Summary of the Electrical Resistivities of Uranium Monocarbide

- (2) The thermal conductivity increases with an increase in carbon content.^(6,13) Wheeler is the only investigator to observe the opposite effect.⁽¹²⁾
- (3) The thermal conductivity decreases as the oxygen content in the UC increases.^(14,15,18,21) Sobon et al.,⁽¹⁴⁾ DeCrescente and Miller,⁽¹⁵⁾ and Hayes and DeCrescente⁽¹⁸⁾ indicate that the effect of combined oxygen and nitrogen in UC (below 2180 ppm) is minor, especially at the higher temperatures, with a general lowering of the conductivity when the oxygen was increased from 1380 to 4000 ppm. The data suggest that much of the wide scatter in thermal conductivity values at low temperatures could possibly result from variations in oxygen content in UC.
- (4) The portion of thermal conductivity due to an electronic process was observed to approach 100% above 1000 °C.^(7,8,13-16,18)

ELECTRICAL RESISTIVITY

The electrical resistivity of UC increases with increasing temperature. With the exception of the data reported by Accary and Caillat⁽²⁾ and Leary et al.^(11,22) the data fall within a scatter of $\pm 8.5 \mu\text{ohm-cm}$.

The following conclusions were made concerning the electrical resistivity of nearly stoichiometric UC:

- (1) The electrical resistivity increases with temperature and is approximately a linear function of temperature above 500 °C. The temperature coefficient is greater below 500 °C.
- (2) The resistivity increases with increasing carbon content.^(4,13) A slight increase is observed from pure uranium to 4.8 wt% carbon and a logarithmic increase from 4.8 to 9.1 wt% carbon.

- (3) The electrical resistivity increases with increases in both oxygen content and combined oxygen nitrogen content.^(14,18,21) Hayes and DeCrescente⁽¹⁸⁾ indicated that resistivity is relatively insensitive to a combined oxygen and nitrogen content to 2300 ppm. However, effects are observed when the oxygen content is significantly larger.⁽²¹⁾
- (4) The resistivity varies with density by the relationship

$$\rho_{HD} = \rho_{LP} (1 - P)^{1.38}$$

where ρ_{HD} is the high density, ρ_{LD} is the low density and P is the porosity fraction.⁽¹⁸⁾

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APPENDIX B

CALIBRATION OF THERMAL DIFFUSIVITY APPARATUS

The accuracy of the thermal diffusivity values and of the pulse technique was evaluated by measuring the thermal diffusivity of Armco iron. Armco iron is considered by many investigators to be a thermal conductivity standard. Recent reviews of the published thermal conductivity data for Armco iron between room temperature and 760 °C indicate a data spread of about 4%.⁽¹⁾

The thermal diffusivity was determined for Armco iron samples of two thicknesses (0.140 and 0.071 cm). Measurements were made during three temperature cycles from 25 to less than 800 °C. The results are illustrated in Figures B-1 and B-2, and tabulated in Table A-1.

Two investigators^(2,3) have measured the thermal diffusivity of Armco iron. These data are the basis for the accuracy evaluations of the pulse technique. The average values of the thermal diffusivity using data of Cody et al.,⁽²⁾ Shanks et al.,⁽³⁾ and this study were used to determine the percent deviation from room temperature to 765 °C (Figure B-3). With the exception of two data points, the data fall within $\pm 3.5\%$ of the average. The data of Cody et al. exhibit a deviation of $\pm 3.0\%$ with all values falling below the average. The data of Shanks et al., show a deviation of $\pm 2.0\%$ with all values above the average.

FIGURE B-1. Thermal Diffusivity of Armco Iron from 200 to 850 °C
Compared with Reported Thermal Diffusivity Data

FIGURE B-2. Thermal Diffusivity of Armco Iron from 680 to 840 °C

TABLE B-1. Thermal Diffusivity of Armco Iron

<u>Temperature,</u> <u>°C</u>	<u>Thermal Diffusivity,</u> <u>cm²/sec</u>	<u>Temperature,</u> <u>°C</u>	<u>Thermal Diffu-</u> <u>sivity, cm²/sec</u>
0.071 cm Thick			
615	0.0620	772	0.0436
690	0.0497	780	0.0451
697	0.0480	781	0.0463
705	0.0483	785	0.0461
714	0.0455	797	0.0478
721	0.0452	799	0.0476
724	0.0444	804	0.0494
734	0.0415	807	0.0490
746	0.0386	815	0.0505
763	0.0346	819	0.0500
765	0.0323	823	0.0501
751	0.0370		
735	0.0410		
733	0.0428		
0.140 cm Thick			
210	0.01386	655	0.0575
366	0.01047	680	0.0515
460	0.0890	685	0.0550
608	0.0655	700	0.0490

FIGURE B-3. *Percent Deviation of Reported Thermal Diffusivity Data*

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APPENDIX C

THERMAL DIFFUSIVITY/CONDUCTIVITY OF URANIUM OXYCARBIDE*TABLE C-1. Thermal Diffusivity/Conductivity
Data of Uranium Oxycarbide*

SPECIMEN UCON-283		U _{0.495} C _{0.485} O _{0.02}
Temperature, °C	Thermal Diffusivity, cm ² /sec	Thermal Conductivity, W/cm-°C
101	0.0723	0.194
408	0.0639	0.191
512	0.0614	0.187
612	0.0596	0.184
672	0.0597	0.185
800	0.0569	0.179
808	0.0568	0.180
660	0.0598	0.185
395	0.0625	0.185
997	0.0559	0.181
1002	0.0546	0.177
1206	0.0564	0.181
1297	0.0512	0.178
1393	0.0521	0.182
1405	0.0546	0.191
1500	0.0530	0.189
1400	0.0544	0.190
1302	0.0547	0.187
1103	0.0530	0.175
1002	0.0558	0.181

SPECIMEN UCON-288

 $U_{0.495}C_{0.355}O_{0.15}$

Temperature, °C	Thermal Diffusivity, cm ² /sec	Thermal Conductivity, W/cm-°C
0.0464	0.128	185
0.0458	0.132	334
0.0457	0.134	400
0.0452	0.135	517
0.0443	0.134	623
0.0447	0.137	711
0.0438	0.136	800
0.0455	0.144	918
0.0449	0.143	998
0.0441	0.139	894
0.0451	0.140	810
0.0442	0.135	674
0.0442	0.133	564
0.0444	0.130	424
0.0453	0.128	285
0.0447	0.124	228
0.0453	0.130	270
0.0462	0.148	996
0.0459	0.149	1102
0.0468	0.155	1196
0.0465	0.157	1301
0.0455	0.156	1402
0.0475	0.163	1401
0.0446	0.147	1202
0.0453	0.147	1109
0.0452	0.144	1000

SPECIMEN UCON-289

 $U_{0.495}C_{0.335}O_{0.017}$

Temperature, °C	Thermal Diffusivity, cm ² /sec	Thermal Conductivity, W/cm-°C
218	0.0429	0.119
360	0.0440	0.126
445	0.0448	0.131
560	0.0439	0.130
660	0.0447	0.134
785	0.0445	0.136
890	0.0454	0.140
998	0.0455	0.143
896	0.0454	0.140
796	0.0437	0.133
706	0.0439	0.132
604	0.0440	0.131
503	0.0439	0.128
408	0.0429	0.123
794	0.0449	0.137
885	0.0466	0.144
1000	0.0473	0.149
980	0.0463	0.145
1071	0.0463	0.147
1194	0.0465	0.151
1287	0.0481	0.159
1407	0.0462	0.156
1400	0.0489	0.165
1297	0.0461	0.153
1202	0.0456	0.148
1001	0.0466	0.147

SPECIMEN UCON-365

 $U_{0.48}C_{0.49}O_{0.03}$

Temperature, °C	Thermal Diffusivity, cm ² /sec	Thermal Conductivity, W/cm-°C
133	0.0563	0.162
333	0.0535	0.163
530	0.0521	0.164
542	0.0528	0.167
671	0.0507	0.162
790	0.0514	0.167
844	0.0507	0.166
891	0.0506	0.167
1101	0.0498	0.169
1200	0.0498	0.173
1302	0.0507	0.179
1401	0.0506	0.182
1501	0.0493	0.181
1298	0.0490	0.173
1204	0.0493	0.171
419	0.0542	0.166
1225	0.0512	0.179
1495	0.0505	0.186
1373	0.0490	0.175
746	0.0525	0.169
421	0.0546	0.167
195	0.0563	0.162

APPENDIX D
ELECTRICAL RESISTIVITY OF URANIUM OXYCARBIDE

TABLE D-1. *Electrical Resistivity Data of Uranium Oxycarbide*

SPECIMEN UCON-458		$U_{0.495}C_{0.485}O_{0.02}$	
Temperature, °C	Electrical Resistivity, $\mu\text{ohm-cm}$	Temperature, °C	Electrical Resistivity, $\mu\text{ohm-cm}$
21	89	763	165
21	92	1008	187
21	103	1011	188
21	88	1111	197
21	86	1114	198
28	89	1201	206
39	88	1210	208
49	93	1303	214
101	97	1301	217
118	101	1370	221
175	107	1376	222
201	111	1419	229
263	116	1449	230
334	125	1490	236
354	124	1505	237
433	133	1358	222
464	134	1181	204
475	135	1032	189
469	135	864	172
574	145	638	148
567	146	482	130
665	156	320	114
664	154	240	105
747	163	146	91
		12	73
		12	80

SPECIMEN UCON-459 $U_{0.495}O_{0.345}O_{0.16}$

Temperature, °C	Electrical Resistivity, μohm-cm	Temperature, °C	Electrical Resistivity, μohm-cm
21	114	1264	284
21	110	1347	291
21	106	1341	292
21	105	1420	300
21	104	1425	300
24	106	1491	305
35	107	1494	305
51	110	1425	298
53	110	1418	299
87	121	1260	281
93	125	1242	282
197	142	1025	248
209	141	942	241
334	161	765	223
339	161	828	229
502	185	688	207
507	187	651	203
630	204	485	178
640	204	454	176
791	231	320	161
823	229	205	143
1023	255	166	140
1033	256	77	127
1126	269	42	102
1133	269	23	101
1205	277	23	107
1201	277		
1264	284		

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