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**Electrical and Thermal Transport Properties of  
Uranium and Plutonium Carbides**  
**A Review of the Literature**

by

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## OFFICE MEMORANDUM

TO : Holders of LA-6096

DATE: May 6, 1976

FROM :

SUBJECT : Errata

SYMBOL : ISD-6

Page 10, Table II. Coefficient  $b$  for  $UC_2$  should read  $+0.205$ .

Page 28, Table V. Temperature range for PuC should read  $50 < T^{\circ}C \leq 1300$ .



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# ELECTRICAL AND THERMAL TRANSPORT PROPERTIES OF URANIUM AND PLUTONIUM CARBIDES

A Review of the Literature

by

H. D. Lewis and J. F. Kerrisk

## ABSTRACT

This report reviews the available literature on electrical and thermal transport properties of uranium carbides, plutonium carbides, and uranium-plutonium carbides. Contributions of many authors are outlined with respect to the experimental measurement methods used and characteristics of the sample materials. Discussions treat the qualitative effects of sample material composition; oxygen, nitrogen, and nickel concentrations; porosity; microstructural variations; and the variability in transport property values obtained by the various investigators. Temperature-dependent values are suggested for the electrical resistivities and thermal conductivities of selected carbide compositions based on a comparative evaluation of the available data and the effects of variation in the characteristics of sample materials.

## I. INTRODUCTION

The cumulative literature on electrical and thermal conductivities of UC, PuC, and (U,Pu)C includes several review articles written since 1967. These articles summarize the experimental efforts and present good data summaries. The newest review articles are by Sheth and Leibowitz<sup>1</sup> and Kerrisk<sup>2</sup> whose works are acknowledged in this report.

Most investigators, in reporting the results of their experimental efforts, compare their data with the results of others. Each reviewer and investigator recognizes the importance of several factors affecting experimentally determined transport properties. These factors include fabrication method, chemistry, microstructural and macrostructural effects, irradiation effects, and method of property measurement. Fabrication methods used in processing carbide fuels involve fusion (fusion-solidification and fusion-solidification-pulverization-pressing-sintering) or solid processing (carbothermic reduction-pulverization-pressing-sintering). Chemistry involves stoichiometric effects (uranium/

carbon ratio, plutonium/carbon ratio, and, in the case of mixed carbides, (uranium + plutonium)/carbon ratio); plutonium/uranium ratio for mixed carbide; and impurity content, oxygen, nitrogen, nickel, and/or other elements. Microstructural and macrostructural effects include presence of second phase (fraction of  $MC_2, M_2C_n$ ) and porosity.

This review proposes to

(a) Summarize the results of experimental efforts which have attempted to relate the measured transport properties to one or more of the factors listed above.

(b) Suggest values for the transport properties of the uranium, plutonium, and uranium-plutonium carbides. Selection is made on the basis of material characterization and agreement among investigators. No attempt is made at quantification of the relation between those factors listed above and experimental results of individual investigators.

(c) Point out those areas in which additional experimental work is needed.

Much of the data considered in this report was available only in graphical form. Errors in reading values from curves were unavoidable. Tabular data

or values calculated from the experimenters' functional fit were used where this information was given.

To simplify the consideration of data and information given by individual investigators, this presentation will first consider electrical resistivity and second thermal conductivity. In each section appropriate properties of uranium carbide (UC), plutonium carbide (PuC), uranium-plutonium carbide ((U,Pu)C), and, where possible, related dicarbides and sesquicarbides will be discussed. A short summary will follow each subsection and suggested values for the property will be given at the end of each section.

## II. ELECTRICAL RESISTIVITY

Most of the resistivity measurements on UC, PuC, and (U,Pu)C reported since 1959 were done at temperatures below 1000°C. Essentially no data are available for UC, PuC, and (U,Pu)C above 1800, 1000, or 1500°C, respectively. Data on dicarbides and sesquicarbides at temperatures above 300°K are limited to very few investigations in the uranium-carbon system.

Since 1967, work on electrical and thermal transport properties of these systems has been reviewed by Fulkerson,<sup>3</sup> Moser,<sup>4</sup> Bates,<sup>5</sup> and Leary.<sup>6</sup> Their original work and conclusions will be included in this discussion where applicable.

### A. Uranium Carbide

1. **Fulkerson**,<sup>3</sup> in his 1970 review, appraised the UC data of Costa,<sup>7</sup> Leary,<sup>8</sup> Moser,<sup>4</sup> Hayes,<sup>9</sup> and Rough.<sup>10</sup> Fulkerson's selected resistivity values were stated to be within  $\pm 2 \mu\Omega\text{cm}$  of seven investigators and, at higher temperatures, to follow the values reported by Hayes, Rough, and Lerner.<sup>11</sup> In evaluating these data, Fulkerson's corrections for porosity were made using the Euken equation.<sup>12</sup> His tabulated preferred values are very close to those reported by Rough, and can be represented by the expression

$$\rho = 32.87 + 0.1607T - 2.586 \times 10^{-5} T^2 \mu\Omega\text{cm} \quad (1)$$

for  $100 \leq T^\circ\text{C} \leq 1350$ .

2. **Moser**,<sup>4</sup> in the discussion of several actinide compounds, reviewed the resistivity data of Hayes<sup>9</sup> and Rough,<sup>10</sup> concluding that there was less than 5% difference between hot-pressed UC at 95% theoretical density (TD) and cast material. He corrected the two sets of data to 100% density using the

Maxwell equation;<sup>13</sup> the resulting resistivity values ranged from  $36 \pm 1 \mu\Omega\text{cm}$  at 25°C to about  $165 \pm 5 \mu\Omega\text{cm}$  at 1000°C.

3. **Bates**<sup>5,14</sup> examined the electrical resistivity (and thermal conductivity) of uranium oxycarbides containing 2 to 17 at.% oxygen, and compared his results with those of Sobon,<sup>15</sup> Mustacchi,<sup>16</sup> and Carniglia,<sup>17</sup> and reviewed the results obtained by Accary,<sup>18</sup> Dayton,<sup>19</sup> Griffiths,<sup>20</sup> Grossman,<sup>21</sup> Leary,<sup>8</sup> Costa,<sup>7</sup> and Hayes.<sup>9</sup> Samples were pressed and sintered at 1700°C from powder prepared by reaction sintering  $U + C + UO_2$  powder. Bates' measurements were made using a four-probe technique with the sample under 1-atm argon containing less than 1 ppm oxygen and less than 5 ppm water vapor. Resistivity was determined from room temperature to 1500°C for two specimens which had been well characterized before testing. The data can be summarized as follows.

$U_{0.495}C_{0.485}O_{0.02}$  at 93.8% density (2 at.%, approximately 2500 ppm):

$$\rho = 86.20 + 0.1031T - 2.515 \times 10^{-6} T^2 \mu\Omega\text{cm} \quad (2)$$

$U_{0.495}C_{0.345}O_{0.16}$  at 92.0% density:

$$\rho = 104.18 + 0.1741T - 2.615 \times 10^{-5} T^2 \mu\Omega\text{cm} \quad (3)$$

for  $25 \leq T^\circ\text{C} \leq 1500$ .

Microstructures of the materials containing 2 and 16 at.% oxygen, respectively, exhibited grain boundary traces of  $UC_2$  and free uranium. Second phase concentrations were not measured. In comparison, results of Sobon<sup>15</sup> for approximately stoichiometric UC at 93% TD containing 5600 ppm oxygen and 5600 ppm nitrogen, indicated resistivities of 43 and 225  $\mu\Omega\text{cm}$  at 20 and 1300°C, respectively.

Bates concluded from his review that for nearly stoichiometric UC, resistivity increases with carbon content, increases with temperature (essentially linearly above 500°C), and increases with increasing oxygen (and nitrogen) content. He suggested that the effect of porosity is expressed by

$$\rho_0 = \rho(1 - P)^{1.38} \quad (4)$$

where

$\rho_0$  = resistivity at 100% TD,

$\rho$  = resistivity of low density material,

and

P = volume fraction porosity.

A summary of resistivities for nearly stoichiometric UC, as determined in several of the investigations mentioned above, is given with Bates' data in Fig. 1.

4. Leary<sup>6</sup> reviewed the same resistivity values here as in Ref. 8. A value of 69  $\mu\Omega\text{cm}$  at room temperature was determined for stoichiometric UC, using a comparative eddy current method. No indication of the sample impurity level was given. For purposes of discussion, Leary considered the data of Secrest,<sup>22</sup> which indicates a linear temperature dependence for case UC<sub>1.0</sub>, with respective resistivities at 100 and 900°C of 50 and 150  $\mu\Omega\text{cm}$ .

5. Kamimoto<sup>23</sup> and Pascard<sup>24</sup> have reported two other room-temperature resistivity values. The Kamimoto value of 53  $\mu\Omega\text{cm}$  at 98.5% TD is in good agreement with results of other investigators. Pascard's value of 30  $\mu\Omega\text{cm}$  was obtained from his study of the resistivities of UC-PuC solid solutions. The carbides were formed by reduction of oxides and direct carburization with carbon. The effect of plutonium addition will be discussed in a subsequent section.

6. Hayes and DeCrescente<sup>9</sup> measured the resistivities of four stoichiometric UC specimens of 82, 87.4, 90.1, and 91.4% TD containing 815, 960, 2100, and 1400 ppm oxygen, and 370, 1200, 170, and 150 ppm nitrogen, respectively. The determinations were made over the range 25 to 1300°C both on heating and cooling the sample in flowing argon, using a standard four-probe method. Specimens were

hot pressed from methane synthesized UC powder. A summary of the data is given in Fig. 2.

The data can be represented by the following equations.

82% TD:

$$\rho = 46.85 + 0.2053T - 3.223 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (5)$$

87.4% TD:

$$\rho = 42.02 + 0.1946T - 2.860 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (6)$$

90% TD:

$$\rho = 39.91 + 0.1731T - 2.119 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (7)$$

91.4% TD:

$$\rho = 38.10 + 0.1604T - 1.447 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (8)$$

for  $25 \leq T^\circ\text{C} \leq 1300$ . Hayes and DeCrescente recommend Eq. (4) for porosity correction and suggest that the resistivity is insensitive to oxygen and nitrogen impurity over the range 1100 to 2300 ppm.

Corrected values for the 82 and 91.4% TD material, using Eq. (4), are also plotted in Fig. 2. The corrected data for the 91.4% TD material are within about  $\pm 2 \mu\Omega\text{cm}$  of the preferred values of Fulkerson<sup>3</sup> and the data of Rough<sup>10</sup> shown in Fig. 1. However, the correction to the data for the low density sample is not consistent with the corrected high density data. This perhaps indicates a combined impurity-temperature-pore morphology effect on resistivity.

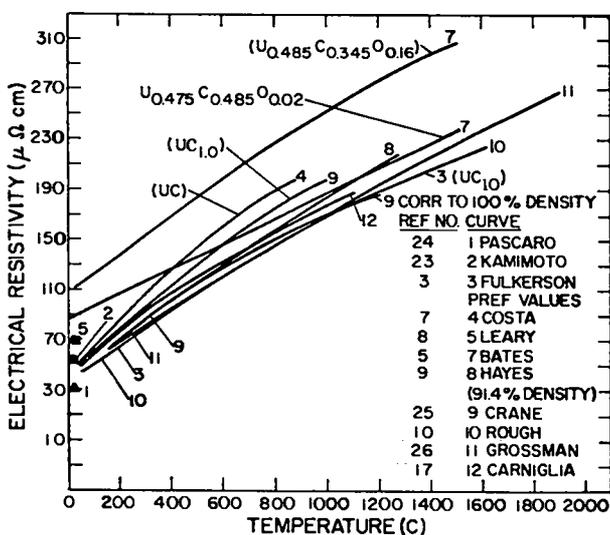


Fig. 1.

Electrical resistivity values for UC (abbreviated summary).

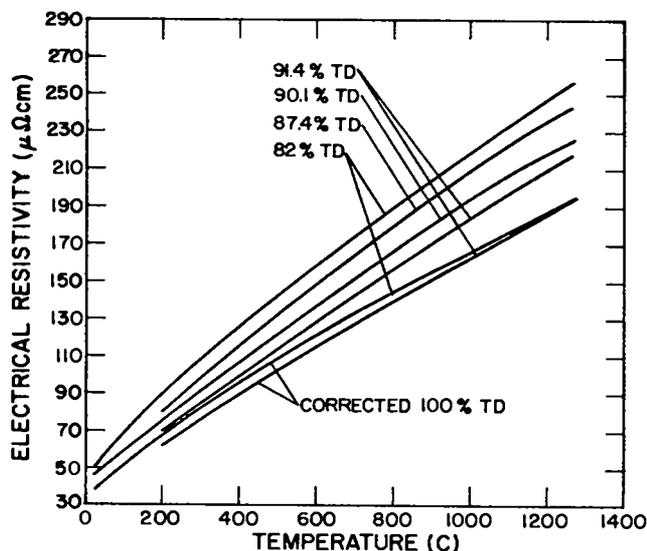


Fig. 2.

Electrical resistivity of UC, according to Hayes.<sup>9</sup>

7. Crane and Gordon,<sup>25</sup> using a dc potentiometric technique, determined the electrical resistivities of specimens prepared for thermal conductivity measurements. The authors estimate experimental error was less than  $\pm 5\%$  to  $500^\circ\text{C}$  and less than  $\pm 10\%$  from  $500$  to  $1000^\circ\text{C}$ . Both cast and sintered materials were examined. Cast materials contained less than 400 ppm oxygen and 100 ppm nitrogen. The pressed and sintered materials contained less than 1000 ppm oxygen and 300 ppm nitrogen. Resistivities were determined from four types of specimens.

- (a) 5.2 wt% carbon, cast, 99% TD,
- (b) 4.8 wt% carbon, sintered, 90% TD,
- (c) 4.4 wt% carbon, cast, 99.8% TD, and
- (d) 4.4 wt% carbon, sintered, 98.1% TD.

Metallographic examination showed free uranium as incomplete grain boundary networks in both cast and sintered hypostoichiometric material, and Widmanstätten  $\text{UC}_2$  precipitate in the hyperstoichiometric material. Second phase concentrations were not measured.

Experimental results are summarized in Fig. 3. The data for the 4.8 wt% carbon sample can be closely represented by

$$\rho = 40.13 + 0.2077T - 5.032 \times 10^{-5} T^2 \mu\Omega\text{cm}, \quad (9)$$

for  $50 \leq T^\circ\text{C} \leq 1000$ . The correction to 100% TD was calculated using Eq. (4). All the data, including the corrected values for the 4.8 wt% sample fall approximately within the experimental error limits stated by the authors, hence perhaps no conclusion should be drawn concerning the effect of carbon concentration on resistivity. However, data for the

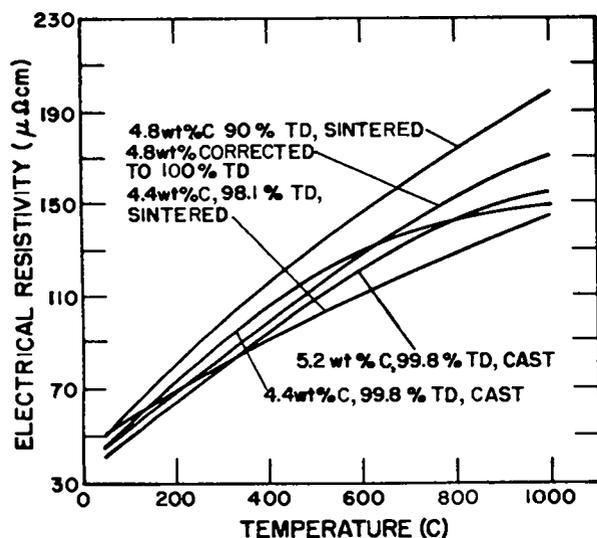


Fig. 3. Resistivity of UC and the effect of variation in carbon concentration, according to Crane.<sup>25</sup>

sintered materials seem to indicate an increase in resistivity with increasing carbon content.

8. Carniglia<sup>17</sup> measured the resistivities of UC specimens containing about 4.1 to 9.1 wt% carbon over the temperature range 25 to  $1000^\circ\text{C}$ , using the dc potentiometric method. Results of these measurements are shown in Fig. 4. Specimens were made from arc-cast rod at 100% TD that have typical impurity levels from 45 to less than 1900 ppm oxygen and from 570 to less than 700 ppm nitrogen. The hypostoichiometric materials contained free uranium in grain boundaries, becoming continuous networks at about 4.3 to 4.4 wt% carbon. The hyperstoichiometric materials contained quenched-in metastable  $\text{UC}_2$  as intragranular platelets.

Carniglia's data for the 4.8 wt% carbon sample are well represented by the expression

$$\rho = 42.29 + 0.1684T - 3.388 \times 10^{-5} T^2 \mu\Omega\text{cm}, \quad (10)$$

for  $50 \leq T^\circ\text{C} \leq 1100$ . His results show increasing resistivity with increasing carbon content. The values for the 9.1 wt% carbon sample over the range 50 to  $400^\circ\text{C}$  are about  $30 \mu\Omega\text{cm}$  higher than those for 4.8 wt% carbon.

9. Rough<sup>10</sup> summarized the BMI resistivity measurements from 25 to  $1600^\circ\text{C}$  on samples prepared from skull arc-cast material containing 4.8, 7.0, and 9.0 wt% carbon. Measurements were by the four-probe potentiometric method. No evaluation of the oxygen and nitrogen impurity level was given. Although densities of the resistivity samples were

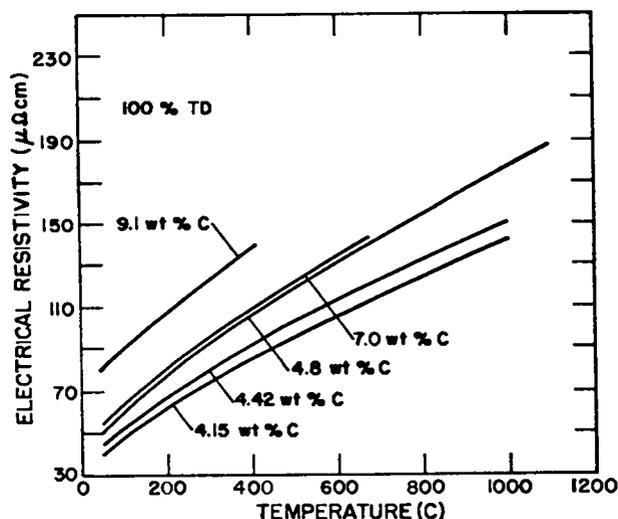


Fig. 4. Resistivity of UC and the effect of variations in carbon concentration, according to Carniglia.<sup>17</sup>

not given, typical values for similar composition used in irradiation tests suggest an average density of about  $13.4 \text{ g/cm}^3$  (98.5% TD) for the 4.8 wt% carbon samples and  $12.63 \text{ g/cm}^3$  (98.3% TD) for the 7.0 wt% carbon samples. Density of the 9.0 wt% carbon material was not reported.

Rough's article gave an excellent discussion of the pre- and postirradiation microstructural study of these materials. Microstructural analysis of the 7.0 wt% carbon composition indicated an intimate mixture of UC + UC<sub>2</sub> in the as-cast condition. Examination of the microstructures of several samples after various heat treatment cycles indicated that the transformation to U<sub>2</sub>C<sub>3</sub> required over 100 h at 1200°C and about 1 h at 1400°C. Therefore, the results of both Rough and Carniglia, for the 7.0 wt% carbon samples, perhaps represent the resistivity of the metastable UC + UC<sub>2</sub> rather than of U<sub>2</sub>C<sub>3</sub>.

The resistivity data reported by Rough are summarized in Fig. 5 with some of the Carniglia data for comparison. Rough commented that the "change in shape of the resistivity and dilation plots for the uranium-9.0 wt% carbon alloy suggests a change in state." The effect is probably related to the observations of Norreys, discussed in a later section.

Rough's data for the 4.8 to 5.3 wt% carbon materials can be expressed as

$$\rho = 33.66 + 0.1623T - 2.752 \times 10^{-5}T^2 \mu\Omega\text{cm}, \quad (11)$$

for  $100 \leq T^\circ\text{C} \leq 1600$ .

Rough's report included a detailed study of irradiation effects on resistivity of these materials. To illustrate the changes in resistivity which may be

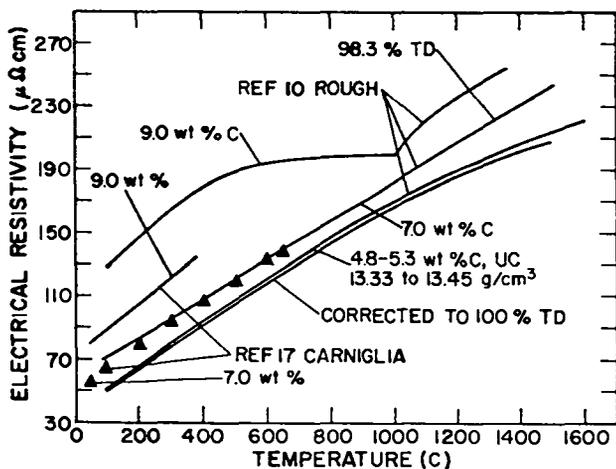


Fig. 5.

Effect of variation in carbon concentration on the resistivity of UC, according to Rough.<sup>10</sup>

expected after irradiation, a very abbreviated data summary is offered in Table I. The increase in electrical resistivity implies a possible corresponding decrease in thermal conductivity under irradiation.

Rough concluded that a "saturation" of point defects that might affect electronic characteristics was attained very early (i.e., at low burnup) during irradiation. The significantly smaller increase in resistivity for specimens irradiated at higher temperatures (i.e., above 400°C) indicated damage annealing during the higher temperature irradiation.

10. Grossman<sup>26</sup> measured the electrical resistivity (and thermal conductivity) of UC, UC<sub>2</sub>, and U<sub>2</sub>C<sub>3</sub> over the temperature range 975°C to about 1800°C. The UC sample material was 100% dense arc-cast rod containing 5.3 wt% carbon and less than 200 ppm oxygen. The UC<sub>2</sub> specimens were made by hot pressing UC<sub>2</sub> powder +1.0 vol% nickel at 1700°C. The UC<sub>2</sub> powder contained 8.7 wt% carbon and 0.3 wt% oxygen. The as-pressed material was approximately 90% TD. Grossman reported that metallographic examination of the UC indicated a "slight second phase" which was identified as "either UC<sub>2</sub> or U<sub>2</sub>C<sub>3</sub>." Examination of the UC<sub>2</sub> specimens showed single phase UC<sub>2</sub> containing a trace phase of UC.

Resistivity was measured by a potentiometric technique. The author estimated an error of  $\pm 2$  and  $\pm 5\%$ , respectively, for the UC and UC<sub>2</sub> data. No error estimate was suggested for the U<sub>2</sub>C<sub>3</sub> data.

The data for U<sub>2</sub>C<sub>3</sub> were obtained by "soaking" the UC<sub>2</sub> specimen at about 1275°C for 30 min, followed by 6 min at 1175°C. The author indicated there may have been residual UC<sub>2</sub> present as second phase during the measurement.

Grossman reported his results in the form of linear fits to the UC and UC<sub>2</sub> data, as given by the following expressions.

UC: for  $827 < T^\circ\text{C} < 1777$  ,

$$\rho = 51.74 + 0.1148 T \mu\Omega\text{cm} , \quad (12)$$

UC<sub>2</sub>: for  $1277 < T^\circ\text{C} < 1797$  ,

$$\rho = 204.8 + 0.0580T \mu\Omega\text{cm} . \quad (13)$$

A data summary is given in Fig. 6, with extrapolation of the UC data to room temperature. The "U<sub>2</sub>C<sub>3</sub>" data are about  $40 \mu\Omega\text{cm}$  higher than the UC data from 1400 to 1800°C.

11. Norreys,<sup>29</sup> in a study of the transformation of U + UC<sub>2</sub> to U<sub>2</sub>C<sub>3</sub>, measured the electrical resistivity of uranium carbides containing 54.2 at.% (5.6 wt%)

**TABLE I**  
**SOME MEASURED EFFECTS ON RESISTIVITY OF UC**

wt% Carbon (nominal)	Pre Heat-Treat		Total Uranium Burnup (at.%)	Av Temp Irradiation (°C)	Density (g/cm <sup>3</sup> )		Resistivity (μΩcm)	
	Temp (°C)	Time (h)			Pre	Post	Pre	Post
5.0	1800	1	0.004	~150	13.45	13.45	32	88
5.0	1800	1	0.016	~150	13.36	13.32	37	101
6.7	1400	15	0.33	~150	12.62	12.72	50	211
6.7	1400	15	0.35	~150	12.63	12.76	48	265
5.0	1450	5	0.47	430-720	13.52	13.35	38	59

and 59.4 at.% (6.9 wt%) using a four-probe potentiometric technique. Measurements were made in vacuum at  $10^{-5}$  torr. The specimens were prepared by arc melting and drop casting. Resulting impurity levels were less than 500 ppm total metallic impurity other than tungsten, up to 1 wt% tungsten (for some samples), less than 200 ppm oxygen, and less than 100 ppm nitrogen.

Norreys heated a specimen containing 5.6 wt% carbon at  $13.2 \text{ g/cm}^3$ , with a resistivity of  $39.5 \mu\Omega\text{cm}$  at 20 to  $1400^\circ\text{C}$ , then measured the resistivity at 10-min intervals. He found the resistivity at  $1400^\circ\text{C}$  remained constant at about  $202 \mu\Omega\text{cm}$  for about 40 min, then increased to about  $218 \mu\Omega\text{cm}$  at 80 min and remained constant. On cooling to  $20^\circ\text{C}$  the resistivity had increased to  $66.8 \mu\Omega\text{cm}$ , indicating transformation to  $\text{U}_2\text{C}_3$ . The specimen was then heated to  $1900^\circ\text{C}$  and cooled rapidly to  $20^\circ\text{C}$  where the resistivity was measured as  $37.5 \mu\Omega\text{cm}$ , in-

dicating transformation to  $\text{UC} + \text{UC}_2$ . It was subsequently found that slight mechanical stressing of the sample was required to initiate the  $\text{U}_2\text{C}_3$  transformation at  $1400^\circ\text{C}$ .

The resistivities in Fig. 6 are from Norreys' plots showing the temperature dependence of the resistivity for two samples of  $\text{U}_2\text{C}_3 + \text{UC}$  on heating and subsequent cooling. Note the range of temperature independence during the  $\text{U}_2\text{C}_3$  to  $\text{UC}_2$  transformation.

**12. Summary: Electrical Resistivity of Uranium Carbide.** Examination of all the data summarized in Fig. 1 for nearly stoichiometric UC, 4.8 wt% carbon, suggests that, except for the data of Costa, resistivities are in agreement to within about 8%. Correction of the Hayes data, using Eq. (4), gives about 5% maximum variation among all of the results. The combined oxygen and nitrogen impurity levels of the samples studied varied from about 200 to 2000 ppm. Bates' data<sup>5</sup> show a significant increase in resistivity for oxygen concentrations greater than about 2500 ppm.

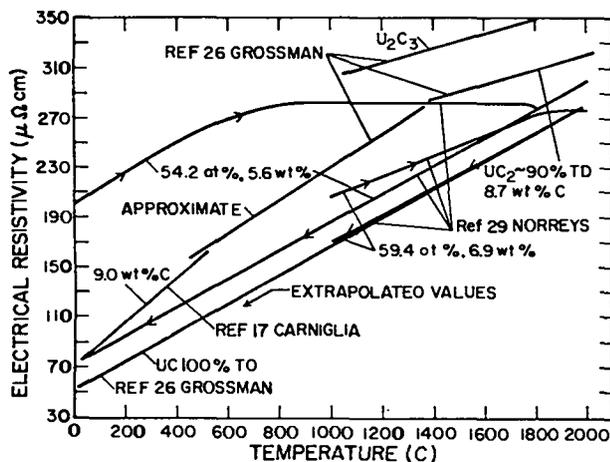
Hayes data<sup>9</sup> suggest that Eq. (4) can be used as a correction for porosity of samples over 90% TD, giving corrected resistivities within 5% of other reported values at 100% TD. The approximation is not as satisfactory for material less than 90% dense.

Although Rough<sup>10</sup> did not discuss impurity levels in his samples, his results agree well with data of others for samples containing less than 2000 ppm total impurity. Grossman's data<sup>26</sup> represents the only information available above  $1600^\circ\text{C}$ ; however, his samples contained second phase  $\text{UC}_2$  or  $\text{U}_2\text{C}_3$ .

It is suggested that Rough's data, corrected by Eq. (4), be used as the resistivity of 100% dense UC. The resistivity is expressed as

$$\rho = 32.9 + 0.159T - 2.688 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (14)$$

for  $25 \leq T^\circ\text{C} \leq 1600$ .



**Fig. 6.**  
*Resistivity of UC, UC<sub>2</sub>, and U<sub>2</sub>C<sub>3</sub>, according to Grossman.<sup>26</sup>*

Examination of the data shown in Figs. 3-6 brings the conclusion that the resistivity of UC increases with carbon content. Quantitative evaluation of this effect is not possible from available data. All authors reported two phase UC + U structures in hypostoichiometric samples and two phase UC + UC<sub>2</sub> or UC + U<sub>2</sub>C<sub>3</sub> structures in hyperstoichiometric materials. It is suspected that Rough and Carniglia's data (Fig. 5) for 7 wt% carbon represent the resistivity of a UC + UC<sub>2</sub> mixture.

The UC<sub>2</sub> data of Grossman seem to give the most reasonable estimate of the resistivity of UC<sub>2</sub>. The apparent agreement, shown in Fig. 6, of the Grossman data for UC<sub>2</sub> (8.7 wt% carbon) below 1300°C and the Carniglia data for 9.0 wt% carbon, a mixture of UC + UC<sub>2</sub>, seems to be substantiated by Norreys' data. However, this illustrates the necessity for relating the transport properties of nonstoichiometric carbides to both the phase concentration and carbon content as well as thermal history.

The Grossman data seem to indicate the electrical resistivity of UC<sub>2</sub> is approximately 25% higher than that of single phase UC.

Rough's data, summarized in Table I, show an increase in resistivity of about three times the normal value after irradiation of approximately stoichiometric UC from 0.004 to 0.016 at.% burnup.

## B. Plutonium Carbide

There have been relatively few determinations of the electrical resistivity of PuC. However, results of all measurements on PuC<sub>1-x</sub> from room temperature to 1000°C show agreement within about ±7% of the value, 250 μΩcm.

1. Fulkerson<sup>3</sup> reviewed the PuC resistivity data of Costa,<sup>7</sup> Leary,<sup>8</sup> and Moser,<sup>4</sup> concluding the resistivity of PuC was essentially constant at 260 μΩcm from ambient to 1000°C.

2. Moser's review<sup>4</sup> included a discussion of the data of Kruger,<sup>27</sup> Leary,<sup>8</sup> and Costa,<sup>7</sup> which show less than ±5% variation from the value 260 μΩcm from 25 to 500°C.

3. Kruger<sup>28</sup> reported a resistivity of about 260 μΩcm for cast PuC containing 47 at.% carbon over the temperature range 25 to 700°C. The four-probe potentiometric method was used in the measurements. No discussion of specimen density or impurity level was included.

4. Kruger<sup>27</sup> reported the results of resistivity measurements on single phase, arc-cast PuC<sub>0.85</sub>

(45.95 at.% carbon of about 100% TD. The specimens contained 0.01 wt% oxygen and less than 0.005 wt% nitrogen. Measurements were made by a four-probe potentiometric method which used several determinations at various dc current levels for each temperature and which gave a measurement precision of 0.5%. The data showed a decrease in resistivity from 257 μΩcm at about 27°C to 254 μΩcm at 727°C. The data are plotted in Fig. 7. The UC data of Rough<sup>10</sup> are shown for comparison. Kruger fitted the following polynomial to the data.

$$\rho = 263.30 - 1.966 \times 10^{-2}T - 1.171 \times 10^{-5}T^2 + 2.246 \times 10^{-8}T^3 \text{ } \mu\Omega\text{cm} \quad (15)$$

for 300 ≤ T°K ≤ 1000. Equation (16) also fits the tabulated experimental data within ±0.2%.

$$\rho = 258.0 - 0.0287T + 3.232 \times 10^{-5}T^2 \text{ } \mu\Omega\text{cm} \quad (16)$$

for 25 ≤ T°C ≤ 750.

5. Costa<sup>7</sup> measured the resistivity of single phase PuC<sub>0.905</sub> (47.5 at.% carbon) over the temperature range 10 to 1200°K by a potentiometric method. Discussion of impurity level, density, or method of sample fabrication was not included in the article. The data plot showed the resistivity decreasing to about 240 μΩcm at 150°K with an increase to 260 μΩcm at 1200°K. The data over the range 27 to 927°C are shown in Fig. 7 and are well represented by

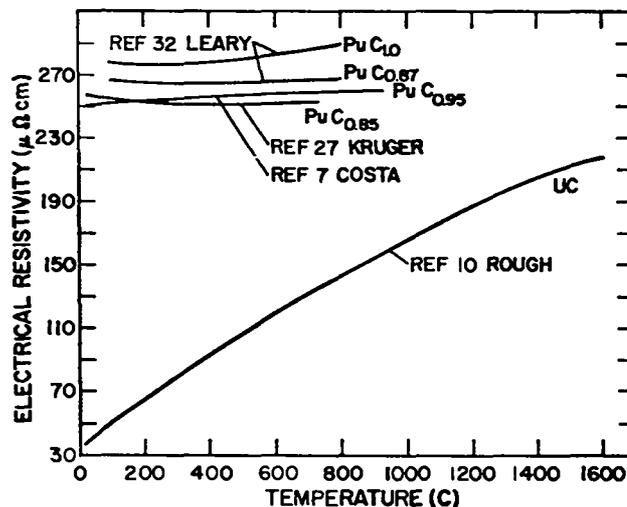


Fig. 7. Resistivity of PuC, according to Kruger,<sup>27</sup> Leary,<sup>32</sup> and Costa.<sup>7</sup>

$$\rho = 249.4 + 1.427 \times 10^{-2}T - 2.624 \times 10^{-6}T^2 \mu\Omega\text{cm} \quad (17)$$

for  $27 \leq T^\circ\text{C} \leq 927$ .

6. **Leary et al.**<sup>8,30-32</sup> reported progressive phases of resistivity measurements on PuC in several articles; the work is summarized in Ref. 8. Experimental results over the range 100 to 800°C were reported for single phase PuC<sub>0.86</sub> and PuC<sub>1.0</sub> specimens which were prepared by multiple arc melting of high purity (less than 200 ppm impurity) plutonium and ground spec grade graphite. The arc-melted buttons were subsequently arc cast into rod, then solution heat treated at 1300°C for 6 h prior to testing. Neither density nor residual impurities were reported. It was stated that the PuC<sub>1.0</sub> was a mixture of PuC<sub>1-x</sub> + Pu<sub>2</sub>C<sub>3</sub>. Measurements were made by a comparative eddy current method. The estimated error was  $\pm 5\%$  of sample resistivity.

The data, plotted in Fig. 7, are closely approximated by

PuC<sub>1.0</sub>:

$$\rho = 278.5 - 1.661 \times 10^{-2}T + 3.869 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (18)$$

PuC<sub>0.87</sub>:

$$\rho = 267.20 - 1.327 \times 10^{-2}T + 1.726 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (19)$$

for  $100 \leq T^\circ\text{C} \leq 800$ .

7. **Summary: Electrical Resistivity of Plutonium Carbide.** Kruger,<sup>33</sup> in his discussion of the phase diagram and properties of PuC, cited work of Mulford<sup>34</sup> and Kruger<sup>35</sup> which shows the presence of Pu<sub>2</sub>C<sub>3</sub> in as-cast compositions containing more than 46 at.% carbon. Heat treatment at 1000°C did not cause solution to the Pu<sub>2</sub>C<sub>3</sub> phase. Kruger, in the course of specimen preparation for compatibility studies, noted about 25 vol% Pu<sub>2</sub>C<sub>3</sub> in pressed and sintered PuC<sub>1.0</sub>.

Costa<sup>36</sup> reported 510  $\mu\Omega\text{cm}$  at 300°K as the resistivity of 78% TD Pu<sub>2</sub>C<sub>3</sub>. The material was prepared by vacuum pressing and sintering, at 1600°C, powder prepared by reacting plutonium hydride with carbon at 1400°C.

Considering the above, and the relation between the data of Leary et al. for PuC<sub>1.0</sub> and the data of Kruger for PuC<sub>0.87</sub>, simplistic mixing of the single

phase PuC + Pu<sub>2</sub>C<sub>3</sub> would give a resistivity of about 280  $\mu\Omega\text{cm}$  at 27°C. This is close to the value reported by Leary for PuC<sub>1.0</sub>.

We suggested that Eq. (16), which represents the data of Kruger,<sup>28</sup> be used for the resistivity of single phase PuC, and that the data of Leary<sup>8</sup> for PuC<sub>1.0</sub> (Eq. (18)) be used as the current best approximation for the resistivity of PuC<sub>1-x</sub> + Pu<sub>2</sub>C<sub>3</sub>.

On the basis of existing information, no quantitative estimate of the effects of oxygen and nitrogen impurity or carbon content can be given.

Lack of information on porosity effects precludes quantitative porosity correction. Equation (4) can be used as an approximation.

### C. Uranium-Plutonium Carbide

Since 1961 there have been fewer than 12 reports of studies on the resistivity of (U,Pu)C. The 1970 review by Fulkerson<sup>3</sup> discussed results of VanCraeynest,<sup>37</sup> Milet,<sup>38</sup> and Leary.<sup>8</sup> The discussion by Leary<sup>8</sup> summarized the data reported in Refs. 29, 31, and 32.

1. **Pascard**,<sup>24</sup> using a potentiometric method, examined the room temperature resistivities of pressed and sintered mixtures of UC and PuC containing 5, 10, 30, and 50 mol% PuC at 90 to 97% TD. Additions of 0.5 wt% nickel to these mixtures were found to decrease the resistivity about 10 to 15%. Pascard's data are approximated by Eq. (20) to within 10% of the reported values.

$$\rho = 54.7 + 4.12 (\text{cPuC}) - 2.38 \times 10^{-2} (\text{cPuC})^2 \mu\Omega\text{cm} \quad (20)$$

where cPuC = concentration of PuC in mol% for  $5 \leq \text{cPuC} \leq 100$ . The reported values for 100% UC and 100% PuC are, respectively, 30 and 230  $\mu\Omega\text{cm}$ .

2. **VanCraeynest**<sup>37</sup> measured the resistivity of (U<sub>0.85</sub>Pu<sub>0.15</sub>)C at 91.5% TD from 100 to 1300°C. No discussion of measurement method or impurity level was given. The data tabulated in this article are plotted in Fig. 8 and can be approximated by

$$\rho = 92.7 + 0.173T - 4.018 \times 10^{-5}T^2 \mu\Omega\text{cm} \quad (21)$$

for  $100 \leq T^\circ\text{C} \leq 1300$ .

3. **Leary**<sup>8</sup> reported resistivities measured on (U<sub>0.8</sub>Pu<sub>0.2</sub>)C specimens over the range 100 to 800°C. Samples were prepared by arc casting and solution heating at 1300°C for 6 h prior to testing. Total impurity in the component materials was reported to

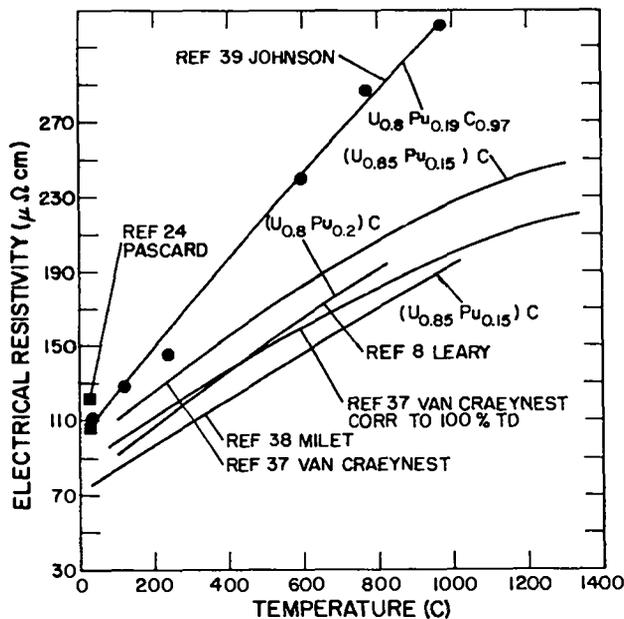


Fig. 8.  
Summary of resistivity values reported for (U,Pu)C.

be less than 200 ppm. Residual oxygen and nitrogen impurity was not discussed. Measurement was by a comparative eddy current technique resulting in an error of  $\pm 5\%$  of sample resistivity. The data plotted in the article were linear with temperature and can be represented by

$$\rho = 78.6 + 0.142T \mu\Omega\text{cm} \quad (22)$$

for  $100 \leq T^\circ\text{C} \leq 800$ .

4. Johnson<sup>39</sup> reported resistivity measurements on a sample of nominal composition  $\text{U}_{98}\text{Pu}_{0.19}\text{C}_{0.97}$  which had been prepared by cold pressing and sintering powder made by grinding the arc-melted alloy (Ref. 39, pp. 24-26). The specimen was apparently about 95% TD. Measurements were by a four-probe dc potentiometric method. Actual sample composition was reported as  $\text{U}_{0.8}\text{Pu}_{0.19}\text{C}_{0.97}\text{O}_{0.028}\text{N}_{0.013}$  containing  $\text{M}_2\text{C}_3$  tangles and platelets. Surface oxidation of the sample during measurement was reported. The relatively high values of resistivity from this work, shown in Fig. 8, probably are explained by the relatively high impurity level and oxidation which occurred during test.

5. Milet's<sup>38</sup> data, reported for about 100% TD  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  prepared by arc casting, was obtained by a dc potentiometric method. Total impurity in

the sample was less than 100 ppm. These data, shown in Fig. 8, are well represented by

$$\rho = 72.0 + 0.122T \mu\Omega\text{cm} \quad (23)$$

for  $25 \leq T^\circ\text{C} \leq 1000$ .

6. Summary: Electrical Resistivity of Uranium-Plutonium Carbide. On the basis of the above information, the following suggestions are made.

(a) There are not enough data available to recommend a quantitative correction to the electrical resistivity of (U,Pu)C for effects of fabrication method, impurity level, porosity, or stoichiometry.

(b) Although the data shown in Fig. 8 agree to within about 10%, including the corrected VanCraeynest data, the differences in resistivity indicated by the  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  data of Leary and the  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  data of Milet seem to reflect the proportionate increase resulting from PuC content determined by Pascard. Equations (22) and (23) are suggested as approximations to the respective resistivities over the range 25 to  $1000^\circ\text{C}$  for high purity material of about 100% TD.

#### D. Recommendations and Conclusions: Electrical Resistivity

Suggested values for the electrical resistivity of UC, PuC, and (U,Pu)C are summarized in Table II. Tabulated values are calculated by equations of the form  $\rho = a + bT + cT^2$ .

The following conclusions may be drawn.

(a) Although the resistivity of 100% dense single phase UC is apparently well defined to  $1500^\circ\text{C}$ , there is a need for measurements at higher temperatures. The same is true for PuC and (U,Pu)C above  $1000^\circ\text{C}$ .

(b) It can be assumed that combined oxygen and nitrogen impurity less than about 2000 ppm does not significantly increase the resistivity of UC. However, even considering the well-designed experiments by Bates, quantitative assessment of the effect cannot be made. Essentially no data exist which indicate the quantitative effect of oxygen and nitrogen levels on PuC or (U,Pu)C.

(c) The effect of carbon concentration on resistivity can be known qualitatively at best until experiments are performed to correlate thermal treatment cycles with second and third phase concentrations and resistivity.

(d) Corrections for porosity, which are accurate at other than ambient temperature, are probably dependent both upon temperature and pore morphology. These effects have not been studied.

**TABLE II**  
**SUGGESTED VALUES FOR ELECTRICAL RESISTIVITY OF UC, PuC, AND (U,Pu)C**

Equation Coefficients <sup>b</sup>	Electrical Resistivity ( $\mu\Omega\text{cm}$ ) <sup>a</sup>					
	UC <sub>1.0</sub> 100% TD	UC <sub>2</sub> <sup>c</sup>	PuC <sub>0.85</sub> Single Phase	PuC <sub>1.0</sub> PuC <sub>1-x</sub> + Pu <sub>2</sub> C <sub>3</sub>	(U <sub>0.85</sub> Pu <sub>0.15</sub> )C 100% TD	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C 100% TD
a	32.9	68.14	258.0	278.5	72.0	78.6
b	0.159	-0.205	-0.0287	$-1.66 \times 10^{-2}$	0.122	0.142
c	$-2.680 \times 10^{-5}$	$-3.93 \times 10^{-5}$	$3.232 \times 10^{-5}$	$3.869 \times 10^{-5}$	---	---
<b>Temperature (°C)</b>						
25	37.0	73.0	257.0	---	75.0	---
100	49.0	88.0	255.0	277.0	84.0	93.0
300	78.0	126.0	252.0	277.0	109.0	121.0
500	106.0	161.0	252.0	280.0	133.0	150.0
700	131.0	192.0	254.0	286.0	157.0	178.0
900	154.0	221.0	255.0	290.0	182.0	192.0
1100	175.0	246.0	---	---	(1000°) <sup>d</sup> 194.0	---
1300	194.0	268.0	---	---	---	---
1500	211.0	287.0	---	---	---	---
1700	---	303.0	---	---	---	---
1900	---	316.0	---	---	---	---
2000	---	321.0	---	---	---	---

<sup>a</sup> Values obtained by extrapolation to temperatures higher than indicated in Table II should be used with caution.

<sup>b</sup> For equation  $\rho = a + bT + cT^2$ .

<sup>c</sup> The data for UC<sub>2</sub> are very speculative. These values were calculated for carbon content of about 9 wt% from the equation obtained by combining the data of Carniglia and Grossman shown in Fig. 6. Norreys' data indicate the effect of changing carbon concentration and thermal cycling.

<sup>d</sup> Indicates temperature in degrees Celsius at which the last data point was observed.

(e) Small additions of nickel apparently reduce electrical resistivity. Quantitative effects are not known.

(f) Rough's data<sup>10</sup> essentially provide the only information on the effects of irradiation on carbide resistivity.

(g) The effect of differences in fabrication methods on resistivity cannot be quantitatively evaluated on the basis of existing data.

### III. THERMAL CONDUCTIVITY

Many of the articles considered in this section were cited in the section on electrical resistivity; of those articles concerned only with thermal conductivity, several materials have been considered. To simplify the discussion, information for UC, PuC, and (U,Pu)C is presented in separate subsections.

Reviews of the work on thermal conductivity written since 1967 are by Sheth,<sup>1</sup> Washington,<sup>41</sup> Fulkerson,<sup>3</sup> Bates,<sup>5</sup> and Leary.<sup>6</sup>

#### A. Uranium Carbide

1. **Sheth and Leibowitz**<sup>1</sup> considered the reviews of Fulkerson<sup>3</sup> and Washington<sup>41</sup> in suggesting the preferred values of Washington be used for the thermal conductivity of UC.

2. **Washington**<sup>41</sup> reviewed the work of the authors cited in Refs. 3, 5, 6, 8, 9, 14, 15, 17, 21, and 43-48. He concluded that for 100% dense stoichiometric UC in the temperature range from about 500 to 2000°C, the thermal conductivity increases linearly from about 20 to 22 W/m·°C. There is considerable variability in results below 700°C, some data indicating a significant phonon contribution. Washington's recommended values for 100% dense stoichiometric UC are given by the following equation.

$$\lambda = 20.0 + 1.30 \times 10^{-3} (T-500) \text{ W/m} \cdot \text{deg} \quad (24)$$

for  $500 \leq T^{\circ}\text{C} \leq 2000$ .

Washington concluded that the effects of stoichiometry were very uncertain, and offered what he considered "very speculative values" for hypostoichiometric material based on extrapolation of Carniglia's<sup>17</sup> results for UC at 4.0 wt% C. He expressed the conductivity as

$$\lambda = 17.0 + 2.9 \times 10^{-3} (T-500) \text{ W/m} \cdot \text{deg} \quad (25)$$

for  $500 \leq T^{\circ}\text{C} \leq 2000$ . Because neither a general data pattern nor quantitative effects had been established for hyperstoichiometric material, Washington concluded it was impossible to suggest preferred values.

His assessment of the effect of oxygen impurity was based on the published work of Wheeler,<sup>42,44</sup> Bates,<sup>5,41</sup> Hayes and DeCrescente,<sup>9</sup> and unpublished data (1971) of King and Waite (Harwell). All investigators agreed that thermal conductivity is decreased by oxygen impurity, the greatest effect being observed below 1000°C. The conductivity values for the oxycarbides approach the UC values at 2400°C. Washington concluded that the existing data do not permit separation of effects of stoichiometry, impurities including nickel and fraction of second phase present, therefore, accurate conductivity values could not be given. His tentative suggestions were expressed in equation form as

$$\lambda = 16.0 + 3.4 \times 10^{-3} (T-500) \text{ W/m} \cdot ^{\circ}\text{K}$$

$$\text{for } 5 \text{ at.}\% \text{ O} \quad (26)$$

and

$$\lambda = 14.0 + 4.5 \times 10^{-3} (T-500) \text{ W/m} \cdot ^{\circ}\text{K}$$

$$\text{for } 7.5 \text{ at.}\% \text{ O (1 wt\%),} \quad (27)$$

for  $500 \leq T^{\circ}\text{C} \leq 2400$ .

Washington examined the data of Moser and Kruger,<sup>45</sup> Hayes and DeCrescente,<sup>9</sup> and Crane and Gordon<sup>47</sup> for the effect of porosity. His conclusion again was that disagreement among results indicated that a quantitative separation of effects was not possible and that a true porosity correction would be temperature and pore-shape dependent. He suggested that at the present state of knowledge the porosity correction given by Eq. (28) be used for all compositions.

$$\lambda_M = \lambda_{TD} (1-P)/(1+P) \quad (28)$$

where P = porosity fraction and  $\lambda_{TD}$  = conductivity for P = 0, and  $\lambda_M$  = measured conductivity. The preferred values of Washington are plotted in Fig. 9.

3. **Fulkerson**<sup>3</sup> reviewed the work by Bates,<sup>5</sup> Sobon,<sup>15</sup> Hayes and DeCrescente,<sup>9</sup> DeCrescente and Miller,<sup>49</sup> Moser and Kruger,<sup>45</sup> Chubb and Dickerson,<sup>50</sup> and Pascard,<sup>43</sup> arriving at preferred conductivity values, plotted in Fig. 9, which he fitted by the expression

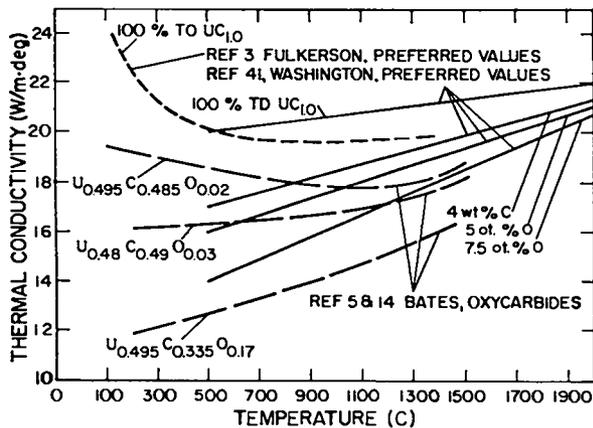


Fig. 9.

Comparison of some selected values for the thermal conductivity of UC with values for UCO reported by Bates.<sup>5,14</sup>

$$\lambda = LT/\rho + \eta_1/T \quad (29)$$

where  $L = 0.933 L_0$ ,  $L_0$  = the Sommerfield Lorenz number,  $\eta_1^{-1} = 3.58 \times 10^{-4} \text{ m} \cdot \text{W}^{-1}$ , and  $\rho$  = preferred resistivities [see Eq. (1)].

Most of the thermal conductivity data considered were calculated from thermal diffusivities using the combined specific heat data of Storms,<sup>51</sup> Levinson,<sup>52</sup> DeCrescente,<sup>49</sup> and Moser.<sup>53</sup> These values are within 2% of the recommended values of Olson.<sup>54</sup> Fulkerson did not discuss the effect of impurities or stoichiometry in any detail.

4. Bates' study<sup>5,14</sup> of uranium oxycarbides included measurements of the thermal diffusivity of four oxycarbide compositions from which he calculated the thermal conductivity using heat capacity data for UC reported by Godfrey et al.<sup>55</sup> Bates' results are summarized here to indicate the large effect of a relatively small amount of oxygen impurity on the thermal conductivity of UC. The experiments considered the four compositions listed below which were prepared by carbothermic reduction, pulverization, pressing, and sintering.

(a)  $\text{U}_{0.495}\text{C}_{0.485}\text{O}_{0.02}$ : density of  $12.7 \text{ g/cm}^3$  and a microstructure,  $\text{UC}_x\text{O}_y$  + traces of free U and  $\text{UC}_2$ .

(b)  $\text{U}_{0.480}\text{C}_{0.490}\text{O}_{0.03}$ : density  $13.1 \text{ g/cm}^3$  and a microstructure, two-phase  $\text{UC}_x\text{O}_y$  +  $\text{UC}_2$  + trace of  $\text{UO}_2$ .

(c)  $\text{U}_{0.495}\text{C}_{0.335}\text{O}_{0.170}$ : density of  $12.3 \text{ g/cm}^3$  and a microstructure  $\text{UC}_x\text{O}_y$  +  $\text{UO}_2$  + traces of free U.

(d)  $\text{U}_{0.495} + \text{C}_{0.355} + \text{O}_{0.150}$ : density of  $12.5 \text{ g/cm}^3$  and a microstructure  $\text{UC}_x\text{O}_y$  +  $\text{UO}_2$  + traces of free U.

The samples were given a "homogenization" treatment to  $1000^\circ\text{C}$  before diffusivity measurements were made from 100 to  $1500^\circ\text{C}$ .

Measurements were performed by the flash method using a pulsed laser as the energy source. The apparatus was calibrated using Armco iron as the reference sample. The uranium oxycarbide samples were subjected to microstructural examination both before and after test.

Bates' results are summarized by Eqs. (30)-(32) which were fitted to data points read from his graph in Ref. 5. These data are shown in Fig. 9.

$$\lambda = 20.0 - 4.38 \times 10^{-3}T + 2.26 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg} \quad (30)$$

for  $\text{U}_{0.495}\text{C}_{0.485}\text{O}_{0.02}$ ,  $100 \leq T^\circ\text{C} \leq 1500$ ,

$$\lambda = 16.4 - 8.02 \times 10^{-4}T + 1.30 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg} \quad (31)$$

for  $\text{U}_{0.48}\text{C}_{0.49}\text{O}_{0.03}$ ,  $200 \leq T^\circ\text{C} \leq 1500$ ,

$$\lambda = 11.4 + 2.51 \times 10^{-3}T + 5.67 \times 10^{-7}T^2 \text{ W/m} \cdot \text{deg} \quad (32)$$

for  $\text{U}_{0.495}\text{C}_{0.335}\text{O}_{0.17}$ ,  $200 \leq T^\circ\text{C} \leq 1450$ . Data for the sample containing 15 at.% oxygen show about  $0.8 \text{ W/m} \cdot \text{deg}$  higher conductivity at  $200^\circ\text{C}$  than for the sample containing 17 at.% oxygen. The data indicate the conductivities for these two compositions were about the same from  $800$  to  $1450^\circ\text{C}$ .

Bates concluded that mixed conduction occurs in the uranium oxycarbides, and that the presence of oxygen in the lattice affects the lattice conductivity more than the electronic conductivity. His results indicated that, although electronic conduction is predominant above about  $1000^\circ\text{C}$ , lattice conduction was significant for those samples containing small amounts of oxygen.

Bates<sup>5</sup> also reviewed the results of some 19 other investigators. These data, much of which will be discussed, showed wide variability between about  $17 \text{ W/m} \cdot \text{deg}$  and  $32 \text{ W/m} \cdot \text{deg}$  from  $50$  to  $500^\circ\text{C}$ . The author concluded this was probably a result of variation in oxygen content of the materials tested.

5. Leary's<sup>6</sup> review considered the data of Crane,<sup>25</sup> Dayton,<sup>19</sup> Russell,<sup>56</sup> Leary,<sup>8</sup> Wheeler,<sup>44</sup> Wittenberg,<sup>59</sup> and Mustacchi<sup>16</sup> on the thermal conductivity of UC. Because results reported by

these investigators (except for those of Mustacchi) will be considered individually, only Leary's conclusions will be included here as follows.

(a) The thermal conductivity data over the temperature range 200 to 2000°C fall within about  $\pm 20\%$  of the value 21 W/m·deg. (For example, the Mustacchi data show essentially a constant value,  $\lambda = 17.4$  W/m·deg from 1000 to 2000°C, while the data of Wheeler shows an increase from about 16.7 to 26.0 W/m·deg over the same range.)

(b) Maximum thermal conductivity is observed in the near stoichiometric range for UC.

(c) Nickel sintering aid, even in the 0.1% by weight concentration range, lowers the conductivity of UC.

(d) Excessive amounts of oxygen and nitrogen lower the thermal conductivity.

(e) There does not appear to be a satisfactory quantitative method for adjusting for effects of porosity on thermal conductivity of carbides. These conclusions also applied in general to PuC and (U,Pu)C and will be considered in the conductivity summary.

6. Wheeler<sup>42</sup> performed diffusivity measurements on several types of material using a modulated electron beam technique. Calibration methods were not discussed. The thermal conductivities were calculated from the determined diffusivity values using the specific heat for UC from Krikorian.<sup>57</sup> The following UC materials were considered by Wheeler.

(a) Sintered, unalloyed UC containing about 0.3 wt% oxygen, or approximately 2.4 at.% oxygen, prepared by sintering uranium powder and graphite.

(b) Cold-pressed and sintered carbothermic reduction product to which 0.1 and 0.3 wt% nickel was added. This material contained 0.1 to 0.2 wt% oxygen.

(c) Cold pressed and sintered U + UO<sub>2</sub> + graphite powders. The UO<sub>2</sub> addition was controlled to yield material containing 0.5, 1.0, and 2.0 wt% oxygen.

(d) Arc-melted uranium + graphite controlled to produce materials containing 49.9, 50.8, and 51.8 at.% carbon. These materials contained about 200-ppm oxygen and 100-ppm nitrogen.

Densities of the sintered materials were reported to range from 94 to 97% of theoretical. Chemical and microstructural examinations were performed both before and after diffusivity measurements were made. Though detectable amounts of second phase were observed in the samples, notably free uranium in the hypostoichiometric samples, no judgment as to effect was made.

The thermal conductivities calculated by Wheeler for these materials are summarized in Fig. 10.

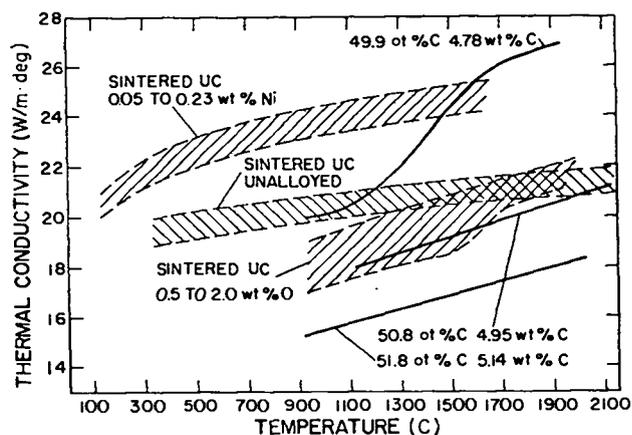


Fig. 10.  
Thermal conductivity of UC, according to Wheeler.<sup>42</sup>

Wheeler's conclusions concerning the UC data were as follows.

(a) The presence of excess carbon or oxygen decreases the thermal conductivity of UC. The data indicate about a 20% decrease in conductivity at oxygen concentrations of 2 wt%. Wheeler attributed differences in his conductivity values and those of Bates to differences in carbon concentration of their respective materials.

(b) The thermal conductivity of UC is probably increased by small amounts of nickel. His data indicate about a 20% increase in conductivity resulting from up to 0.23 wt% nickel.

(c) The inflection in the conductivity curve above about 1600°C for the hypostoichiometric UC may be attributed to a change in vacancy distribution.

(d) Phonon transport forms a significant portion of the thermal conductivity of UC, and (U,Pu)C, at high temperatures.

7. Pascard<sup>43</sup> measured the diffusivity, from 100 to 2000°C, of UC prepared by pressing and sintering carbide powder which was prepared by comminution of carbide formed from the metal hydride. Two types of the phase shift measurement methods were used: modulated electron beam above 1000°C and sinusoidal heat wave from ambient to 1000°C. The apparatus was calibrated against known standards. The accuracy of diffusivity measurement was estimated as  $\pm 10\%$ .

The reported conductivities were calculated from the measured diffusivity using  $C_p$  values selected by Pascard. These values are about 10% lower than Olson's recommended values<sup>54</sup> at 1500°C, but within 2% below 1000°C. The samples were reported to contain 800 ppm oxygen and 130 ppm nitrogen and to be 95% TD. Pascard's conductivity curve,

shown in Fig. 11, can be represented by the following expressions.

$$\lambda = 25.2 - 2.32 \times 10^{-2}T + 2.21 \times 10^{-5}T^2 \text{ W/m} \cdot \text{deg} \quad (33)$$

for  $100 < T^{\circ}\text{C} < 700$ ,

$$\lambda = 17.2 + 3.52 \times 10^{-3}T \text{ W/m} \cdot \text{deg}$$

for  $700 < T^{\circ}\text{C} \leq 2000$ .

8. Hayes<sup>9</sup> measured the conductivity of three stoichiometric UC samples using a steady state radial heat flow method. Two of these samples were 95% TD containing 2600 ppm oxygen and 1400 ppm nitrogen, and 860 ppm oxygen and 520 ppm nitrogen, respectively. The third sample of 91% TD contained 1260 ppm oxygen and 930 ppm nitrogen. The samples were prepared by hot pressing methane synthesized UC powder. Density was adjusted by "particle size selection."

The data tabulated in Hayes' article are plotted in Fig. 11 with curves as he showed them. He concluded that

- Porosity effects can be corrected by Eq. (4).
- Thermal conductivity is relatively independent of temperature in the range 1000 to 2000°C.
- Increasing the oxygen + nitrogen impurity level from about 1400 to about 2200 ppm had a minor effect on conductivity.
- Thermal conductivity above 1000°C is 100% electronic. (Hayes' estimate from Wiedemann-Franz relation using his resistivity values is discussed in Sec. II.A.6.)

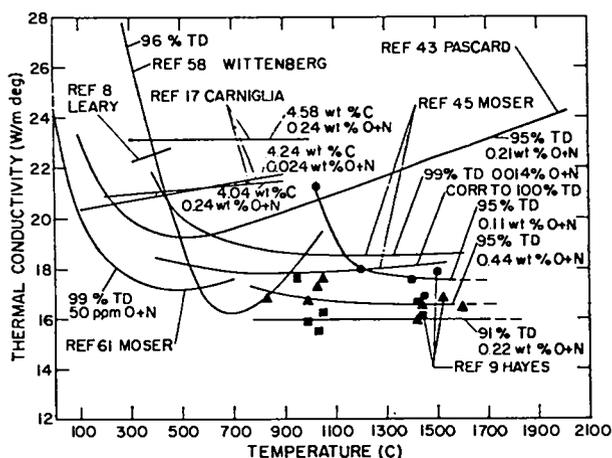


Fig. 11.

Variability in reported values for the thermal conductivity of UC.

9. Carniglia<sup>17</sup> measured the thermal diffusivity of several arc-cast UC samples containing 4.04 to 4.58 wt% carbon by the flash diffusivity technique using a laser as the energy source. The apparatus was calibrated against known standards. Accuracy of the diffusivity measurements was reported to be  $\pm 4\%$ . Conductivities were calculated from the measured diffusivities, measured densities (99.3% TD), and  $C_p$  values of Krikorian.<sup>57</sup>

The materials tested were apparently about 99% TD containing about 2400 ppm oxygen + nitrogen. Carniglia reported values of about 23.0 to 23.4 W/m·deg for stoichiometric UC over the range 300 to 1000°C and about 21 to 21.3 W/m·deg for hypostoichiometric UC containing 4.04 wt% carbon.

10. Moser<sup>45</sup> measured thermal diffusivities of UC, PuC, and (U,Pu)C prepared by arc-casting, pressing, and sintering at 1800°C powder prepared from the arc-cast material. The flash diffusivity equipment was calibrated against an  $\text{Al}_2\text{O}_3$  standard. The maximum estimated error in the measurements was reported as  $\pm 7.5\%$ , with an accuracy of  $\pm 5\%$ . Chemical analyses of the sample materials showed 4.93 wt% carbon, 0.0117 wt% oxygen, and 0.0021 wt% nitrogen for the arc-cast sample and 4.69 wt% carbon, 0.106 wt% oxygen, and 0.0228 wt% nitrogen for the pressed and sintered sample. Moser reported an equivalent carbon concentration of 4.94 wt% or 50.8 at.% and 99.0% TD for the cast sample, and 4.79 wt% or 49.9 at.% equivalent carbon and 78.8% TD for the pressed and sintered sample.

The arc-cast material contained a small amount of intragranular  $\text{UC}_2$ , while the sintered sample was single phase. This was in apparent agreement with Moser's "equivalent carbon" concept for UC, PuC, and (U,Pu)C. Equivalent carbon content is expressed by summing the oxygen, nitrogen, and carbon as

Equivalent wt% C

$$= \text{wt}\% \text{ C} + 12/16 \text{ wt}\% \text{ O} + 12/14 \text{ wt}\% \text{ N} \quad (34)$$

Moser states that single phase uranium monocarbide (and monocarbide of uranium/plutonium ratio 4:1) can only be obtained if the nonmetal atom content is less than 50 at.%, and less than 46.5 at.% for PuC.

Moser did not calculate the thermal conductivity of these materials from his diffusivity values because he believed the samples, except for the arc-cast UC, contained enough oxygen and nitrogen to make calculations unrealistic. He recommended the diffusivity be corrected for porosity using a modified Maxwell correction

$$\alpha_T = (1 + 0.5P)\alpha_M, \quad (35)$$

where  $\alpha_T$  = diffusivity at 100% TD,  
 $\alpha_M$  = measured diffusivity, and  
 $P$  = volume fraction porosity.

For comparison with other data, this review calculated conductivity from the diffusivity values of Moser using  $C_p$  values recommended by Olson,<sup>54</sup> and expansion data recommended by Andrew.<sup>59</sup> The values for the 78.8% TD sintered sample were corrected to 100% TD using expression (35). The data are plotted in Fig. 11. Thermal conductivity for the 99% TD cast material is approximated by

$$\lambda = 25.1 - 1.13 \times 10^{-2}T + 4.65 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg} \quad (36)$$

Moser, in three other articles,<sup>53,60,61</sup> also reported the conductivity from 0 to 700°C of stoichiometric 99+ % TD, single phase UC containing 50 ppm oxygen, and no nitrogen. Specific heat data obtained from his laser pulse experiment were used to calculate the conductivity. Moser estimated his experimental precision as about 10%. The specific heat values from the experiment:

$$C_p = 12.32 + 3.329 \times 10^{-3}T - 1.130 \times 10^{-5}T^2, \quad (37)$$

for 273 < T°K < 1000, tabulated in Refs. 53 and 61 are about 3% higher at 1000°K than those recommended by Olson.<sup>54</sup> The conductivities, copied from Moser's curve, are replotted in Fig. 11.

Moser,<sup>4</sup> in a review of electrical and thermal properties, tabulated conductivity values from Ref. 60, adding a value of  $\lambda = 18.0 \text{ W/m} \cdot \text{deg}$  at 1000°C.

11. **Wittenberg**,<sup>58</sup> using a rather unique modulated heat wave technique to measure diffusivity, determined the thermal conductivity of a near stoichiometric UC (4.78 wt% carbon) sample of 96% TD containing 0.1 wt% nickel. Samples were pressed and sintered from powder prepared by carbothermic reduction. Chemical analysis of the material was not discussed; however, results on x-ray analysis indicated the presence of  $\text{UO}_2$ ,  $\text{U}_2\text{C}_3$ , and  $\text{UC}_2$ . The apparatus was calibrated against Armco Iron. The stated measurement uncertainty was 10 to 15%.

In calculating conductivity, Wittenberg used  $C_p$  values from Ref. 62. Using Olson's recommended values to calculate the conductivity results in an increase of about 5% compared to those reported. Wittenberg's tabulated data are plotted in Fig. 11.

12. **Leary**<sup>8</sup> summarized thermal conductivity data on UC, PuC, and (U,Pu)C presented in Refs.

31, 32, and 63. The UC samples were arc cast and solution heat treated at 1300°C for 6 h. Impurity or second phase content of the samples and sample density were not discussed. The conductivity measurements were by an axial heat flow method. Calibration procedures were not reported. The conductivity plotted in Fig. 11 was reported to be expressed by

$$\lambda = 0.0512 + 7.3 \times 10^{-6}T \text{ cal/cm} \cdot \text{s} \cdot \text{deg} \quad (38)$$

for 300 < T°C < 500,

$$(\lambda = 21.42 + 3.054 \times 10^{-3}T \text{ W/m} \cdot \text{deg}) \quad .$$

13. **DeCrescente**<sup>49</sup> reported results of conductivity measurements by a radial heat flow method on hot pressed UC of 9.1% TD, containing 4.85 wt% carbon and 1250 ppm oxygen. No discussion of second phase concentration in the thermal conductivity samples was given. The conductivity, uncorrected for porosity, ranged from 18.7 W/m·deg at 880°C to 17.9 W/m·deg at 1440°C. Applying a simple (1 - P) correction to these data results in values ranging from 20.6 W/m·deg to 19.7 W/m·deg at 880 and 1440°C, respectively, in close agreement with the data of Rough, discussed in the next paragraph.

14. **Rough**<sup>10</sup> reported the thermal conductivity of one 99% TD sample of hyperstoichiometric UC, 5.0 wt% carbon, to be 20. W/m·deg up to 1140°C. Measurements were made by comparative radial heat flow. There was no discussion of concentration of oxygen or nitrogen in the sample. However, the report contained an excellent microstructural study of the various materials used in the general study, of which the conductivity measurement was a rather trivial part. Very interesting observations were made on the time required for transformation of hyperstoichiometric as-cast UC +  $\text{UC}_2$  containing 7.0 wt% carbon to  $\text{U}_2\text{C}_3$ . The time required, for example, at 1200°C was greater than 100 h. Rough's conductivity value is shown in Fig. 12.

15. **Meerson**<sup>66</sup> reported thermal conductivity values ranging from 11.7 to 16.7 W/m·deg at 200 and 700°C, respectively. These values were from measurements on 90% TD hot pressed UC after correction for porosity using a simple 1/(1-P) correction factor. No specifics were given on measurement method or impurity levels in the samples.

16. **Accary**<sup>18</sup> reported conductivity values for 98% TD sintered UC of 19.7 W/m·deg to 18.4 W/m·deg over the temperature range 120 to 236°C. No other experimental details were discussed.

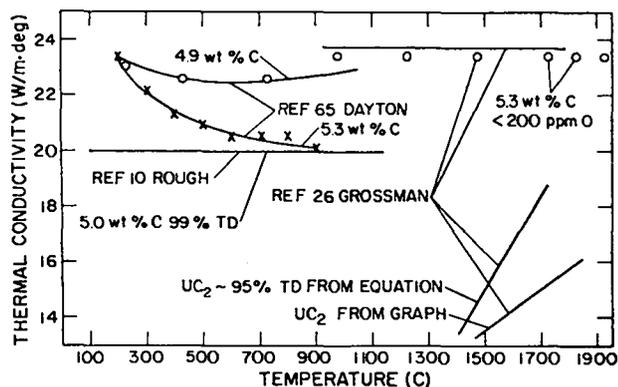


Fig. 12.  
Thermal conductivity values reported for hyperstoichiometric UC.

17. Russell<sup>56</sup> studied the effect of carbon concentration on the thermal conductivity of UC and (U,Pu)C at 70°C. Measurements were made by the Shroder modulated heat wave method.<sup>79</sup> The equipment was calibrated against standards whose conductivities were known to  $\pm 3\%$ . The sample material was prepared by arc melting, hot pressing, and sintering, which resulted in samples of less than 92% TD. No other details concerning samples were given, other than the observation of a uranium grain boundary phase in the hypostoichiometric material.

The data for UC samples, taken from Russell's curve, are summarized in Table III. The data indicate a sharp rise, about 18%, in conductivity from 21.8 W/m·deg at the stoichiometric composition, 4.8 wt% carbon, to 25.9 W/m·deg at 5.0 wt% carbon, and a slight decrease with increasing carbon content to 25.1 W/m·deg at 5.5 wt% carbon. The reported data were corrected to 100% TD using a simple (1-P) correction.

Reference 56 contains an excellent micrographic study of the MC<sub>2</sub> and M<sub>2</sub>C<sub>3</sub> phases which was not directly related to the conductivity study.

18. Grossman<sup>26,21</sup> used a steady state method to measure the thermal conductivity of 100% TD arc-cast hyperstoichiometric UC containing 5.3 wt% car-

bon and less than 200 ppm oxygen. Data reported in tabular form and points taken from Grossman's conductivity curve are shown in Fig. 12. The data below 950°C are apparently results of Dayton and Tipton (see Refs. 21, 65). Microstructural examination made after measurement of conductivity showed slight concentration of UC<sub>2</sub> or U<sub>2</sub>C<sub>3</sub>. Grossman's data summary gave the thermal conductivity as 0.057 cal/cm·s·deg  $\pm 12\%$  (23.85 W/m·deg), from 925 to 1775°C.

The conductivity of UC<sub>2</sub>, 8.7 wt% carbon, was also measured. The samples were prepared by hot pressing UC<sub>2</sub> powder containing 1 vol% nickel. The sample rod was 95% TD before conductivity measurement. After conductivity measurement, the central 1-cm length of rod was found to be 90% TD. Microstructural examination indicated loss of nickel from the central portion of the rod and some UC precipitate.

The conductivity was reported in the data summary as  $\lambda = -0.035 + 4. \times 10^{-5}T$  cal/cm·s·deg  $\pm 15\%$  from 1500 to 2000°K. Values calculated from this expression and taken from Grossman's plotted data are shown in Fig. 12.

19. Dayton<sup>65</sup> reported the conductivity of cast UC samples containing 4.9 wt% carbon and 5.3 wt% carbon measured by a steady state longitudinal heat flow method. Impurity levels and densities of the conductivity specimens were not discussed. Reported tabular data are shown in Fig. 12.

20. Secret<sup>22</sup> reported the thermal conductivity from 100 to 735°C, measured by a steady state heat flow method, of UC containing 5.2 wt% carbon. Samples were prepared from arc-cast material typically 98% TD. Impurity levels were not discussed.

These conductivity values were somewhat higher than values reported by Dayton. The values decreased from 25.1 W/m·deg at 100°C to 22.2 W/m·deg at 400°C, and increased to 25.5 W/m·deg at 735°C.

21. Crane<sup>25,47</sup> measured the conductivity of UC samples containing 4.4 to 5.2 wt% carbon. The samples were prepared by two methods: (a) casting

TABLE III

EFFECT OF CARBON CONTENT ON CONDUCTIVITY<sup>a</sup> AT 70°C

wt% carbon	4.6	4.7	4.8	4.86	4.9	5.0	5.1	5.2	5.3	5.5
$\lambda$ (W/m·deg)	18.5	21.1	21.8	25.4	25.7	25.9	25.9	25.6	25.4	25.1

<sup>a</sup>From Russell's curve.

and pressing and (b) sintering powder made by carburization of uranium hydride with propane. The cast samples ranged in density from 99.0 to 99.8% TD and contained less than 300 ppm oxygen and 100 ppm nitrogen. The sintered materials ranged in density from 90.1 to 98.1% TD, and contained 400 to 1000 ppm oxygen and 100 to 400 ppm nitrogen.

The measurements were made by a comparative steady state longitudinal heat flow method. Microstructural examination showed uranium in grain boundaries of both the cast and sintered hypostoichiometric materials, and Widmanstätten  $UC_2$  in hyperstoichiometric materials.

Crane's tabulated data are plotted in Fig. 13. These data seem atypical in that both cast and sintered hypostoichiometric materials show the higher conductivity above 500°C. Density corrections do not alter the relative conductivity values. The sharp increase of conductivity with temperatures above 500°C also is atypical as compared to results of other investigators.

22. Kubota<sup>67</sup> measured the thermal conductivity of sintered UC and  $UC_2$  from 125 to 400°C using a steady state longitudinal heat flow method. No information about sample material was given other than both the UC and  $UC_2$  samples were 95% TD. The  $UC_2$  data are plotted in Fig. 14.

Grossman's graphed  $UC_2$  data,<sup>26</sup> from 1300 to 1700°C, seem to give a reasonable high temperature extrapolation of the Kubota data. An approximation for the  $UC_2$  conductivity can be written as

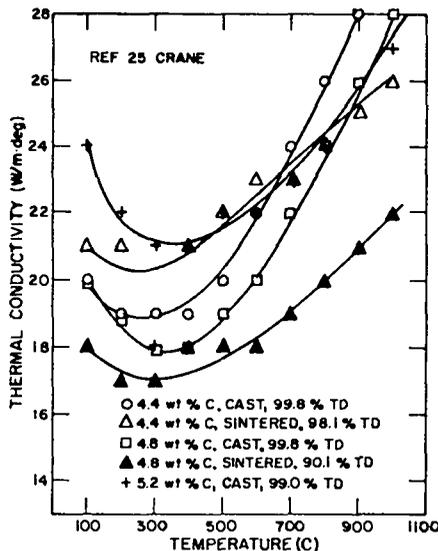


Fig. 13.

Effect of variation in carbon concentration on the thermal conductivity of UC, according to Crane.<sup>25</sup>

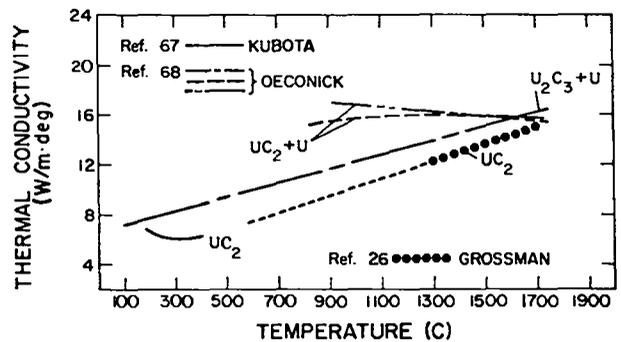


Fig. 14.

Some reported values for the thermal conductivity of  $UC_2$  and  $U_2C_3$ .

$$\lambda = 3.235 + 6.915 \times 10^{-3} T \text{ W/m} \cdot \text{deg} \quad (39)$$

for  $300 < T^\circ\text{C} < 2000$ .

23. DeConick<sup>68</sup> reported the conductivity of  $U_2C_3$  from ambient to 1800°C. Conductivity values were calculated from diffusivities determined by a modulated electron beam method. The specific heat used in the calculation of conductivity was determined by extrapolation of existing literature values below 350°K.<sup>69</sup> The samples contained 7.02 wt% carbon. Quantitative microstructural examination showed the specimens were 85 vol%  $U_2C_3$  + 15 vol% UC. Measured densities were about 100% TD for all samples.

The thermal conductivity was expressed by DeConick as

$$\lambda = 5.05 \times 10^{-2} + 5.63 \times 10^{-5} T \text{ W/cm} \cdot \text{deg} \quad (40)$$

for  $300 < T^\circ\text{K} < 2050$ ,

$$(\lambda = 6.58 + 5.63 \times 10^{-3} T \text{ W/m} \cdot \text{deg}$$

for  $25 < T^\circ\text{C} < 1750$ ). After heat treatments of 1 h at 2170°K (about 1900°C), the conductivity was redetermined for one sample. Metallographic examination after heat treatment showed the specimen consisted of  $UC_2$  + UC. DeConick's data showed the conductivity  $UC_2$  + UC to be significantly higher than his  $U_2C_3$  + UC values. Subsequent determinations on the same sample showed a general decrease in conductivity due to transformation back to  $U_2C_3$ .

24. Summary: Uranium Carbide Thermal Conductivity. The reported thermal conductivity data show wide variation in values over the entire range of temperature in which measurements were made. Such variability is the result of the combined effects

of carbon concentration and volume concentration of second phase dicarbide and sesquicarbide, concentration of oxygen and nitrogen, porosity, and presence of nickel introduced as sintering aid. A quantitative evaluation of these factors is impossible at this time.

The measurements made by Russell<sup>56</sup> at 70°C indicate a maximum in thermal conductivity for nearly hypostoichiometric material with about 3 to 4% decrease for hyperstoichiometric material at 5.5 wt% carbon and about 28% decrease in conductivity for hypostoichiometric material at 4.6 wt% carbon. Russell's value of about 26. W/m·deg was one of the highest reported at lower temperatures. The data of Washington,<sup>41</sup> Wheeler,<sup>42</sup> Carniglia,<sup>17</sup> and Dayton<sup>65</sup> are in general agreement with the results of Russell although Russell's conductivity values are higher. Crane<sup>25</sup> shows a minimum in conductivity for stoichiometric UC. His data seem to show excessive increase in conductivity above 300 or 400°C.

The effects of porosity cannot be qualitatively evaluated. A simple (1-P) correction is perhaps justified at the present state of knowledge.

$$\lambda_M = (1 - P)\lambda_{TD} \quad (41)$$

where  $\lambda_M$  = measured conductivity,  
 $\lambda_{TD}$  = conductivity at 100% TD, and  
 P = volume fraction porosity.

Conflicting data exist on the effects of nickel additions to UC. This will be discussed in the section on (U,Pu)C. However, the data of Wheeler<sup>42</sup> suggest that for pressed and sintered UC, increases in conductivity on the order of 10 to 20% can be expected with nickel additions of 0.1 to 0.2 wt%.

Most investigators believe that oxygen concentrations below about 2500 ppm do not significantly affect the thermal conductivity. Bates<sup>5</sup> and Wheeler<sup>42</sup> provide the most comprehensive study for oxygen concentrations in the range 2 at.% (about 0.3 wt%) to 17 at.% (about 2.0 wt%) oxygen. The results of these two investigations are in reasonable agreement, showing conductivities of about 18 to 19 W/m·deg and 12 to 13 W/m·deg at 100°C, and 18 to 20 W/m·deg and 15 to 18 W/m·deg at 1200°C, respectively, for UC containing about 2 at.% and 17 at.% oxygen.

Although the effects of nitrogen concentration in UC have not been discussed, the literature (e.g., Ref. 70) on nitrides and carbonitrides shows significantly lower thermal conductivities than for UC. Moser<sup>45</sup> indicated an effect of impurity level nitrogen with respect to second phase dicarbide concentration in Eq. (34).

Based on general data trends, qualitative evaluation of porosity levels, and using the data of Bates, Wheeler, and Russell as a qualitative guide to the evaluation of oxygen and carbon effects the following suggested values are given for the thermal conductivity of UC. The calculated conductivities are plotted in Fig. 15.

100% TD UC<sub>1.0</sub>:

$$\lambda = 21.7 - 3.04 \times 10^{-3}T + 3.61 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg} \quad (42)$$

for 50 < T°C < 700,

$$\lambda = 20.2 + 1.48 \times 10^{-3}T \text{ W/m} \cdot \text{deg}$$

for 700 < T°C ≤ 2300.

100% TD UC (0.3 wt% oxygen):

$$\lambda = 21.3 - 4.66 \times 10^{-3}T + 2.40 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg} \quad (43)$$

for 100 < T°C ≤ 2300.

100% TD UC (2.0 wt% oxygen):

$$\lambda = 12.4 + 2.73 \times 10^{-3}T + 6.55 \times 10^{-7}T^2 \text{ W/m} \cdot \text{deg} \quad (44)$$

for 100 < T°C ≤ 2300.

Based on the values of Kubota,<sup>67</sup> DeConick,<sup>68</sup> and Grossman,<sup>26</sup> suggested values for the approximate conductivity of UC<sub>2</sub> and U<sub>2</sub>C<sub>3</sub> are as follows.

100% TD UC<sub>2</sub>:

$$\lambda = 3.24 + 6.92 \times 10^{-3}T \text{ W/m} \cdot \text{deg} \quad (45)$$

for 300 < T°C ≤ 2000.

100% TD U<sub>2</sub>C<sub>3</sub> + 15 vol% UC:

$$\lambda = 6.58 + 5.63 \times 10^{-3}T \text{ W/m} \cdot \text{deg} \quad (46)$$

for 25 < T°C < 1750.

Suggested values of conductivity vs carbon concentration for hypo and hyperstoichiometric UC cannot be given. Effect of carbon content is obviously related to resultant concentration and morphology

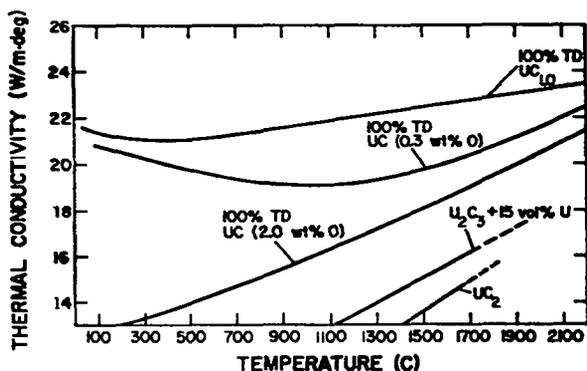


Fig. 15.

Suggested values for the thermal conductivity of UC.

of second phase free uranium,  $UC_2$  and/or  $U_2C_3$ . Concentrations of second phase are also dependent upon thermal history of the sample.

No quantitative evaluation of the effects of measurement method or sample fabrication techniques can be given. However, conductivities reported for pressed, sintered UC when corrected for porosity are, in general, lower than for cast material. This is probably because of generally higher impurity levels of the sintered material and uncertainty in the correction for porosity.

There is disagreement among investigators as to the mode of conduction in the UC system. Wheeler,<sup>42</sup> for example, concluded the lattice contribution to the conductivity is significant at high temperature. Hayes<sup>9</sup> concluded the conductivity above 1000°C is 100% electronic.

## B. Plutonium Carbide

Very few studies of the thermal conductivity of PuC have been reported. Of these, only one investigation has been at temperatures over 500°C.

1. Sheth<sup>1</sup> recommended conductivity values calculated from diffusivity measurements (possibly those of Kruger<sup>45</sup>) on a sample of 84% TD. In calculating conductivities, he used the values 249.269 for the molecular weight of  $PuC_{0.87}$ ,  $C_p$  values from Ref. 71, and thermal expansion data of Pallmer.<sup>72</sup> Sheth expressed the conductivity of 84% TD  $PuC_{0.87}$  as

$$\lambda = 0.00918 + 2.181 \times 10^{-5}T - 6.1819 \times 10^{-9}T^2 \text{ cal/cm} \cdot \text{s} \cdot ^\circ\text{C} \quad (47)$$

for  $400 < T^\circ\text{C} < 1370$ ,

$$\begin{aligned} (\lambda = 3.84 + 9.13 \times 10^{-3}T \\ - 2.59 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg}). \end{aligned}$$

These values are plotted in Fig. 16.

2. Fulkerson<sup>3</sup> reviewed the data of Moser<sup>45</sup> and Leary,<sup>8</sup> and suggested values for the conductivity calculated from Moser's data using the specific heat values of Kruger.<sup>73</sup> These values ranged from about 7.5 W/m·deg at 400°C to 12.5 W/m·deg at 1300°C.

3. Moser<sup>4</sup> reported two conductivity values for PuC. He calculated a value of 7.5 W/m·deg at 25°C from diffusivity measurements on an isostatically pressed sample containing 0.01 wt% (about 100 ppm) oxygen. The value was corrected for porosity using the Maxwell equation. The other value, 14.2 W/m·deg at 500°C, he attributed to measurements by Leary.<sup>8</sup>

4. Leary<sup>8</sup> reported the conductivity of  $PuC_{0.87}$  from 200 to 400°C measured by comparative axial heat flow. Samples were prepared by arc melting and casting plutonium containing less than 200 ppm total impurity and spectrographic grade graphite. No other details were given. Leary's values shown in Fig. 16 were expressed in linear form as

$$\lambda = 0.0155 + 38.5 \times 10^{-6}T \text{ cal/cm} \cdot \text{s} \cdot ^\circ\text{C} \quad (48)$$

for  $200 < T^\circ\text{C} < 400$ ,

$$(\lambda = 6.5 + 1.61 \times 10^{-2}T \text{ W/m} \cdot \text{deg}).$$

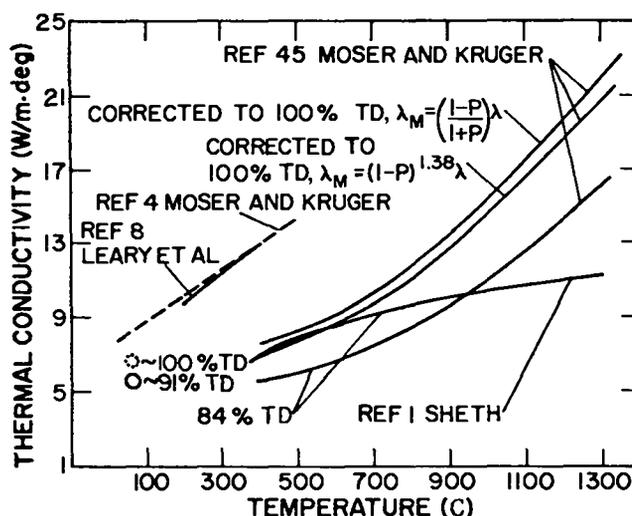


Fig. 16.

Reported thermal conductivities for PuC.

5. Russell<sup>56</sup> reported a conductivity value of 5.56 W/m·deg at 70°C for one sample of PuC<sub>1-x</sub> of about 91% TD (12.2 g/cm<sup>3</sup>). This value, corrected to 100% TD using the correction given by expression (49) is shown in Fig. 16.

$$\lambda_M = \left( \frac{1-P}{1+P} \right) \lambda_{TD} \quad (49)$$

6. Moser<sup>45</sup> reported the thermal diffusivity of 83.8% TD sintered PuC containing 4.12 wt% carbon and 0.61 wt% oxygen + nitrogen over the temperature range 400 to 1300°C. Microstructural examination of the sample material showed the presence of up to 10 vol% Pu<sub>2</sub>C<sub>3</sub>.

The apparatus was calibrated against an Al<sub>2</sub>O<sub>3</sub> standard, indicating the reported ±5% measurement accuracy and ±5% precision. Moser did not calculate conductivity from his measurements.

Conductivities calculated using the specific heat values suggested by Olson<sup>54</sup> and expansion values suggested by Andrew are plotted in Fig. 16. Values corrected for porosity by expressions (4) and (49) are also shown.

7. **Summary: Thermal Conductivity of Plutonium Carbide.** The diffusivity data of Moser offer the only information above 500°C. The conductivity values calculated from Moser's data show approximately the same temperature dependence as the data of Leary, but are considerably lower than Leary's data would be, extrapolated from 400 to 1300°C. This is probably because of the significant Pu<sub>2</sub>C<sub>3</sub> concentration in Moser's sample. However, because no other data exist above 500°C, it is suggested that conductivity of 100% TD PuC be used, as calculated from Moser's data and corrected by expression (49). The values are expressed by

$$\lambda = 7.45 - 4.04 \times 10^{-3}T + 1.20 \times 10^{-5}T^2 \text{ W/m} \cdot \text{deg} \quad (50)$$

for 100 < T°C ≤ 1300. Values for high purity single phase PuC<sub>1-x</sub> are probably significantly higher.

No quantitative evaluation of the effects of impurity level, porosity, carbon concentration, or fabrication method can be given.

### C. Uranium-Plutonium Carbide

Since 1967 several of the investigators previously discussed have reviewed the available data on the thermal conductivity of (U,Pu)C, namely, Sheth,<sup>1</sup>

Kerrisk,<sup>74</sup> Washington,<sup>41</sup> Fulkerson,<sup>3</sup> and Leary.<sup>6</sup> All of these authors have recognized apparently inconsistent information or areas of lack of information which prevent quantitative assessment of the effects of composition, impurities, and second phase concentration on thermal conductivity. Observations and conclusions of these reviews relating to (U,Pu)C will be summarized and most of the individual articles will be subsequently discussed.

1. Sheth<sup>1</sup> only considered the recent reviews of Kerrisk<sup>74</sup> and Washington<sup>41</sup> in suggesting the values of Kerrisk.

2. Kerrisk<sup>74</sup> reviewed the articles of Johnson,<sup>75</sup> Washington,<sup>41</sup> Bocker,<sup>76</sup> VanCraeynest,<sup>37</sup> Pascard,<sup>43</sup> Moser,<sup>45</sup> and Leary.<sup>8</sup> After correcting reported values for porosity using expression (49) he concluded that Washington's values were too low. Kerrisk suggested that the conductivity for 100% TD (U<sub>0.8</sub>Pu<sub>0.2</sub>)C, as shown in Fig. 17, be expressed as

$$\lambda = 0.03036 + 2.537 \times 10^{-5}T - 6.685 \times 10^{-9}T^2 \text{ cal/cm} \cdot \text{s} \cdot \text{C} \quad (51)$$

for 100 < T°C < 1900,

$$\lambda = 0.0545 \text{ cal/cm} \cdot \text{s} \cdot \text{C}$$

for T°C > 1900. Equivalent values are given by expression (52).

$$\lambda = 12.70 + 10.62 \times 10^{-3}T - 2.797 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg} \quad (52)$$

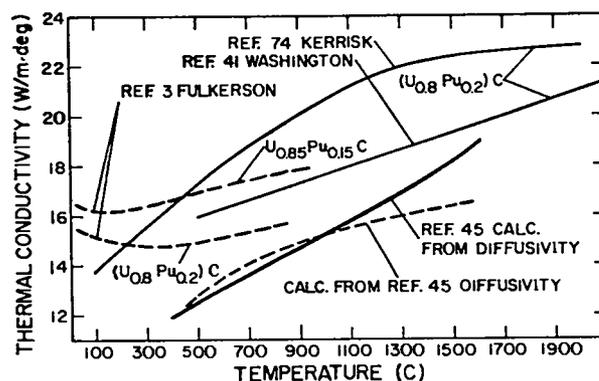


Fig. 17.

Values for the thermal conductivity of (U,Pu)C recommended by recent reviews compared to values calculated from thermal diffusivity data of Moser.

for  $100 < T^{\circ}\text{C} < 1900$  ,

$$\lambda = 22.80 \text{ W/m}\cdot\text{deg}$$

for  $T^{\circ}\text{C} > 1900$ . It was also concluded that sufficient data do not exist to permit quantitative evaluation of the effect of varying the Pu/(U + Pu) ratio.

3. **Washington**<sup>41</sup> reviewed the work of Leary,<sup>6,8</sup> Johnson,<sup>75</sup> Pascard,<sup>43</sup> VanCraeynest,<sup>37</sup> Wheeler,<sup>42</sup> Bocker,<sup>76</sup> Tachis,<sup>48</sup> and Wittenberg.<sup>58</sup> The data from these references represented both arc-cast and sintered specimens ranging from 91% TD to about 98% TD, from 15 to 30% PuC, and results of both steady state and diffusivity measurement methods. The conductivity values for  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  ranged from about 15.5 W/m·deg to about 21 W/m·deg at 1000°C. In-reactor results of Tachis for a 75% TD sample ranged from 10 W/m·deg at 1050°C to about 10.5 W/m·deg at 1600°C. Only Leary and Wheeler investigated the effect of varying the Pu/(U + Pu) ratio.

Washington concluded that expression (49) should be used for porosity correction, and that it was not possible to give preferred values for non-stoichiometric material. His preferred values, shown in Fig. 17, for the conductivity of 100% TD  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  were expressed as

$$\lambda = 16. + 3.4 \times 10^{-3} (T-500) \text{ W/m}\cdot\text{deg} \quad (53)$$

for  $500 < T^{\circ}\text{C} < 1900$  .

4. **Fulkerson**<sup>3</sup> reviewed information on  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  and  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  published by VanCraeynest,<sup>37</sup> Leary,<sup>8</sup> Johnson,<sup>75</sup> Moser,<sup>45</sup> and Milet.<sup>38</sup> Thermal conductivities were calculated from diffusivities using calculated  $C_p$  values for the mixed carbide and the TD of  $\text{UC}_{1.0}$ .<sup>4</sup> The conductivity data of Ref. 75 was corrected to 100% TD (method not indicated) and an average curve established. After analysis of these data, Fulkerson suggested the thermal conductivity of  $(\text{U,Pu})\text{C}$  be given by

$$\lambda = \frac{L_0 T}{\rho} + \frac{1970}{T} \text{ W/m}\cdot^{\circ}\text{K} , \quad (54)$$

where  $L_0$  is the theoretical Lorenz number, and  $T = ^{\circ}\text{K}$ . The resistivity values of Leary<sup>8</sup> for  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  and Milet<sup>38</sup> for  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  were used to calculate the conductivity curves shown in Fig. 17. Fulkerson concluded that the conductivity calculated from Moser's data was supported by these calculations. He also concluded that more reliable high

temperature information, for both thermal conductivity and electrical resistivity, would be required to resolve the apparent lack of agreement among the existing data on conductivity.

5. **Leary**<sup>6</sup> discussed the  $(\text{U,Pu})\text{C}$  work of Russell,<sup>56</sup> Wittenberg,<sup>58</sup> and Leary.<sup>8</sup> Leary,<sup>6</sup> in this review, did not propose recommended values. His conclusions are summarized as follows.

(a) Replacement of uranium by plutonium in UC causes a significant decrease in thermal conductivity in the temperature range 250 to 1000°C.

(b) Maximum conductivity is observed for near-stoichiometric  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$ .

(c) Nickel sintering aid lowers the conductivity of  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}_{0.95}$ .

(d) Excessive amounts of oxygen and nitrogen lower the thermal conductivity of the carbides.

(e) There is no satisfactory method for quantitative adjustment for the effects of porosity on the conductivity.

(f) The presence of fission products probably decreases the conductivity.

(g) Additional work should be done on the conductivity of fully characterized carbides, and should be related to measurement of the electrical resistivity on the same materials.

6. **Moser**<sup>45</sup> did not calculate thermal conductivity values from his flash diffusivity measurements on sintered, 74.8% TD  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$ . Sample preparation and apparatus calibration was briefly discussed in Sec. III.A.10. The mixed carbide specimens contained 4.66 wt% carbon, 0.202 wt% oxygen, and 0.0206 wt% nitrogen. The effective carbon concentration, calculated by expression (34), was reported as 4.83 wt% or 50.2 at.%. Metallographic analysis revealed no significant concentration of second phase dicarbide or sesquicarbide. Moser used Eq. (35) to correct the diffusivity values for porosity.

The conductivity values plotted in Fig. 17 were calculated from Moser's results using the specific heat recommended by Olson<sup>54</sup> and thermal expansion recommended by Andrew.<sup>59</sup> These values were corrected for porosity using expression (35). The validity of this correction for samples of low density is probably questionable.

7. **Russell**<sup>56</sup> in the work discussed in part in Sec. III.A.17, also determined the thermal conductivity at 70°C of  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  having carbon contents ranging from about 4.5 to 5.5 wt%. The samples containing 4.5 to about 5.3 wt% carbon were arc-cast material, and those containing more than 5.3 wt% carbon were prepared by hot pressing. One hypostoichiometric specimen was prepared by cold

pressing and sintering. The conductivity values, other than that for the cold pressed sample, fell on a smoothly varying curve from 12.6 W/m·deg at 4.5 wt% carbon to a maximum of 17. W/m·deg at 5.03 wt% carbon (approximately stoichiometric) then to 12.6 W/m·deg at 5.5 wt% carbon. Densities of the pressed specimens were reported as greater than 95% TD. It was not clear that the data were corrected for porosity. (Russell used a simple  $(1-P)$  correction to his UC data, Sec. III.A.17.) He concluded that the low conductivity values compared to arc-cast specimens of some of the sintered specimens were the result of porosity and that oxygen concentrations less than 0.2 wt% did not significantly affect the conductivity. This was not clear because conductivity values reported for samples containing 0.57 and 0.17 wt% oxygen at two carbon concentrations were higher than conductivities reported for samples containing 0.17 and 0.14 wt% oxygen, respectively, at the same respective carbon concentrations.

8. Horspool<sup>77</sup> made only a brief comment on the conductivity of (U,Pu)C in reference to the work of Wheeler<sup>42</sup> and Wittenberg.<sup>58</sup> However, a pertinent reference was made to work of Browning et al.,<sup>78</sup> concerning phase equilibria. Horspool suggests that "the two-phase region containing a solid solution of the monocarbides and a solid solution of the sesquicarbides should exist up to temperatures over 2000°C for plutonium concentrations  $(Pu/U + Pu) \approx 0.15$  to 0.40."

9. Bocker<sup>76</sup> reported the thermal conductivity, calculated from measured values of thermal diffusivity, specific heat, and density. The samples were sintered  $(U_{0.8}Pu_{0.2})C$  at 92% TD. The material, prepared from powder processed from the hydride, contained 800 ppm oxygen and 800 ppm nitrogen. No indication was given as to the  $C_p$  values or porosity corrections used in the calculation of the conductivity. Values taken from Bocker's graph are plotted in Fig. 18. Assuming these data are uncorrected for porosity, the conductivities corrected for porosity using expression (28), are also shown.

Bocker referenced Milet<sup>40</sup> for a description of the measurement methods used. The work described by Milet, in Refs. 40 and 38, calculated the thermal conductivity from resistivity, density, and specific heat using the Ewing formula, Eq. (55).

10. Lorenzelli<sup>80</sup> exhibited the conductivities reported by VanCraeynest<sup>37</sup> in the study of nitrides, carbonitrides, and carbides. (This work will be discussed in the next subsection.) Lorenzelli made the pertinent observation that additions of nickel sintering aid to pressed and sintered carbides cause high

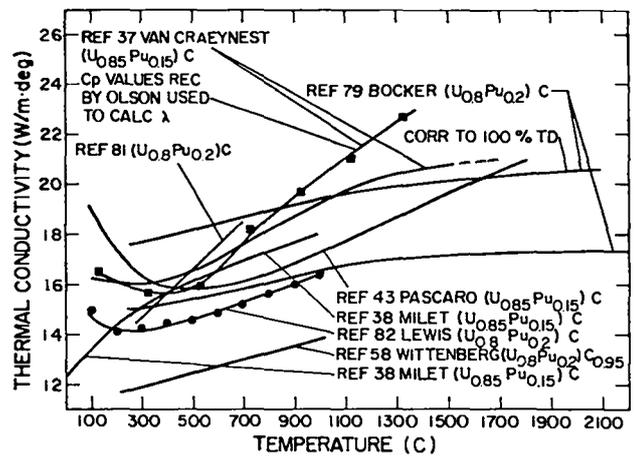


Fig. 18. Variability in thermal conductivity values reported for (U,Pu)C.

enough densification at 1300°C to prevent CO release, resulting in  $M_2C_3$  concentrations of 10 to 15 vol% in materials having equivalent carbon-to-metal ratios of 1.0. Without nickel sintering aid, less than 3 vol%  $M_2C_3$  is guaranteed.

11. VanCraeynest<sup>37</sup> reported thermal conductivity calculated from thermal diffusivity measurements, over the temperature range 100 to 1700°C, on several (U,Pu)C samples having a uranium/plutonium ratio of 85/15. Sample fabrication methods, oxygen and nitrogen impurity, and second phase dicarbide or sesquicarbide concentrations were not discussed. Samples were 91.5% TD.

Two diffusivity measurement methods were used: a modulated heat wave technique below 900°C and a modulated electron beam technique at higher temperatures. The methods were discussed in this reference.

In calculating the conductivity of (U,Pu)C, VanCraeynest used the specific heat of UC reported by Krikorian.<sup>57</sup> It is not clear whether his values were corrected for density or thermal expansion. A simple  $(1-P)$  correction to this data would give about 17.5 and 23 W/m·deg at 300 and 1700°C, respectively. The conductivity values tabulated by VanCraeynest are plotted in Fig. 18. The value shown at 1700°C is estimated from his plotted diffusivity and the  $C_p$  and density values which were apparently used to calculate his value at 1300°C.

A plot of conductivity values calculated from VanCraeynest's diffusivities using specific heat estimates calculated from Olson's suggested values for

UC and PuC is shown in Fig. 18. The higher conductivity above 700°C reflects the relatively high  $C_p$  recommended for PuC.

Using the Wiedemann-Franz law, VanCraeynest calculated the electronic and lattice conductivities from the measured thermal conductivities and electrical resistivity. The reported resistivity values used in the calculations were not discussed. The electronic and lattice conductivities were, respectively, 8.3 W/m·deg and 8.0 W/m·deg at 100°C and 15.4 W/m·deg and 5.0 W/m·deg at 1300°C, indicating a significant lattice contribution above 1000°C.

12. **Pascard's** discussion<sup>43</sup> of nitrides and carbonitrides included the plot of thermal conductivity of (U,Pu)C shown in Fig. 18. The data points shown were taken from Pascard's curve. Although no specifics were given as to composition or impurity levels, the discussion seems to indicate the samples were pressed and sintered approximately stoichiometric,  $(U_{0.85}Pu_{0.15})C$  of about 95% TD.

Diffusivity measurements were by two phase shift methods, modulated heat wave and modulated electron beam described in Ref. 43. Corrections for porosity apparently were not made. A simple (1-P) correction would produce about a 5% increase in the values shown.

Pascard gave an important comment on the use of nickel sintering aid. Nickel-free carbides generally do not exhibit any trace of higher carbides  $M_2C_3$  and  $MC_2$ . Starting with the same powder, nickel addition can result in the presence of more than 10%  $M_2C_3$ , even at 1600°C; this demonstrates that nickel-containing carbides are not in thermodynamic equilibrium, the high density associated with nickel prevents CO outgassing.

13. **Wittenberg**,<sup>58</sup> in conjunction with the work described in Sec. III.A.11, also measured the thermal diffusivity of two sintered (U,Pu)C specimens. One of these was  $(U_{0.8}Pu_{0.2})C_{0.95}$  + 0.1 wt% nickel at 96.8% TD. This sample contained 4.82 wt% carbon and concentrations of  $M_2C_3$ ,  $MC_2$ , and  $MO_2$  detectable by x-ray analysis. The second sample of  $(U_{0.8}Pu_{0.2})C_{0.97}$  at 92.2% TD contained 4.46 wt% carbon, no nickel, and showed a barely detectable concentration of  $MC_2$  by x ray.

The diffusivity data were plotted as constant values from 200 to 1000°C; the diffusivity of the sample containing nickel was 0.028 cm<sup>2</sup>/s as compared to 0.039 cm<sup>2</sup>/s for the nickel-free sample. Wittenberg did not calculate thermal conductivities from these data.

The data plotted in Fig. 18 were calculated from the diffusivity data using specific heat values calculated from the values for UC and PuC

recommended by Olson.<sup>54</sup> The thermal conductivity of the sample containing nickel ranged from 8.4 W/m·deg at 250°C to 9.9 W/m·deg at 1000°C.

14. Data from Ref. 81 are included in Fig. 18 to show their similarity to the conductivity of  $(U_{0.85}Pu_{0.15})C$  calculated from VanCraeynest's data using Olson's recommended  $C_p$  values. These results were from arc-cast and annealed  $(U_{0.8}Pu_{0.2})C_{1.0}$  specimens, no other specifics being given. Although the measurement method was not discussed, the comparative longitudinal heat flow apparatus was probably used.

15. **Lewis and Kerrisk**<sup>82</sup> made flash diffusivity measurements on  $(U_{0.8}Pu_{0.2})C$  containing 13 vol%  $(U_{0.8}Pu_{0.2})_2C_3$ , 5.15 wt% carbon, 255 ppm nitrogen, 230 ppm oxygen, and less than 4000 ppm nickel (added as sintering aid). The apparatus was calibrated against a known molybdenum reference sample.

Conductivities plotted in Fig. 18 were calculated from the diffusivity using the expansion data for  $(U_{0.8}Pu_{0.2})C_{0.95}$  recommended by Andrew<sup>59</sup> and  $C_p$  values calculated from the specific heat of UC and PuC recommended by Olson.<sup>54</sup>

16. **Milet**<sup>38</sup> calculated the thermal conductivity of  $(U_{0.85}Pu_{0.15})C$  from his electrical resistivity measurements discussed in Sec. II.C.5 from Ewing's<sup>83</sup> expression

$$\lambda = 2.61 \times 10^{-8} \left( \frac{T}{\rho} \right) - 2 \times 10^{-17} \left( \frac{T}{\rho} \right)^2 \left( \frac{1}{C_p d} \right) + 97 \left( \frac{C_p d^2}{MT} \right) \text{cal/cm} \cdot \text{s} \cdot \text{deg} \quad (55)$$

where  $T = ^\circ K$ ,  $\rho = \Omega \text{cm}$ ,  $C_p = \text{cal/g} \cdot \text{deg}$ ,  $d = \text{g/cm}^3$ , and  $M = \text{average molecular weight}$ . The specific heat was calculated from Newman and Kopp's rule,  $C_p = \sum_i x_i C_{pi}$  where  $x_i$  is the concentration of the  $i$ th species. The temperature-dependent specific heat of UC was taken from Kubaschewski<sup>84</sup> ( $C_p = 13.4 + 1.02 \times 10^{-3}T - 1.46 \times 10^{-5}T^{-2}$ ), and of PuC from Kruger<sup>73</sup> ( $C_p = 13.08 + 11.44 \times 10^{-4}T - 3.232 \times 10^{-5}T^{-2}$ ). Milet's calculated values are plotted in Fig. 18.

17. **Leary**<sup>8</sup> summarized the conductivity measurements determined by comparative axial heat flow (see also Refs. 30 and 32) on  $(U_{0.9}Pu_{0.1})C$ ,  $(U_{0.8}Pu_{0.2})C$ , and  $(U_{0.7}Pu_{0.3})C$  over the temperature range 200 to 400°C. It is assumed the arc-cast specimens were greater than 95% TD. Part of this

work on UC and PuC were discussed in Secs. III.A.12 and III.B.4. Leary found that additions of plutonium to the UC lattice decreased the thermal conductivity in a manner analogous to the resistivity increase discussed by Pascard.<sup>24</sup> Concentrations of 20 and 30 mol%, respectively, lowered the thermal conductivity of UC (at 400°C) to about 78 and 67% of the value for the unalloyed carbide. Leary expressed the conductivity data plotted in Fig. 19 by the following linear equations.

(U<sub>0.9</sub>Pu<sub>0.1</sub>)C:

$$\lambda = 0.0426 + 18.0 \times 10^{-6}T \text{ cal/cm} \cdot \text{s} \cdot \text{deg} \quad (56)$$

for 265 ≤ T°C ≤ 460.

(U<sub>0.8</sub>Pu<sub>0.2</sub>)C:

$$\lambda = 0.0340 + 20.0 \times 10^{-6}T \text{ cal/cm} \cdot \text{s} \cdot \text{deg} \quad (57)$$

for 200 ≤ T°C ≤ 400.

(U<sub>0.7</sub>Pu<sub>0.3</sub>)C:

$$\lambda = 0.0220 + 35.0 \times 10^{-6}T \text{ cal/cm} \cdot \text{s} \cdot \text{deg} \quad (58)$$

for 200 ≤ T°C ≤ 400. Multiplication of these expressions by 418.4 will give the conductivity in W/m·deg.

18. Wheeler,<sup>42</sup> in addition to the studies on UC discussed previously, measured the diffusivity of

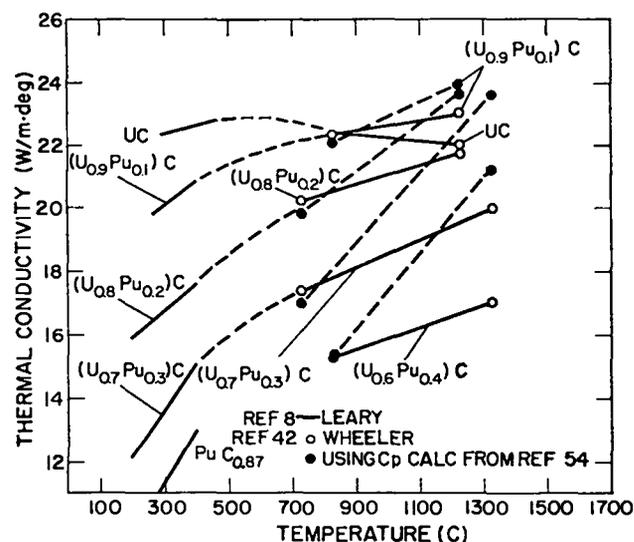


Fig. 19.

Effect of plutonium/uranium ratio on the thermal conductivity of (U,Pu)C, according to Leary<sup>8</sup> and Wheeler.<sup>42</sup>

(U,Pu)C of varying Pu/U ratios over the range 800 to 1300°C. The specimens were fabricated by multiple arc casting, and densities are assumed to have been greater than 95% TD. Impurity levels were not discussed. The reported carbon concentrations and nominal compositions are listed in Table IV.

The reported conductivity was calculated from the diffusivity using Krikorian's specific heat for UC. These values, taken from Wheeler's curves, are plotted in Fig. 19. The dashed lines were drawn to aid in relating values for similar compositions reported by Leary.

Using the calculated C<sub>p</sub> from UC and PuC data of Ref. 54 for the various Pu/U ratios, and C<sub>p</sub> for UC calculated from Krikorian's equation, Wheeler's conductivity values were modified to reflect the specific heats given in Ref. 54 as shown in Fig. 19. The indicated temperature dependence seems excessive.

19. Johnson<sup>75</sup> studied the effect of density on the thermal conductivity of (U<sub>0.8</sub>Pu<sub>0.2</sub>)C using a comparative heat-flow apparatus calibrated against known standards. Samples were prepared by pressing and sintering. The materials contained less than 100 ppm total oxygen and nitrogen. Metallographic examination of the specimens indicated the materials were single phase (U,Pu)C except for two samples which showed trace concentrations of second phase (not identified). This article summarized the work reported in Ref. 85.

Johnson fitted Eq. (59) to his data. The porosity correction seems quite high on considering the resultant values for 100% TD material.

$$\lambda = (3.94 \times 10^{-2} + 2.53 \times 10^{-5} T) \left( \frac{1-P}{1+4.01P} \right) \text{ cal/cm} \cdot \text{s} \cdot \text{deg} \quad (59)$$

TABLE IV  
COMPOSITION OF  
DIFFUSIVITY SPECIMENS

Composition (nominal)	wt% Plutonium		at.% Carbon Before
	Before	After	
(U <sub>0.9</sub> Pu <sub>0.1</sub> )C	9.4	9.2	44.8
(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	19.5	18.8	48.5
(U <sub>0.7</sub> Pu <sub>0.3</sub> )C	28.3	27.4	45.9
(U <sub>0.6</sub> Pu <sub>0.4</sub> )C	36.9	37.3	48.8

or,

$$\lambda = (16.48 + 1.059 \times 10^{-2} T) \left( \frac{1-P}{1+4.01P} \right) \text{ W/m}\cdot\text{deg} \quad (60)$$

where  $100 < T^{\circ}\text{C} < 1000$ . The conductivities calculated from Eq. (59) and experimental data tabulated in Ref. 75 are shown in Fig. 20. The temperature dependence of the calculated values for 100% TD material seems excessive. Note that these values are higher than shown by Leary<sup>8</sup> in Fig. 19. This result is perhaps another indication of the need to consider the porosity correction as dependent upon temperature and pore morphology.

20. Bradbury<sup>86</sup> assessed the thermal conductivity of  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  from in-pile experiments designed to study fuel swelling and fission product migration. The two samples used in the conductivity study were arc-cast at about 99% TD and sintered at less than 96% TD. The arc-cast material contained less than 900 ppm oxygen + nitrogen, and the sintered material less than 2000 ppm. The in-reactor sample temperatures at an operating power level of 58 MW were calculated based on a conductivity of  $15 \text{ W/m}\cdot\text{deg}$  deduced from Ref. 8, and com-

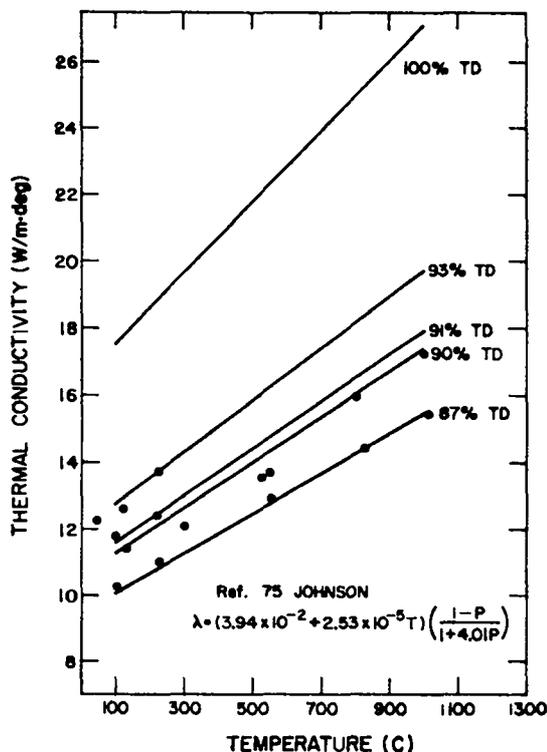


Fig. 20.

Effect of porosity on the thermal conductivity of  $(\text{U,Pu})\text{C}$ , according to Johnson.<sup>75</sup>

pared with measured temperature. The author concluded the value of  $15 \text{ W/m}\cdot\text{deg}$  gave fairly accurate estimates of fuel operating temperature.

21. Stahl<sup>87</sup> reported in-pile measurement of the thermal conductivity of  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}_{0.95}$  specimens containing 2000 to 4000 ppm oxygen. The experiment consisted of measuring the temperature drop from fuel center to clad surface of identical UC and  $(\text{U,Pu})\text{C}$  specimens under like irradiation conditions. The combined fuel and clad-gap conductances were estimated and, using a conductivity value of  $23 \text{ W/m}\cdot\text{deg}$  for UC to estimate the gap conductance, the thermal conductivity of  $(\text{U,Pu})\text{C}$  was calculated.

Specimens were fabricated of pressed and sintered powder produced by carbothermic reduction of oxides. Specimens of 95% TD containing 0.1 wt% nickel sintering aid and specimens of 89% TD without the sintering aid were tested.

The in-pile measurements over a temperature range of 700 to  $1010^{\circ}\text{C}$  resulted in conductivity values from 12.1 to  $24.7 \text{ W/m}\cdot\text{deg}$  and an average of  $19.3 \text{ W/m}\cdot\text{deg}$  for the three specimens tested.

22. Summary: Thermal Conductivity of Uranium-Plutonium Carbide. Introductory remarks in the summary (Sec. III.A.24) for the thermal conductivity of UC are applicable here. The interplay of effects of carbon, oxygen, nitrogen and sesquicarbide or dicarbide concentration, porosity, the apparent anomalous effect of nickel additions, and the effect of varying the plutonium/uranium ratio are impossible to separate and evaluate quantitatively at the present state of knowledge.

The work reported by Leary<sup>8</sup> and Wheeler<sup>42</sup> provide the only systematic study of the effect of varying the Pu/U ratio over a wide range. Leary's data over the range 200 to  $450^{\circ}\text{C}$  indicate about a 20 to 33% decrease in thermal conductivity for  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  and  $(\text{U}_{0.7}\text{Pu}_{0.3})\text{C}$ , respectively, compared to his UC data at  $400^{\circ}\text{C}$ . The higher temperature data of Wheeler from 800 to  $1300^{\circ}\text{C}$  show the same effect but with decreases on the order of 10 and 20% at  $900^{\circ}\text{C}$ . The conductivities reported by Leary and Wheeler for  $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$  are in general higher than reported by other investigators. The results of these investigations should be used only as a guide to estimating the effects of variation in the plutonium/uranium ratio.

A study by Russell<sup>56</sup> on the effect of carbon concentration showed a 20% decrease in thermal conductivity for  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  containing 4.5 or 5.5 wt% carbon compared to a maximum conductivity determined for approximately stoichiometric  $(\text{U}_{0.85}\text{Pu}_{0.15})\text{C}$  containing 5.03 wt% carbon.

Johnson's data represent the only systematic study of the effect of porosity. However, the data for samples having densities over 90% TD cover a very limited temperature range although conductivities of 87 and 90% TD materials were investigated to 1000°C. The porosity correction used in conjunction with the reported linear temperature dependence results in conductivity values for 100% TD ( $U_{0.8}Pu_{0.2}C$ ) which seem to be excessive above 300°C.

Porosity corrections used by the various authors represent a wide range of values of the correction factor for a given volume fraction of void. For example, values calculated for 90% TD material range from 0.90 for the simple  $(1-P)$  correction to 0.64 for the correction used by Johnson.

The limited information available seems to indicate that small amounts of nickel sintering aid lower the conductivity of (U,Pu)C. Most investigators agree that concentrations of oxygen above about 2000 ppm significantly lower the conductivity. The (U,Pu)N compounds, not discussed in this review, show lower conductivity than the (U,Pu)C. If these effects are considered in conjunction with the observations of Horspool (Sec. III.C.8), Lorenzelli (Sec. III.C.10), Pascard (Sec. III.C.12), and Moser (Sec. III.C.6) and Eq. (34) on the effects of oxygen and nitrogen content on resultant  $M_2C_3$  concentrations for approximately stoichiometric compositions, it is not surprising that effects of porosity, composition, and impurity levels have not been resolved quantitatively.

The information available on effects of irradiation<sup>86,87</sup> on the thermal conductivity does not permit a quantitative evaluation at elevated temperature.

There is a qualitative difference in the general shape of the thermal conductivity curves. In general, results obtained by transient methods show a decrease in conductivity of (U,Pu)C from ambient to 300 or 400°C while the few results obtained by steady state methods do not. Exceptions are the data of Moser and Wittenberg.

On consideration of the information presented in Figs. 17 through 20, it is clear that there is disagreement as to the conduction mode in (U,Pu)C. Leary's Lorenz number calculations, over a limited temperature range, and Moser's data seem to indicate pure electronic conduction. However, the work of Wheeler, Fulkerson, and VanCraeynest indicates a significant lattice contribution at elevated temperature. All investigators seem to agree that the thermal conductivity of (U,Pu)C approaches that of UC above 2000°C.

It is believed that, at the present state of knowledge, the thermal conductivity of 100% TD

( $U_{0.8}Pu_{0.2}C$ ) or ( $U_{0.85}Pu_{0.15}C$ ) can be expressed by Eq. (61). Differences due to the plutonium/uranium ratio fall within the limits of uncertainty. A decrease in conductivity from ambient to about 400°C is given by this equation illustrated in Fig. 21. However, the uncertainty in conduction mode is illustrated by the cross-hatched area. The suggested values were chosen to reflect roughly a 20% lower conductivity than UC at temperatures below 700°C, as indicated by most investigations; a positive temperature dependence above 500°C such that the conductivity approaches that of UC above 2000°C; and high enough overall values to reflect less than 2500 ppm total oxygen and nitrogen concentrations.

For 100% TD ( $U_{0.8}Pu_{0.2}C$ ):

$$\lambda = 17.5 - 5.65 \times 10^{-3}T + 8.14 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg} \quad (61)$$

for  $50 < T^\circ\text{C} \leq 500$ , and

$$\lambda = 12.76 + 8.71 \times 10^{-3}T - 1.88 \times 10^{-6}T^2 \text{ W/m} \cdot \text{deg}$$

for  $500 < T^\circ\text{C} \leq 2300$ .

Values for the thermal conductivity of (U,Pu)<sub>2</sub>C<sub>3</sub> or (U,Pu)C<sub>2</sub> above 300°K cannot be given at the present state of knowledge.

#### D. Recommendations and Conclusions: Thermal Conductivity

Suggested values for the thermal conductivity of UC, PuC, and (U,Pu)C are summarized in Table V. The tabulated values are calculated by equations of the form  $\lambda = a + bT + cT^2$ . Reliability of the suggested values probably falls within  $\pm 10\%$  for UC and  $\pm 15\%$  for PuC and (U,Pu)C.

Suggested values for the conductivity of ( $U_{0.8}Pu_{0.2}C$ ) are lower than the values recommended by Kerrisk<sup>74</sup> (Fig. 17) at temperatures above 1200°C. At 1200°C, the suggested value is approximately 5% lower than Kerrisk's curve. The values are consistently higher than those recommended by Washington<sup>41</sup> (Fig. 17). At 1200°C, the Washington curve is approximately 20% lower than the value suggested by this review. These concluding remarks are applicable to the available information on UC and PuC as well as (U,Pu)C; however, we emphasize the relation to the mixed carbides. On consideration of the information

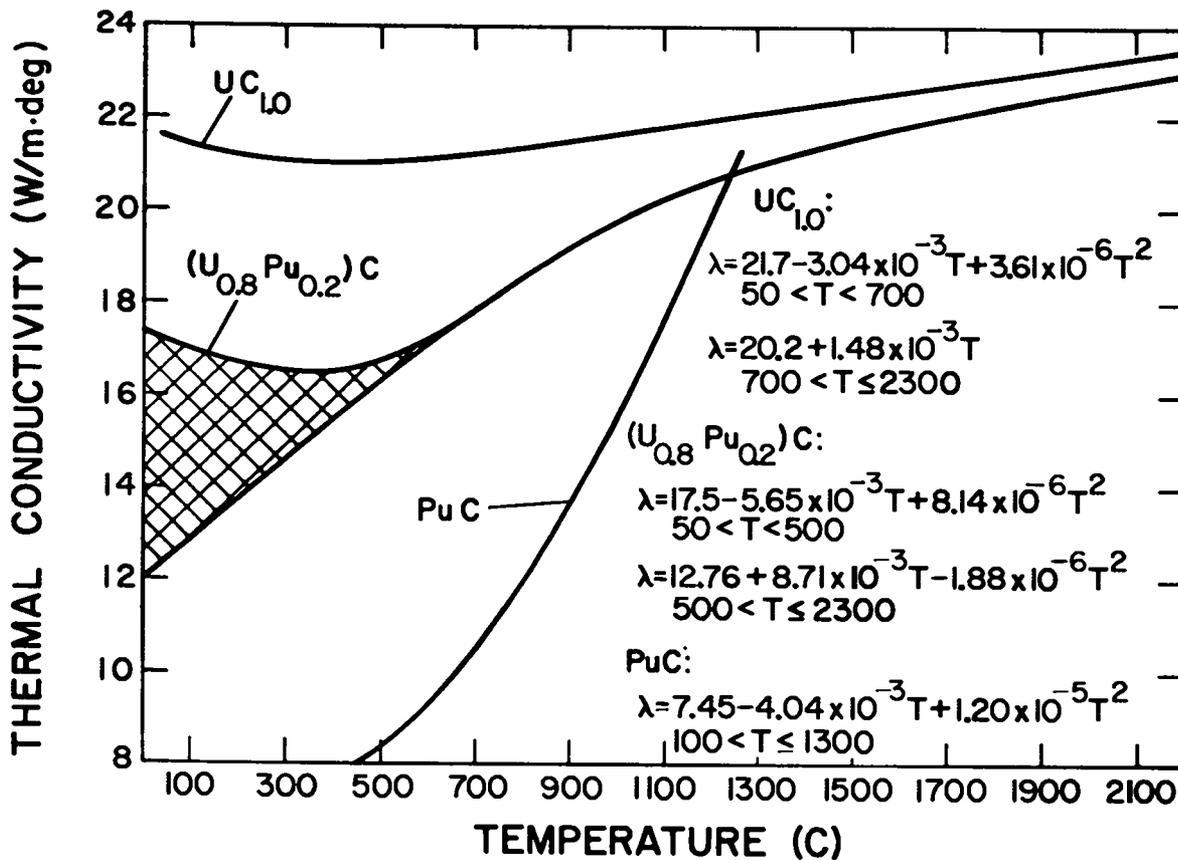


Fig. 21.  
Suggested values for the thermal conductivity of UC, PuC, and (U,Pu)C.

discussed in this review, the conclusions of Fulkerson<sup>3</sup> (Sec. III.C.4) and Leary<sup>6</sup> (Sec. III.C.5) are still valid. These and the conclusions of this review can be briefly summarized as follows.

(a) There is a lack of definitive information on the thermal conductivity of (U,Pu)C, especially at temperatures much above 1400°C (in the case of PuC there is very little data from ambient to 1500°C).

(b) The effects of porosity on thermal conductivity are probably dependent upon temperature, pore size distribution, and pore morphology and are believed related to the combination of variables in (c).

(c) Effects of nickel additions, densification phenomena, carbon, oxygen, and nitrogen concentrations, and concentrations and morphology of second phase higher carbides are interdependent. The quantitative evaluation of these effects will require conductivity measurements on tailored materials which have been well characterized on the basis of

compositional and impurity level control and quantitative microstructural analysis. Baseline studies on the effect of second phase concentration should definitely include resistivity measurements and metallographic (ceramographic) evaluation of, if possible, the sample or part of the sample used for conductivity measurements.

There exists an important adjunct to these conclusions. Deficiencies in the present knowledge of the temperature dependence of the thermal conductivity of uranium-plutonium carbides should be corrected so that reliable benchmark data, both for unirradiated and irradiated material, can be made available to the organizations involved in engineering, design, and evaluation phases of the Division of Reactor Development and Demonstration, Advanced Fuels Irradiation Testing Program. Determination of accurate high temperature values is of special importance to off-normal testing and reactor safety investigations.

**TABLE V**  
**THERMAL CONDUCTIVITY (W/m·deg)**

Equation Coefficients <sup>a</sup>	UC <sub>1.0</sub> 100% TD		PuC 100% TD	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C 100% TD	
	50 < T°C ≤ 700	700 < T°C ≤ 2300	50 < T°C ≤ 2300	50 < T°C ≤ 500	500 < T°C ≤ 2300
a	21.7	20.2	7.45	17.5	12.8
b	-3.04 × 10 <sup>-3</sup>	1.48 × 10 <sup>-3</sup>	-4.04 × 10 <sup>-3</sup>	-5.65 × 10 <sup>-3</sup>	8.71 × 10 <sup>-3</sup>
c	3.61 × 10 <sup>-6</sup>	---	1.20 × 10 <sup>-5</sup>	8.14 × 10 <sup>-6</sup>	-1.88 × 10 <sup>-6</sup>
<b>Temperature (°C)</b>					
25	21.6 (19.7) <sup>b</sup>		---	17.4	
100	21.4 (18.7)		7.2 (3.3) <sup>b</sup>	17.0 (9.8) <sup>b</sup>	
300	21.1 (18.0)		7.3 (5.1)	16.5 (11.6)	
500	21.1 (17.9)		8.4 (6.8)	16.7 (12.6)	
700	21.3 (18.2)		10.5 (8.3)	17.9 (13.4)	
900	21.5 (18.7)		13.5	19.1 (15.0)	
1100	21.8 (19.2)		17.5	20.1	
1300	22.1 (19.9)		22.5	20.9	
1500	22.4 (20.6)		---	21.6	
1700	22.7		---	22.1	
1900	23.0		---	22.5	
2100	23.3		---	22.8	
2300	23.6		---	22.9	

<sup>a</sup>For equation  $\lambda = a + bT + cT^2$ .

<sup>b</sup>Values in parentheses are  $\lambda_e$  values calculated from the theoretical Lorenz number [2.45 × 10<sup>-8</sup> (V/°K)<sup>2</sup>] and suggested values for resistivity.

## REFERENCES

1. A. Sheth and L. Leibowitz, "Thermal Conductivity Values for Advanced Fuels," Interim Report, Argonne National Laboratory report ANL-AFP-3 (November 1974).
2. J. F. Kerrisk, "Uranium-Plutonium Carbide Fuel Properties," Los Alamos Scientific Laboratory internal document (June 5, 1974).
3. W. Fulkerson, T. G. Kollie, S. C. Weaver, J. P. Moore, and R. K. Williams, "Electrical and Thermal Properties of the NaCl Structured Metallic Actinide Compounds," in *Plutonium 1970 and Other Actinides*, W. N. Miner, Ed., Proc. 4th Intern. Conf. Plutonium and Other Actinides, Santa Fe, NM, October 5-9, 1970, Nuc. Met., Met. Soc. AIME 17, (1970), pp. 374-384.
4. J. B. Moser, O. L. Kruger, and J. H. Handwerk, "Electrical and Thermal Properties of Some Actinide Compounds," Proc. Brit. Ceram. Soc. 10, 129-139 (1968).
5. J. L. Bates, "Thermal Conductivity and Electrical Resistivity of Uranium Oxycarbide," Battelle Memorial Institute report BNWL-989 (1969).
6. J. A. Leary and K. W. R. Johnson, "Thermal Conductivity of Uranium-Plutonium Carbide Fuels," in *Plutonium Fuels Technology*, Proc. 1967 Nuclear Met. Symp., Scottsdale, AZ, October 4-6, 1967, Nuc. Met., Met. Soc. AIME 13, (1968), pp. 309-321.
7. P. Costa and R. Lallement, "Resistivity and Thermoelectric Power of the Carbides of Thorium, Uranium, and Plutonium," Phys. Lett. 7, No. 1, 21-22 (1963).
8. J. A. Leary, R. L. Thomas, A. E. Ogard, and G. C. Wonn, "Thermal Conductivity and Electrical Resistivity of UC, (U,Pu)C, and PuC," in *Carbides in Nuclear Energy*, Vol. 1, L. R. Russell, Ed. (Macmillan, London, 1964), pp. 365-372.
9. B. A. Hayes and M. A. DeCrescente, "Thermal Conductivity and Electrical Resistivity of Uranium Monocarbide," Pratt and Whitney Aircraft report PWAC-480 (1965).
10. F. A. Rough and W. Chubb, Eds., "Progress on the Development of Uranium Carbide Type Fuels," Battelle Memorial Institute report BMI-1554 (November 1961).
11. P. Lerner and A. Accary, "Colloque International sur les Propriétés Thermodynamiques, Physiques et Structurales des Dérivés Sémimétalliques," Colloques Internationaux du Centre National de la Recherche Scientifique 157. 515 (1965).
12. A. Euken, "Die Wärmeleitfähigkeit Keramischer Feuersfester Stoffe," Forsch. Gebiete Ingenieurw. Bd. 3, Forschungsheft No. 353, (1932), p. 16.
13. J. C. Maxwell, *A Treatise on Electricity and Magnetism*, 3rd Ed. (Oxford University Press, London, 1892), p. 440.
14. J. L. Bates, "Thermal Conductivity of Uranium Oxycarbide," Proc. 7th Conf. on Thermal Conductivity, Gaithersburg, MD, November 13-16, 1967 (National Bureau of Standards Special Pub. 302, 1968), pp. 477-489.
15. J. T. Sobon, A. D. Miller, and M. A. DeCrescente, "The Thermal Conductivity of Uranium Monocarbide," United Aircraft Corp. Pratt and Whitney Aircraft Division report CNLM-5621 (1964). Also abstract in Bul. Amer. Ceram. Soc. 43, 329 (1964).
16. C. Mustacchi and S. Guiliani, "Development of Methods for the Determination of High Temperature Thermal Diffusivity of UC," European Atomic Energy Community report EUR 337e (1963).
17. S. C. Carniglia, "Single Crystal and Dense Polycrystal Uranium Carbide: Thermal, Mechanical and Chemical Properties," in *Carbides in Nuclear Energy*, Vol. 1, L. R. Russell et al., Eds. (Macmillan, London, 1964), pp. 403-426.
18. A. Accary and R. Caillat, "Development of Ceramic Fuels in France: Nuclear Fuel Elements," H. Hausner, Ed. (Reinhold, NY, 1959), pp. 257-266.
19. R. W. Dayton and C. R. Tipton, "Progress Relating to Civilian Application," Battelle Memorial Institute report BMI-1357 (July 1959), p. 53.
20. L. B. Griffiths, "Effect of Oxygen on the Temperature Coefficient of Resistivity of Uranium Monocarbide," Nature 193, No. 4813, 362-366 (1962).

21. L. N. Grossman, "High Temperature Thermophysical Properties of Uranium Monocarbide," *J. Amer. Ceram. Soc.* 46, No. 6, 264-267 (1963).
22. A. C. Secrest, E. L. Foster, and R. F. Dickerson, "Preparation and Properties of Uranium Monocarbide Castings," Battelle Memorial Institute report BMI-1309 (1959).
23. M. Kamimoto, "Thermal Conductivity of Uranium Carbon Nitride ( $U_{1-x}N_x$ ) from 100° to 1000°K," *J. Phys. Chem. Solids* 35, 393 (1974).
24. R. Pascard, "Preliminary Investigations in the Plutonium Carbon System and the Solid Solutions of the Carbides of Uranium and Plutonium," in *Powder Metallurgy in the Nuclear Age*, Plansee Proc., J. Bemesovsky, Ed. (Metallwerk Plansee, Reutte/Tyrol, 1962), pp. 387-416.
25. J. Crane and E. Gordon, "The Development of Uranium Carbide as a Nuclear Fuel," United Nuclear Corporation report UNC-5080 (1964).
26. L. N. Grossman, "Electrical Conductivity, Thermal Conductivity and Thermal Emission for Fuel Bearing Carbides:  $UC_2$ ,  $UC$ ,  $U_{0.5}Zr_{0.5}C$ , and  $ThC_2$ ," General Electric report GEST-2015 (April 1963).
27. O. L. Kruger and J. B. Moser, "Electrical Properties and Electronic Configuration of the Monocarbide, Monophosphide, and Monosulfide of Plutonium," *J. Chem. Phys.* 46, No. 3, 891-900 (1967).
28. O. L. Kruger and J. B. Moser, "Properties of the Carbides, Phosphides and Sulfides of Plutonium," Annual Progress Report for 1965, Argonne National Laboratory report ANL-7155 (1965), pp. 105-109.
29. J. J. Norreys, M. J. Wheeler, and E. Gillam, "The Preparation and Some Properties of Uranium Sesquicarbide," in *Carbides in Nuclear Energy*, Vol. 1, L. R. Russell, Ed. (Macmillan, London, 1964), pp. 1-6.
30. J. A. Leary and A. E. Ogard, "Quarterly Status Report on Solid Plutonium Fuels Program, June 30, 1963," Los Alamos Scientific Laboratory report LAMS-2949 (July 1963), p. 13.
31. J. A. Leary and A. E. Ogard, "Quarterly Status Report on Solid Plutonium Fuels Program for Period July 1 through September 30, 1963," Los Alamos Scientific Laboratory report LAMS-3023 (October 1963), p. 6.
32. J. A. Leary and R. L. Thomas, "Quarterly Status Report on Solid Plutonium Fuels Program for Period October 1 through December 31, 1963," Los Alamos Scientific Laboratory report LAMS-3054 (March 1964), pp. 6-10.
33. O. L. Kruger, "Constitution and Properties of Plutonium Monocarbide," in *Compounds of Interest in Nuclear Reactor Technology*, J. T. Waber et al., Eds., Intern. Conf. Compounds of Interest in Nuclear Reactor Technology, Univ. of Colorado, Boulder, August 3-5, 1964, *Nuc. Met., Nuc. Met. Soc. AIME, IMD Spec. report* 13 (1964), pp. 387-394.
34. R. N. R. Mulford, F. H. Ellinger, G. S. Hendrix, and E. D. Albrecht, *The Plutonium-Carbon System* (Cleaver-Hume Ltd., London, 1961). Also in *Plutonium 1960*, E. Grissom et al., Eds., Proc. 2nd Intern. Conf. on Plutonium Metallurgy, Grenoble, France, April 19-22, 1960, pp. 301-311.
35. O. L. Kruger, "Phase Studies on Arc-Melted Plutonium-Carbon Alloys Near the Monocarbide Composition," *J. Amer. Ceram. Soc.* 46, 80 (1963).
36. P. Costa, R. Lallement, F. Anselin, and D. Rossigrid, "Magnetic Transitions in Uranium and Plutonium Mononitrides, Monocarbides, and Sesquicarbides," in *Compounds of Interest in Nuclear Reactor Technology*, J. T. Waber et al., Eds., Intern. Conf. Compounds of Interest in Nuclear Reactor Technology, Univ. of Colorado, Boulder, August 3-5, 1964, *Nuc. Met., Met. Soc. AIME, IMD Spec. report* 13 (1964), pp. 83-91.
37. J. C. VanCraeynest, J. C. Weilbacher, and J. C. Salbreux, "Thermal Conductivity of Mixed Uranium and Plutonium Carbides, Nitrides and Carbonitrides," Commissariat à l'Energie Atomique report CEA-CONF-1248. Also Proc. 8th Conf. on Thermal Conductivity, Purdue Univ., 1968, C.Y. Ho and R. E. Taylor, Eds. (Plenum, NY, 1969), pp. 587-601.
38. C. Milet, "Etudes des Composés U-Pu-C-Ti." Commissariat à l'Energie Atomique report CEA-R-3201 (1967).
39. K. W. R. Johnson and J. F. Kerrisk, "Transport Properties, Electrical Resistivity," in "Quarterly Status Report on the Advanced Plutonium Fuels Program, April 1 through June 30, 1970" and "Fourth Annual Report FY 1970," Los

Alamos Scientific Laboratory report LA-4494-MS (August 1970), pp. 45-48.

40. C. Milet, "A Contribution to the Study of U-Ti and U-Pu-Ti Carbides," Commissariat à l'Énergie Atomique report CEA-R-3656 (1968).
41. A. B. G. Washington, "Preferred Values for the Thermal Conductivity of Sintered Ceramic Fuel for Fast Reactor Use," The Reactor Group, United Kingdom Atomic Energy Authority report TRG-R-2236 (September 1973).
42. M. J. Wheeler, E. King, C. Manford, and H. J. Hedger, "Thermal Diffusivity of Uranium and Uranium-Plutonium Carbides," J. Brit. Nucl. Energy Soc. 10, No. 1, 55-64 (1971). Also United Kingdom Atomic Energy Authority, Atomic Energy Research Establishment report AERE-R-6499 (1970).
43. R. Pascard, "Properties of Carbides and Carbonitrides," in *Plutonium Fuels Technology*, Proc. 1967 Nuclear Met. Symp., K. E. Horton et al., Eds., Scottsdale, AZ, October 4-6, 1967, pp. 345-368.
44. M. J. Wheeler, "Thermal Conductivity of Uranium Monocarbide," in *Carbides in Nuclear Energy*, Vol. 1, L. R. Russell et al., Eds. (Macmillan, London, 1964), pp. 358-364.
45. J. B. Moser and O. L. Kruger, "Thermal Diffusivity of Actinide Compounds," Proc. 7th Conf. on Thermal Conductivity, National Bureau of Standards, Spec. Pub. 302 (1968), pp. 461-468.
46. H. E. Schmidt, "Thermal Conductivity," in *Carbide Fuels for Fast Breeder Reactors*, K. Kummerer and F. Thuemmler, Eds., Euratom Fast Reactor Exchange Program, Karlsruhe Nuclear Research Center report EUR FNR-722 (1969), Chap. 7.
47. J. Crane and E. Gordon, "Elevated Temperature Properties of Uranium Carbide," Trans. Amer. Nucl. Soc. 7, No. 1, 100-101 (1964).
48. A. M. Tachis, "In-Reacto Thermal Conductivity of Uranium Carbide," Atomic Energy of Canada, Ltd. report AECL-2521 (1966).
49. M. A. DeCrescente and A. D. Miller, "High Temperature Properties of Uranium Carbide," in *Carbides in Nuclear Energy*, Vol. 1, L. R. Russell et al., Eds. (Macmillan, London, 1964), pp. 342-357.
50. W. Chubb and R. F. Dickerson, "Properties of Uranium Carbides," Ceram. Bull. 41, 564-569 (1962).
51. E. K. Storms, *The Refractory Carbides* (Academic Press, New York and London, 1967).
52. L. S. Levinson, "High Temperature Heat Content of Uranium Carbides," in *Carbides in Nuclear Energy*, Vol. 1, L. R. Russell et al., Eds. (Macmillan, London, 1964), pp. 429-434.
53. J. B. Moser and O. L. Kruger, "Thermal Conductivity and Heat Capacity of the Monocarbide, Monophosphide, and Monosulfide of Uranium," J. Appl. Phys. 38. No. 8, 3215-3222 (1967).
54. W. M. Olson, "Thermodynamic Values for the Plutonium Carbon System," Los Alamos Scientific Laboratory report, to be published.
55. T. G. Godfrey, J. A. Wooley, and J. M. Leitnaker, "Thermodynamic Properties of Uranium Carbides," J. Nucl. Mater. 21, 175-189 (1967).
56. L. E. Russell, "The Structure and Properties of UC and (U,Pu)C Alloys," in Proc. Conf. on New Nuclear Materials Technology Including Non-Metallic Fuel Elements, Prague, July 1-5, 1963, pp. 409-423. Also in United Kingdom Atomic Energy Authority, Atomic Energy Research Establishment report AERE-R-4330 (1963).
57. O. H. Krikorian, "Estimation of High Temperature Heat Capacities of Carbides," Univ. of California Lawrence Radiation Laboratory report UCRL-6785 (1962).
58. L. J. Wittenberg and G. R. Grove, "Reactor Fuels and Materials Development—Plutonium Research: 1964 Annual Report," Monsanto Research Corporation Mound Laboratory report MLM-1244 (1964), pp. 52-55.
59. J. F. Andrew and T. W. Latimer, "Review of Thermal Expansion and Density of Uranium and Plutonium Carbides," Los Alamos Scientific Laboratory report LA-6037-MS (November 1975).
60. J. B. Moser and O. L. Kruger, "Thermal Conductivity and Heat Capacity of Actinide Compounds," Argonne National Laboratory report ANL-7299 (1967), pp. 130-133.
61. J. B. Moser and O. L. Kruger, "Heat Pulse Measurements on Uranium Compounds," J. Nucl. Mater. 17, 153-158 (1965).

62. L. C. Harrington and G. H. Rowe, "Enthalpy and Heat Capacity of Uranium Monocarbide to 1200°C," Pratt and Whitney Aircraft report PWAC-426 (January 1964).
63. J. A. Leary and R. L. Thomas, "Quarterly Status Report on Solid Plutonium Fuels Program, June 30, 1963," Los Alamos Scientific Laboratory report LAMS-2949 (July 1963), p. 13.
64. L. N. Grossman, "Thermal Properties Apparatus for Metallics at High Temperatures," General Electric Co., Atomic Power Equipment Pub. R62APE14 (1962).
65. R. W. Dayton and C. R. Tipton, "Thermal Conductivity of Cast UC," in Battelle Memorial Institute report BMI-1377 (September 1959), p.74.
66. G. A. Meerson, R. B. Kotel'nikov, and S. N. Bashlykov, "Uranium Monocarbide," J. Nucl. Energy, Reactor Science and Tech. 16, 485-488 (1962).
67. T. Kubota, Y. Seki, and S. Takahashi, "Thermal Conductivity of UO<sub>2</sub>, UC, and UC<sub>2</sub>," J. Nucl. Sci. and Tech. 1, No. 3, 93-100 (1964).
68. R. DeConick, W. VanLierde, and A. Gijs, "Thermal Diffusivity and Conductivity of U<sub>2</sub>C<sub>3</sub> up to 2200 K." J. Nucl. Mater. 46. 213-216 (1973).
69. J. D. Farr, W. G. Witteman, P. L. Stone, and E. F. Westrum, "Advances in Thermophysical Properties," in *Advances in Thermophysical Properties at Extreme Temperature Pressure*, 3rd ASME Symp. on Thermophysical Properties, Purdue Univ., Lafayette, IN, March 22-25, 1965, p. 162.
70. Y. Kamimoto, Y. Takahashi, and T. Mukaibo, "Thermal Conductivity of UC<sub>1-x</sub>N<sub>x</sub> from 100 to 1000 K," J. Phys. Chem. Solids 35, 393-399 (1974).
71. A. Sheth and L. Leibowitz, "Equation-of-State for Advanced Fuels," Interim Report, Argonne National Laboratory report ANL-AFP-2 (October 1974).
72. P. G. Pallmer, "Thermal Expansion of Plutonium Carbides, General Electric Hanford Atomic Products Operation report 72245 (1962).
73. O. L. Kruger and H. Savage, "Heat Capacity of Plutonium Carbide from 400° to 1300°K," J. Chem. Phys. 40, No. 11, 1324-1328 (1964).
74. J. F. Kerrisk, "Uranium-Plutonium Carbide Fuel Properties, Los Alamos Scientific Laboratory, internal document (June 5, 1974).
75. K. W. R. Johnson and J. A. Leary, "The Thermal Conductivity of Uranium-Plutonium Carbides," Trans. Amer. Nucl. Soc. 12, 591-592 (1969).
76. S. Bocker, R. Boucher, R. Lorenzelli, and C. Milet, "The U-Pu-C-M Carbides," in *Plutonium 1970 and Other Actinides*, W. N. Miner, Ed., Proc. 4th Intern. Conf. Plutonium and Other Actinides, Santa Fe, NM, October 5-9, 1970, 17, Part I, pp. 113-119.
77. J. M. Horspool, N. Parkinson, J. R. Findlay, R. E. Potter, L. E. Russell, and W. Batey, "Fuel Material for Fast Reactors" in *Fuel and Fuel Elements for Fast Reactors*, Proc. International Atomic Energy Agency Symp., Brussels, July 1973.
78. P. Browning, B. A. Phillips, P. E. Potter, and M. H. Rand, "Phase Equilibria and Vaporization Studies in the U-Pu-C System," United Kingdom Atomic Energy Authority Atomic Energy Research Establishment report AERE-R-6883 (1971).
79. J. Shroder, "A Simple Method for Determining the Thermal Conductivity of Solids," Phillips Tech. Rev. 21, 307 (1959/60).
80. R. Lorenzelli, P. Delarouche, M. Housseau, and P. Petit, "Investigation of Carbide Type Compounds: Phase Diagrams and Properties," in *Plutonium 1970 and Other Actinides*, W. N. Miner, Ed., Proc. 4th Intern. Conf. Plutonium and Other Actinides, Santa Fe, NM, October 5-9, 1970, 17, Part I, p. 818.
81. Director's Office, "Quarterly Status Report on Advanced Reactor Technology (ART) for Period Ending October 31, 1965," Los Alamos Scientific Laboratory report LA-3431-MS (November 1965), pp. 44-49.
82. J. F. Kerrisk and H. D. Lewis, "Transport Properties in Los Alamos Scientific Laboratory Contribution to the Advanced Fuels Program; Quarterly Progress Report for the Period April-June 1975," Los Alamos Scientific Laboratory internal document.
83. C. T. Ewing, B. E. Walker, G. Rand, and R. R. Miller, "Thermal Conductivity of Metals, Liquid Metals Technology," Chem. Engr. Prog., Symposium Series 26. 19-24 (1957).

84. O. Kubaschewski and E. L. Evans, *La Thermochimie en Metallurgie* (Gauthier-Villars, Paris, 1964).
85. K. W. R. Johnson and J. F. Kerrisk, "Thermal Conductivity," in "Quarterly Status Report on the Advanced Plutonium Fuels Program, April-June 1969," Los Alamos Scientific Laboratory report LA-4284-MS (October 1969), pp. 13-14.
86. B. T. Bradbury, B. R. T. Frost, and J. R. Findlay, "The Irradiation Behaviour of Plutonium-Uranium Monocarbide," in *Plutonium 1965*, A. E. Kay et al., Eds., Proc. 3rd Intern. Conf. on Plutonium, London, November 22-26, 1965, pp. 963-975.
87. D. Stahl and A. Strasser, "Properties of Solid Solution Uranium-Plutonium Carbides," in *Carbides in Nuclear Energy*, Vol. 1, L. R. Russell et al., Eds. (Macmillan, London, 1964), pp. 373-391.