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The Thermal Conductivity of Uranium Monocarbide ✓

UC

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(Summary of a paper to be presented at the 66th Annual Meeting of  
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MASTER

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Introduction

Uranium carbide shows promise as a fuel material for reactors operating at relatively high temperatures based on its high melting point, high uranium density and high thermal conductivity. Before refined reactor designs can be made, however, good quantitative data on the thermal conductivity at temperatures in excess of 1000C is required. This paper presents data gathered as part of a continuing study aimed at determining the thermal conductivity of refractory uranium fuels as a function of temperature, density and composition over the temperature range 1000-2200C.

At the inception of this program it was felt that an absolute method capable of achieving high temperatures was necessary and that the difficulties encountered in fabricating the large complex specimens needed were justified. The steady state radial heat flow method and apparatus of Rasor and McClelland (Ref. 4) were therefore chosen.

This paper discusses the experimental equipment and presents results of measurements on three specimens of UC over a temperature range 900 to 1600C. An analysis of the data is made with respect to other physical properties of the material and the measured conductivities are compared with the work of other investigators (Refs. 1, 2 & 3).

Experimental Apparatus

The apparatus has been thoroughly described in the literature (Ref. 4) and only the major modifications will be discussed here. Figure 1 shows an external view of the furnace while Figure 2 is a schematic representation of the equipment including modifications.

Heat is conducted radially across the specimen to a water cooled stainless steel heat sink which acts as a calorimeter, Figure 3. The specimen consists of three one inch high disks, two inches in diameter with a 0.5 inch hole in the center to receive the heat sink.

The development of an accurate and dependable water flow calorimeter was the most difficult phase of the program. The calorimeter in use at the present time is similar to the original design of Rasor and McClelland except that fiberglass reinforced epoxy resin tubes are substituted for glass tubing, as shown in Figure 4. The tubes are knurled in the gauge

section of the calorimeter and are provided with centering lugs to insure concentricity of the entire assembly.

The differential thermocouples and the plastic tube upon which they are mounted are coated with a thermal setting electrical insulating varnish (Dow Corning 997). With water flowing in the calorimeter the resistance from thermocouple to ground is greater than 60,000 ohms for a new calorimeter. Over a period of five to six weeks this value decreases to 25-40,000 ohms at which time the calorimeter is rebuilt. With care in placement of the differential thermocouples, calorimeters can be constructed to read within five percent of the true heat flow and can be calibrated to plus or minus one percent accuracy. The emf generated by the differential couples in the calorimeter is amplified by a 50-2000 microvolt direct current amplifier and recorded by a potentiometer type strip chart recorder.

The specimen rests on carburized tantalum pins which are in turn supported by a tungsten pedestal resting on the vacuum chamber base. The entire furnace system is insulated from the vacuum chamber walls with carbon felt.

The system is evacuated using two (one four inch and one two inch) oil diffusion booster pumps operated in parallel, each backed by a mechanical pump. This arrangement gives fairly rapid pump down and operating pressures in the  $10^{-5}$  torr range for temperatures up to 1500C. Pressures less than  $10^{-3}$  torr are maintained at temperatures to 2200C.

All temperature measurement in the work reported here was by optical pyrometer sighted through optically flat sight ports into black body cavities.

### Procedure

The specimens were hot pressed and then machined to finished dimensions by diamond grinding. The density of the finished material ranged from 85 to 95 percent of theoretical density (13.6 gm/cc). Figures 5 and 6 show a typical specimen prior to test and Figure 7 shows the microstructure of the central portion of a test specimen used to obtain some of the reported data. Typical chemical analysis of the hot pressed blanks are shown in Figure 8.

The experimental runs on UC were started at low temperatures and the temperature raised in 100-200C steps. To insure only radial heat flow through the specimen, the guard heaters on either end of the main heater were adjusted to give a vertical heater temperature profile constant within plus or minus 5C.

The calorimeters were calibrated before and after each test run. The calibration was performed by passing a known current through the stainless steel casing and measuring the voltage drop across the gauge length. A comparison of the power dissipated in the casing with the output of the calorimeter gave a calibration curve. To insure that turbulent flow was maintained in the calorimeter over a wide range of water flow rates, the calibration was carried out at various flow rates and an arbitrary limit of five percent was placed on the

calorimeter correction, i.e., if the calorimeter output indicated less than 95 percent of the true power input, the calorimeter was discarded.

To determine if the specimen had densified or picked up carbon during test, the weight and density were measured. Metallographic and chemical analysis were performed before and after test.

Results and Discussion

The table of Figure 9 and the curves of Figure 10 exhibit the results of this investigation. Figure 12 shows the data plotted along with the results of other investigators.

In order to compare the electronic contribution with the total conductivity, it was necessary to measure the electrical resistivity of UC. This was done using a standard four probe method over the temperature range 25 to 1300C. These data are shown graphically in Figure 11. A more detailed account of these measurements is to be published elsewhere.

The electron contribution to the total thermal conduction was calculated using the Weidemann-Franz Law with a Lorentz constant of  $2.45 \times 10^{-8}$  watt-ohm/C and the measured electrical resistivity. Comparison of the electron conduction and the total conduction shown in Figure 13 indicates that UC exhibits a typical metallic behavior and that at temperatures above 1400C the conduction becomes predominantly electronic. This does not rule out the possibility of some other transport mechanism at higher temperatures, e.g., exciton conduction. It is well known that variations in density of the test specimens have a significant effect on the measured conductivity of the specimens. Thermal conductivity has been related to density using the relationship (Ref. 8)  $K_t = \frac{K_{ex}}{1-P}$ .  $K_t$  is

the total conductivity for a 100 percent dense body,  $K_{ex}$  is the measured conductivity for a body of porosity P. The non-linearity of Figure 14 shows that the measured values for conductivity do not follow this relationship. Assuming that the thermal conductivity of UC is nearly all electronic one would expect the electrical resistivity of UC to also follow the 1-P relationship. Figure 15 shows that this is not the case. In summary, neither the total thermal conductivity nor the electrical resistivity appear to follow the 1-P relationship.

Comparison of the measured data and that of other investigations is somewhat difficult. All other investigations have been on fully dense arc cast material and, as already shown above, the linear relationship between density and conductivity does not apply to UC. Keeping in mind that the CANEL data are for less dense material, Figure 12 shows that there is good agreement between the present data for 91 and 95 percent densities and that of Mustacchi (Ref. 3). (The Mustacchi data has been corrected using the heat capacity data of Harrington (Ref. 5) ). An extrapolation of the data of Dayton (Ref. 1) on 5.3 w/o carbon material would also show good agreement with the CANEL data on the higher density materials.

The effects of excess carbon on the thermal conductivity of UC are also somewhat hard to predict. Carniglia (Ref. 6) has shown that the electrical resistivity increases with increasing carbon content. Again assuming that at high temperatures most of the total conductivity is from electron conduction increasing the carbon content should lower the conductivity, although from the data of Figure 12 this does not appear to be true.

The materials used in the CANEL measurements varied in oxygen content and in most cases contained more oxygen than materials of other investigators. The appearance of  $UC_2$  precipitates in specimens containing less than 4.8 w/o carbon indicates that oxygen and nitrogen are entering into substitutional solution in the UC lattice, however, no evidence was seen of a change in lattice parameters under these conditions. Since impurities in solution generally adversely affect the thermal conductivity, one would expect such an effect here, however the large density variations and a general lack of the data necessary to separate these variables precludes direct experimental confirmation at this time.

The data of other investigators (Refs. 1, 2 & 3) show a positive temperature coefficient while the CANEL data show the opposite. In the data of Dayton (Ref. 1) this difference might be accounted for in the method used. A longitudinal method was used and there is a possibility of errors being introduced at higher temperatures because of improper heat guarding of the specimen. The data of Grossman (Ref. 2) also show a very slight positive coefficient and may be accounted for in that the curve shown in Figure 12 is the best fit from the measured data with a scatter much greater than that of the CANEL measurements. No explanation is available for the difference in the CANEL and Mustacchi data.

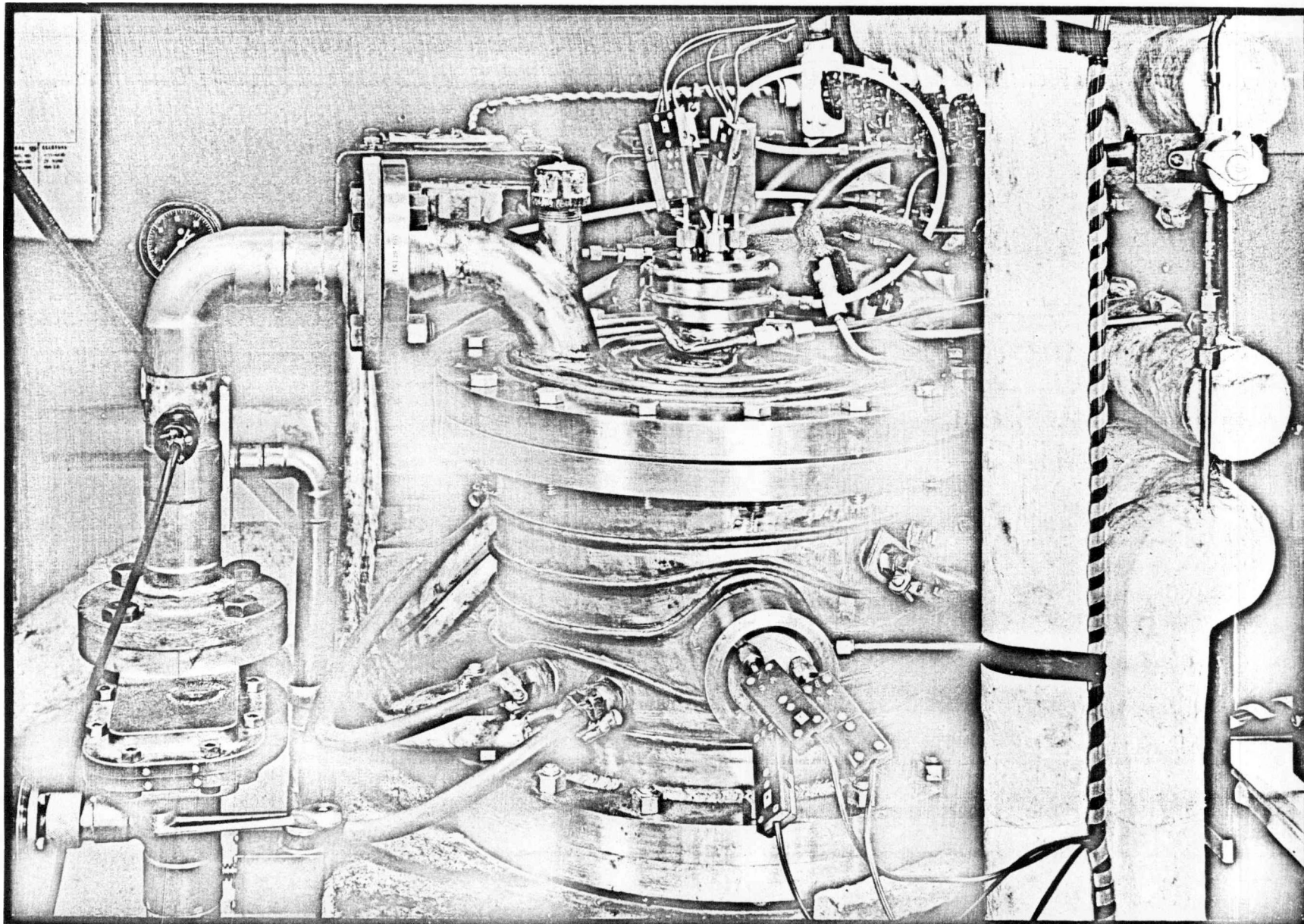
### Conclusions

1. The high temperature thermal conductivity of uranium carbide containing small amounts of excess carbon varies from approximately 0.180 to 0.165 watt/cm-°C over the temperature range 950 to 1600C.
2. The thermal conductivity of UC is typical of metallic materials in that at high temperatures the conductivity is predominantly by electrons.
3. The thermal conductivity decreased with increasing temperature up to 1600C.
4. The measured conductivity is very dependent upon the density and does not follow the 1-P relationship.
5. Although the results of this investigation are in fair agreement with the results of other investigators, a definite correlation cannot be made at this time because most investigators were working with fully dense material or they did not characterize their samples and/or test apparatus well enough to explain variations in their data.

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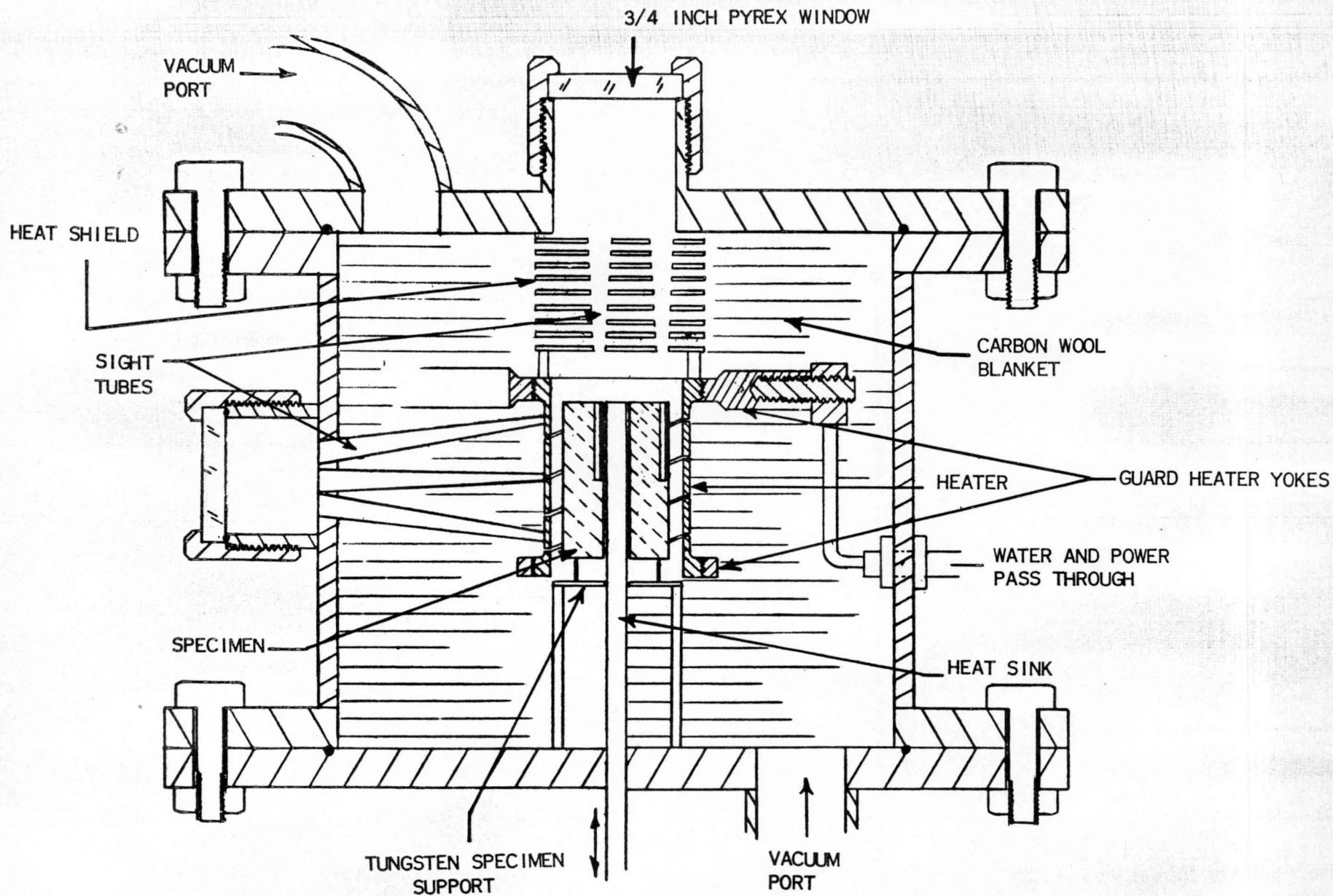
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# THERMAL CONDUCTIVITY FURNACE

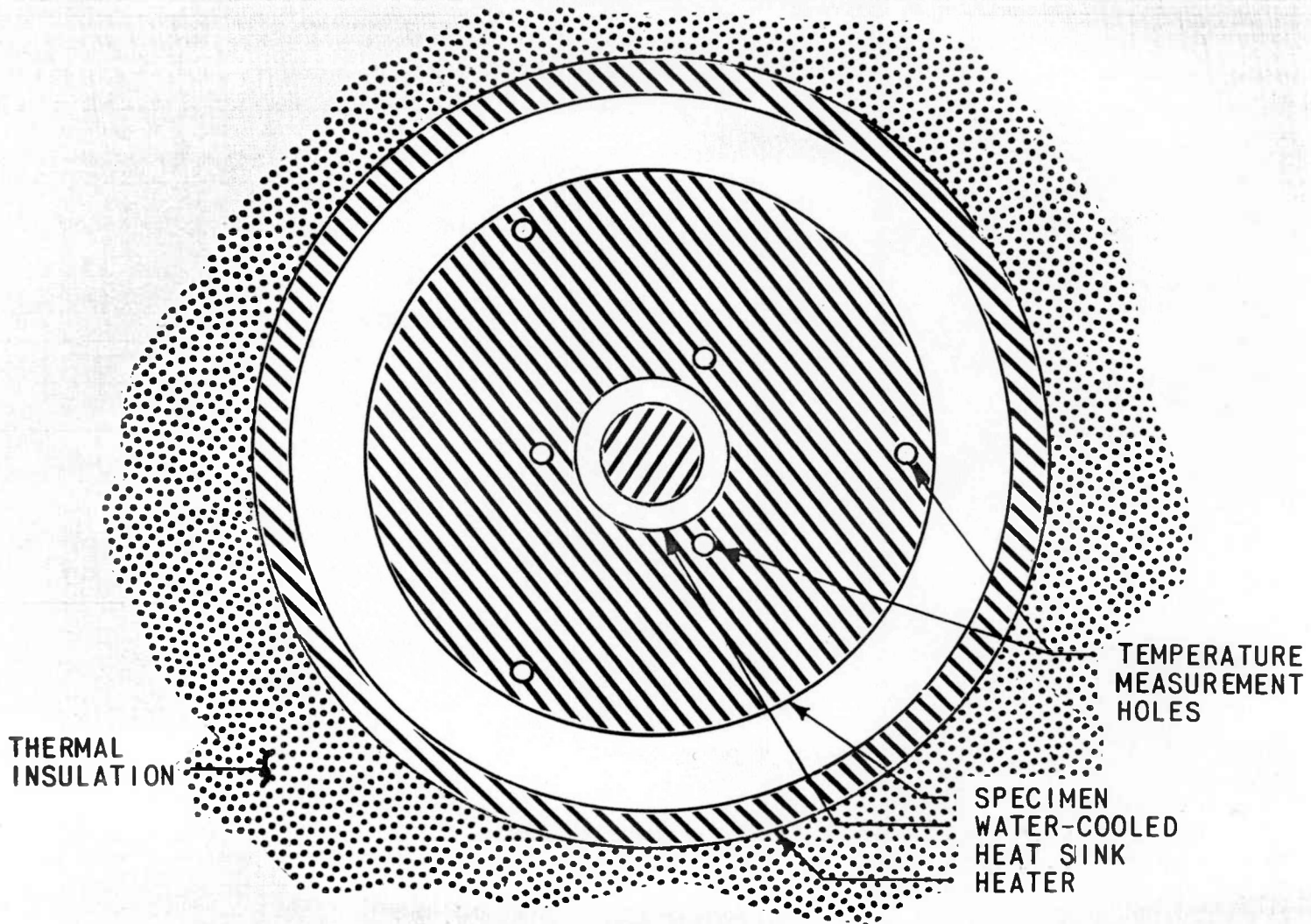


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FIG 1

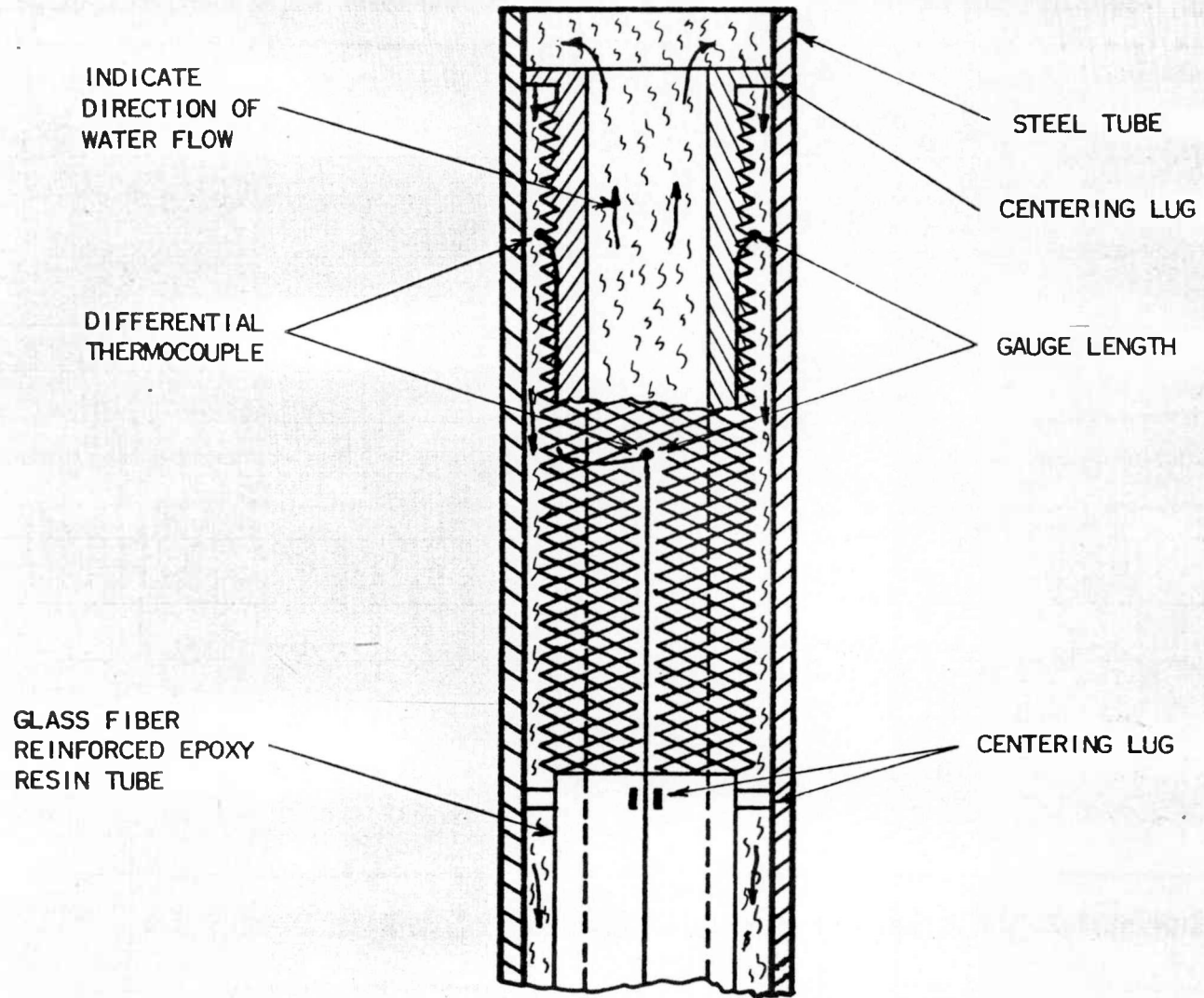
# THERMAL CONDUCTIVITY FURNACE



# HEAT FLOW THROUGH SPECIMEN



# CUTAWAY OF WATER CALORIMETER



THERMAL CONDUCTIVITY SPECIMEN

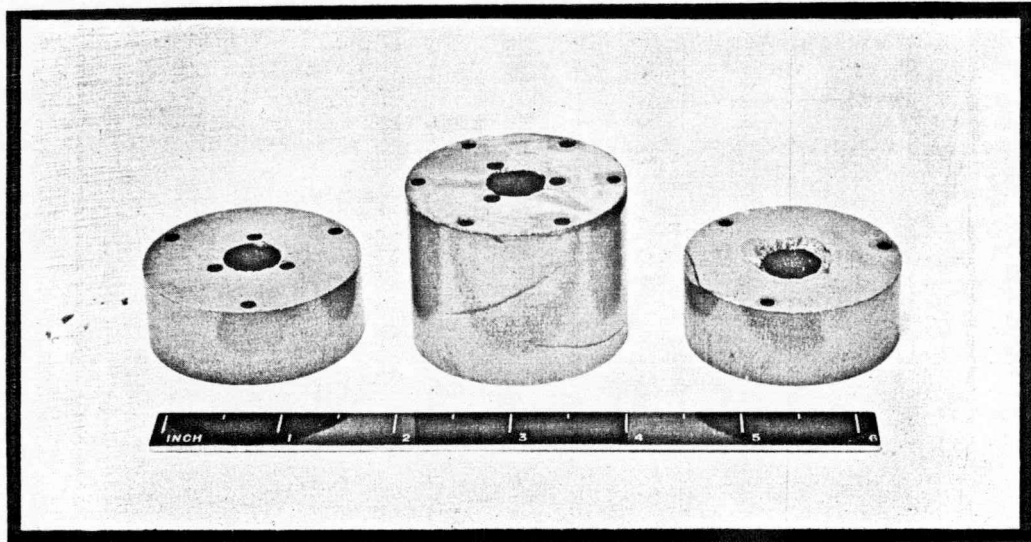
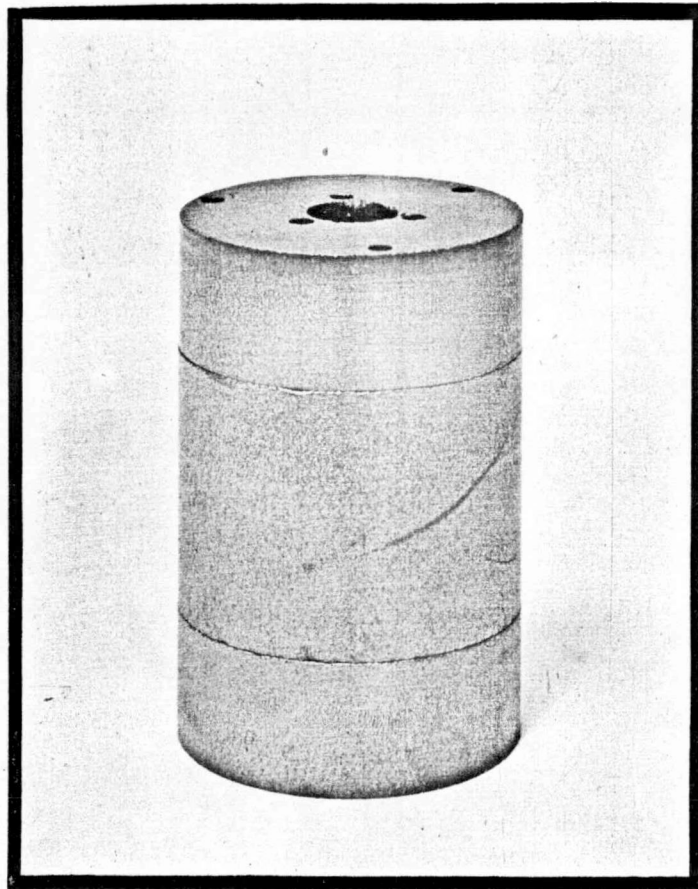


FIG 6

THERMAL CONDUCTIVITY SPECIMEN



MICROSTRUCTURE OF URANIUM CARBIDE



95% T.D.

MAGNIFIED: 200X

## ANALYSES OF HOT PRESSED UC THERMAL CONDUCTIVITY SPECIMENS

SAMPLE NO.	STARTING POWDER CHEMISTRY			DENSITY	AS-PRESSED CHEMISTRY		
	O <sub>2</sub> PPM	N <sub>2</sub> PPM	C W/O		O <sub>2</sub> PPM	N <sub>2</sub> PPM	C W/O
HP-2	4400	450	4.8, 5.1	89.0	3300	310	4.70
HP-15	4400	450	4.8, 5.1	93.7	3600	610	4.81
HP-20	2700	4700	4.85	87.0	2300	6200	4.84
HP-21	2700	4700	4.85	92.1	2500	880	4.72
HP-24	4400	790	4.97	91.0	4200	600	5.01
HP-27	4400	790	4.97	91.5	1250	930	4.85
S-2	1800	185, 230	4.65, 4.75	98.5	4800	160	4.49
S-3	1800	185, 230	4.65, 4.75	97.1	7600	190	4.62
HP-25	900, 1000	175, 210	4.63	--	2800	260	4.76
S-6	900, 1000	175, 210	4.73	98.5	3300	195	4.74

Thermal Conductivity and Electrical Resistivity of UC

Thermal Conductivity

Specimen #UC-2

4.85 w/o carbon  
1250 ppm oxygen  
930 ppm nitrogen

91% Dense

Temp. °C	K (w/cmC)
1025	0.1705
1030	0.1760
1417	0.1590
1440	0.1650
1050	0.1755
880	0.1685
1513	0.1675
1600	0.1640
995	0.1650
995	0.1700

Specimen # UC-3

4.63 w/o carbon  
6800 ppm oxygen  
1350 ppm nitrogen

86.5% Dense

Temp. °C	K (w/cmC)
948	0.1200
1040	0.1130
1050	0.1180
1050	0.1210
1155	0.1200
1160	0.1180
1145	0.1140
920	0.1150
1155	0.1110
945	0.1135
910	0.1150
1225	0.1125
1235	0.1150

Specimen # UC-5

4.75 w/o carbon  
2600 ppm oxygen  
1400 ppm nitrogen

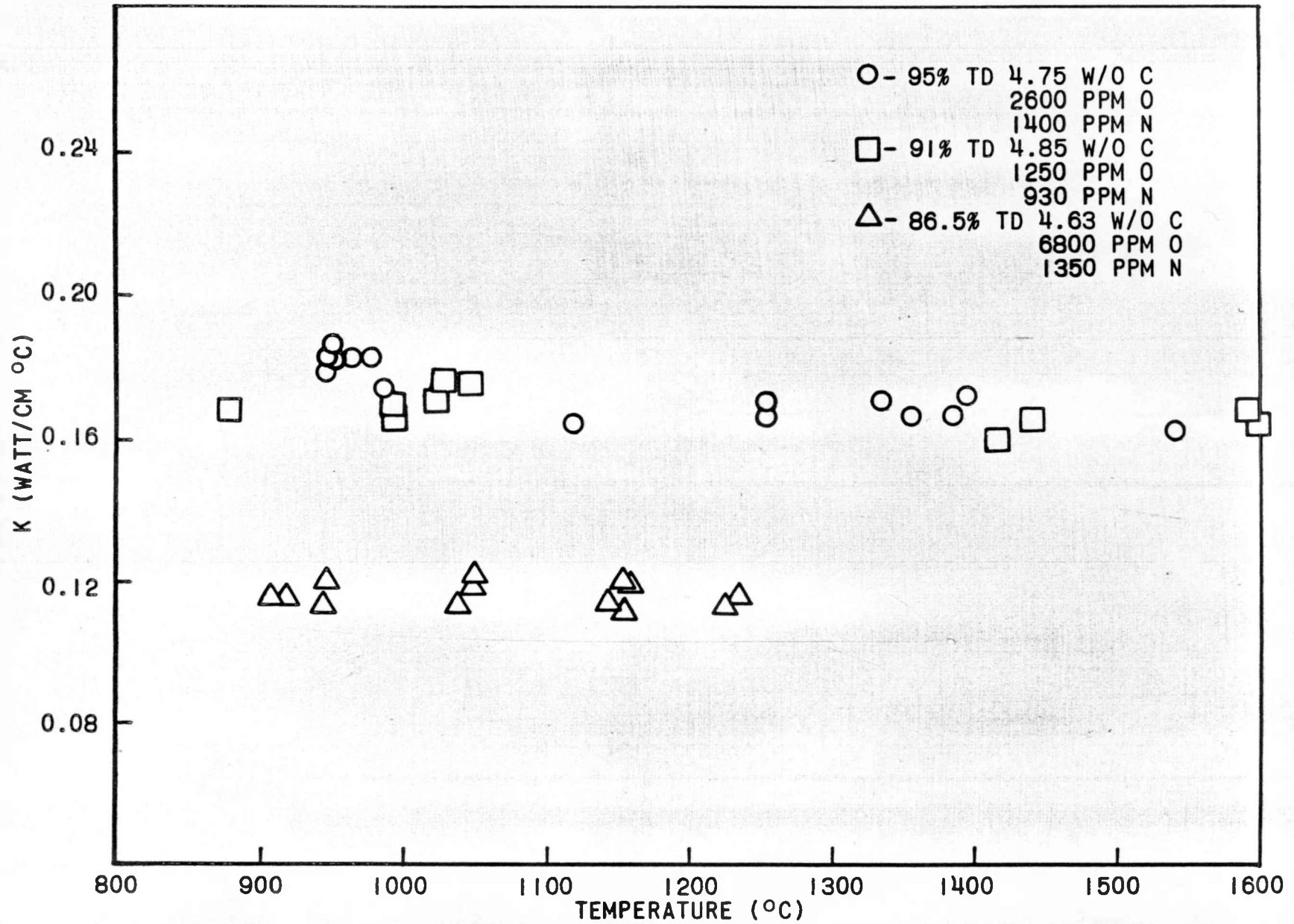
95% Dense

Temp. °C	K (w/cmC)
950	0.1825
987	0.1740
955	0.1820
980	0.1825
967	0.1825
1120	0.1645
1385	0.1655
1540	0.1620
1255	0.1650
953	0.1860
948	0.1780
1335	0.1700
1355	0.1660
1395	0.1715
1255	0.1700

Electrical Resistivity

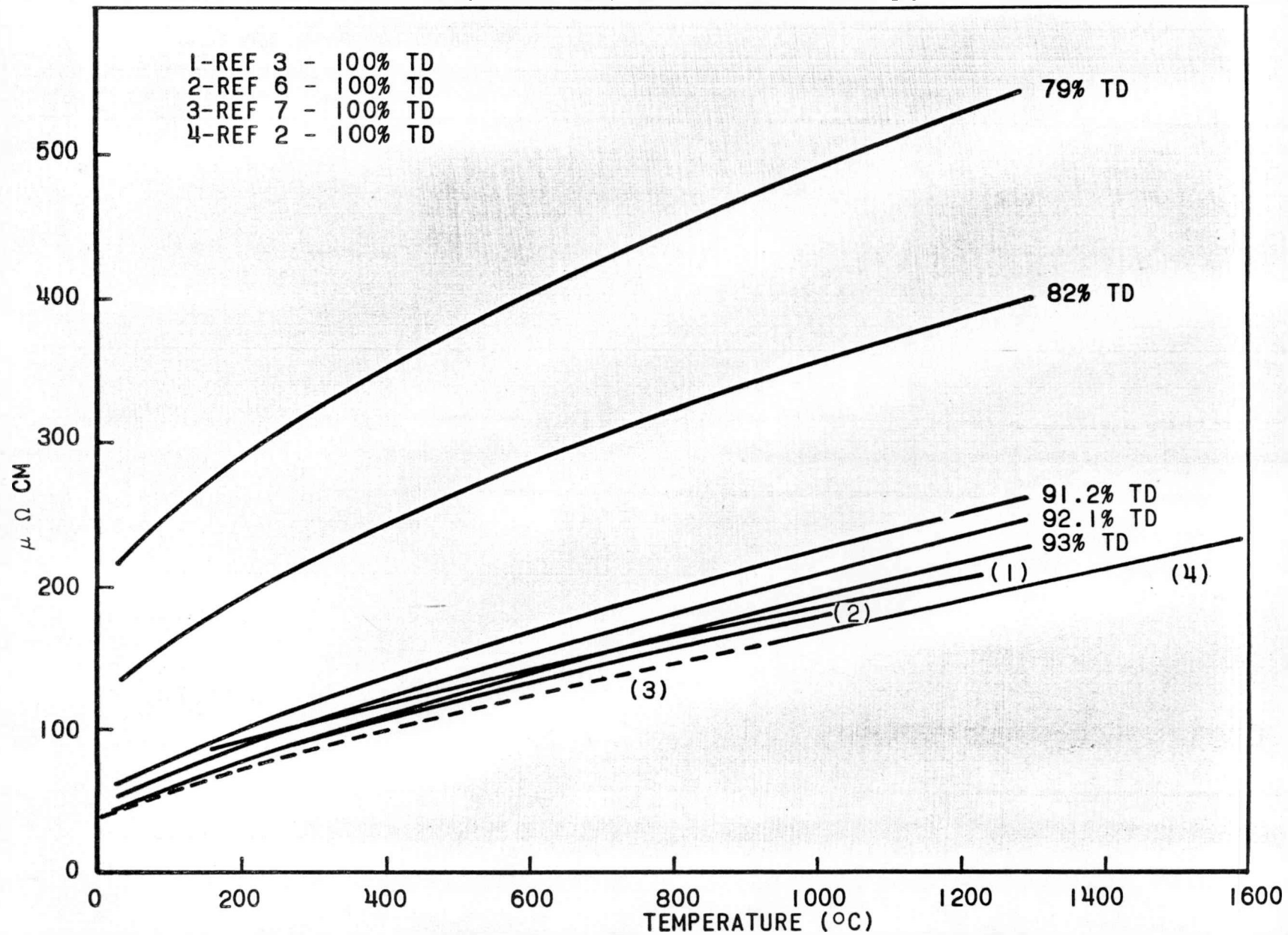
Specimen UCR-1		Specimen UCR-1B		Specimen UCR-2		Specimen UCR-2B		Specimen UCR-3	
4.68 w/o carbon 5600 ppm oxygen 1950 ppm nitrogen		4.67 w/o carbon 1750 ppm oxygen 870 ppm nitrogen		-- w/o carbon -- ppm oxygen -- ppm nitrogen		4.69 w/o carbon 1650 ppm oxygen 5000 ppm nitrogen		4.77 w/o carbon 2100 ppm oxygen 170 ppm nitrogen	
82% Dense		92.1% Dense		79% Dense		91.2% Dense		93% Dense	
Temp. °C	$\mu\Omega$ cm.	Temp. °C	$\mu\Omega$ cm.	Temp. °C	$\mu\Omega$ cm.	Temp. °C	$\mu\Omega$ cm.	Temp. °C	$\mu\Omega$ cm.
21	117.70	24.5	50.92	23	173.52	23	58.55	24.5	42.92
200	174.10	200.5	84.48	206	249.84	200	93.31	200	73.82
427	232.20	422	123.15	430	324.49	427.5	134.00	429.5	110.78
649	277.6	647.5	157.83	650	384.54	645	170.46	660.5	145.26
863	319.3	869	189.73	866	437.88	869	203.85	848	174.41
1089	360.5	1087.5	218.76	1096	488.76	1090	233.23	1094.5	203.50
1308	398.6	1284	245.19	1288	543.01	1293	253.91	1290	225.26

# THERMAL CONDUCTIVITY OF UC



16

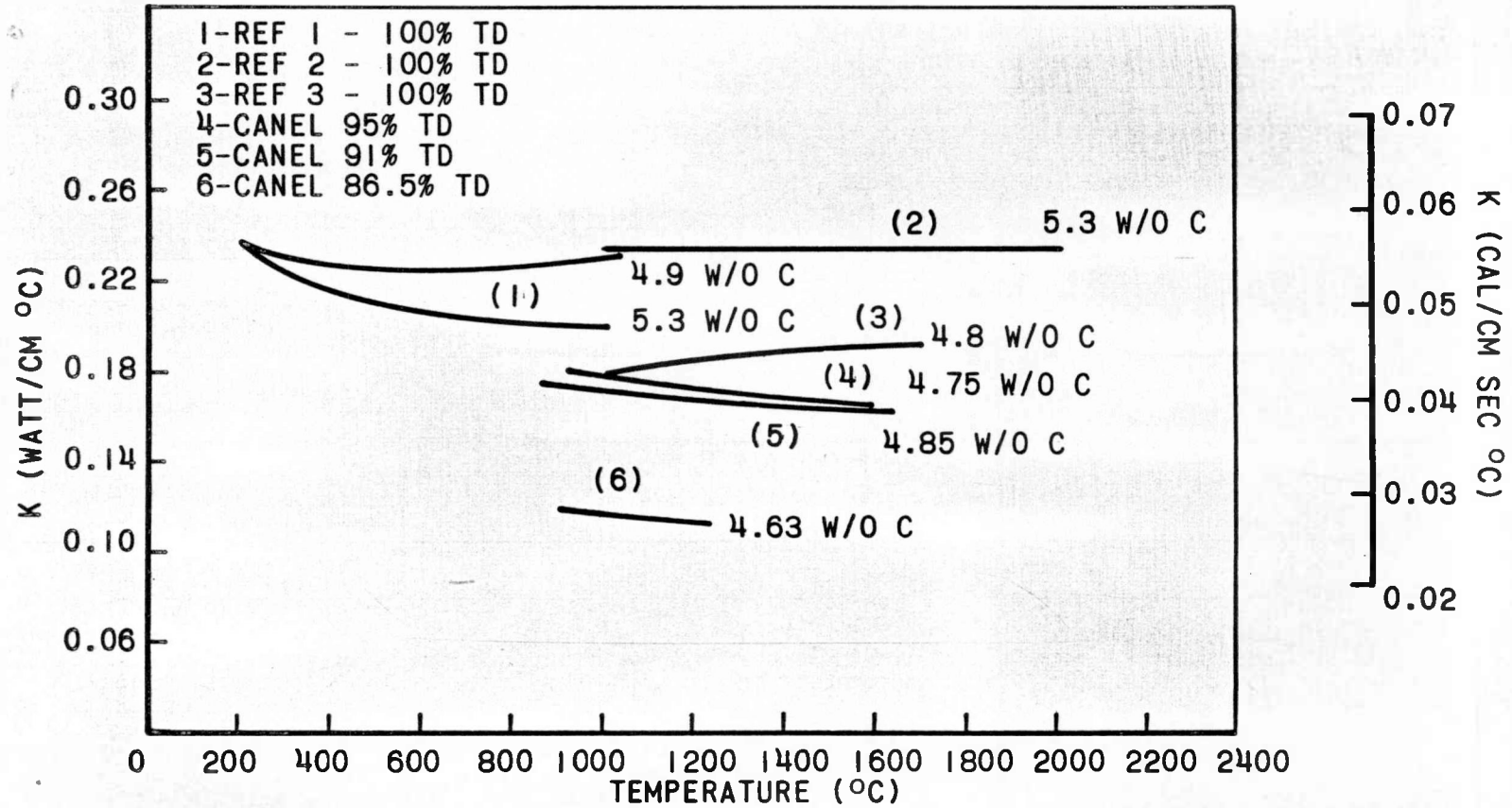
# ELECTRICAL RESISTIVITY OF UC



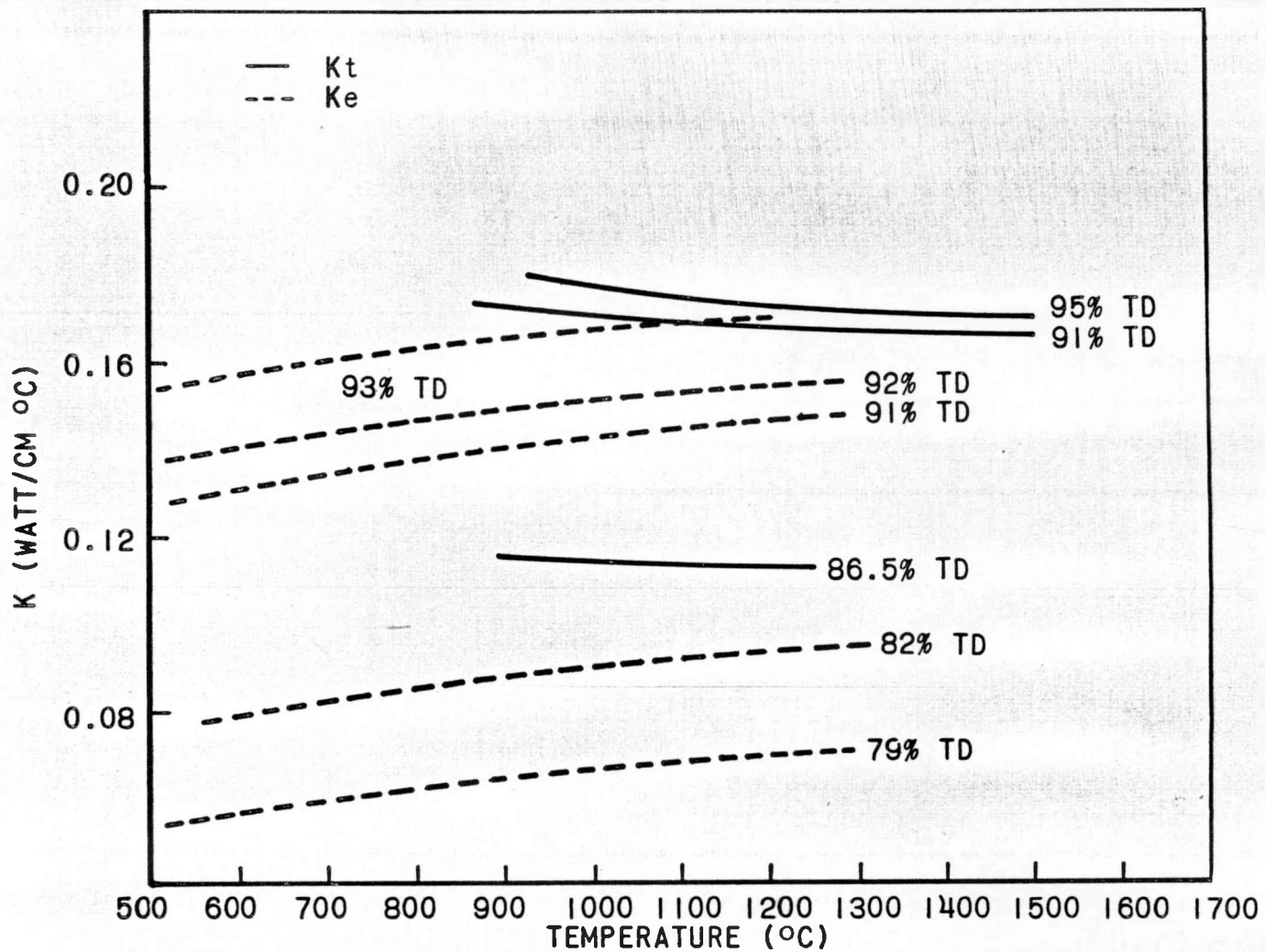
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FIG 11

# THERMAL CONDUCTIVITY OF UC



# ELECTRON CONDUCTIVITY VERSUS TOTAL CONDUCTIVITY



# THERMAL CONDUCTIVITY VERSUS DENSITY

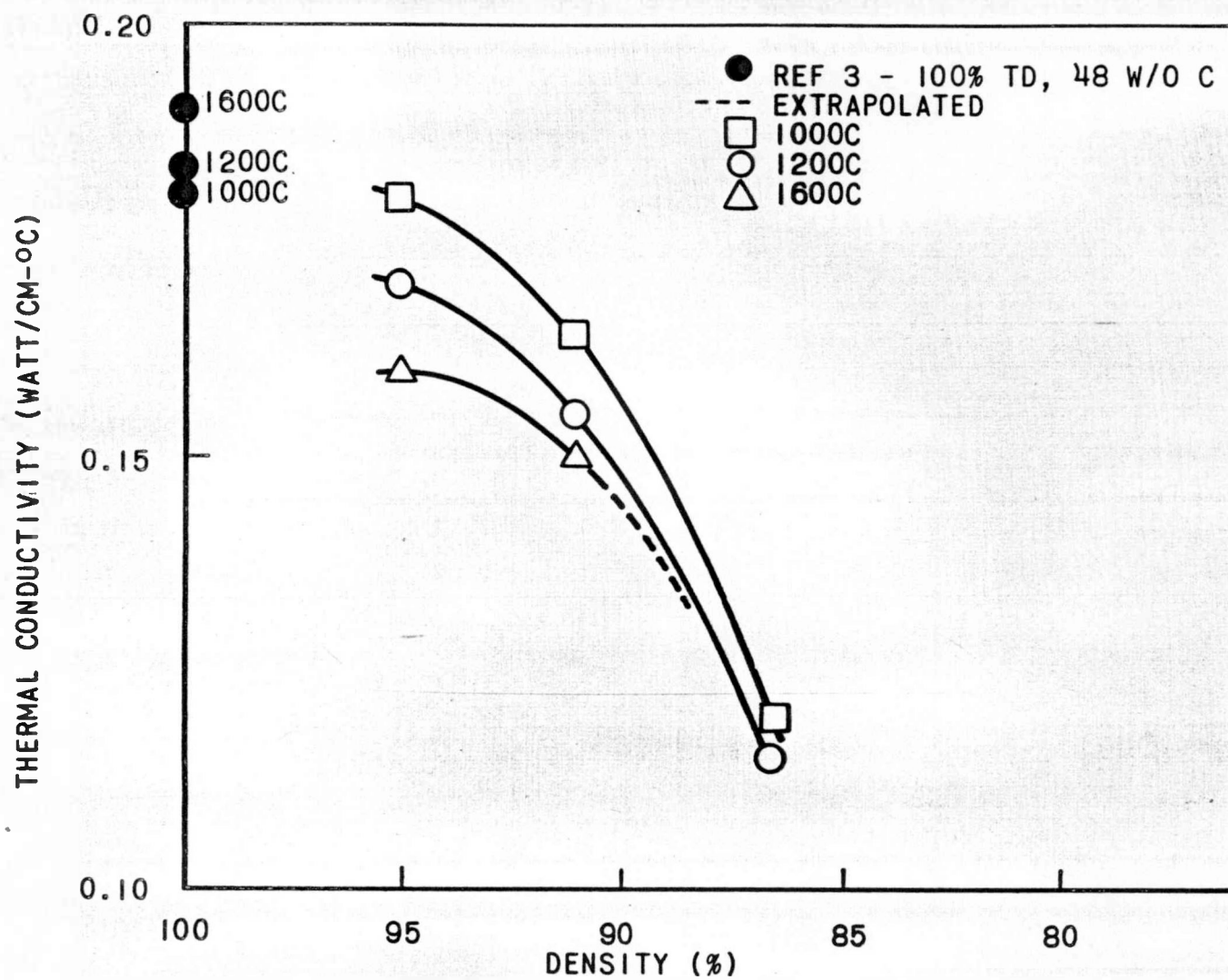


FIG 15

ELECTRICAL RESISTIVITY VERSUS DENSITY

