

Thermal Conductivity Measurements of Uranium Carbide by Electron Bombardment

John P. Greene, Jerry A. Nolen, Maria Petra[†] and Tatiana A. Burtseva[‡]

Physics Division, Argonne National Laboratory, 9700 S. Cass Ave., Argonne, IL 60439, greene@anl.gov

[†]Experimental Facilities Division, Argonne National Laboratory, Argonne, IL

[‡]Energy Technology Division, Argonne National Laboratory, Argonne, IL

Abstract

For the next generation of nuclear physics accelerator facilities, fission targets employing the compounds of uranium are being proposed to produce ion species far from stability. With the 100 kW beams proposed for the RIA production targets, heat dissipation becomes a serious issue. In our two-step target design, neutrons are first generated which then induce fission in a surrounding assembly of uranium carbide. Knowledge of the physical, chemical and, most importantly, the thermal properties of the uranium target material are crucial to the engineering designs under consideration. The interplay of density, grain size, thermal conductivity, and diffusion properties of the uranium carbide must be well understood before a successful target can be built.

We present here recent investigations of uranium carbide samples generated by thermochemical reduction starting from the oxide. Thin sample pellets have been produced for measurements of thermal conductivity using the method of electron bombardment. The experimental apparatus consists of a 10 kV vertical electron beam source, which is used to heat the samples while the thermal radiation is observed using a two-color optical pyrometer. Initial results are given and compared with bulk properties as well as to similar measurements made on pellets of uranium carbide powder. Slight variations of thermal conductivity with density have been observed for the reduced oxide samples. Experimental investigations of the physical and thermal properties for uranium carbide using various binder materials are also being undertaken.

I. INTRODUCTION

The target materials used for ISOL-type radioactive beam facilities are often porous forms of refractory compounds, such as CaO and UC_x (where $x \leq 2$). At high primary beam power, the temperature distribution within such targets depends on the thermal conductivity of these forms of the materials. Very little data exist on the thermal conductivity of such materials at

the temperatures of interest, typically 1500-2000 °C, or even higher. For the next generation of nuclear physics accelerator facilities, targets employing the compounds of uranium are being proposed to produce ion species far from stability. With the 100 kW beams proposed for the RIA production targets, heat dissipation becomes a serious issue. In our two-step target design [1], neutrons are first generated which then induce fission in a surrounding assembly of uranium carbide. Knowledge of the physical, chemical and, most importantly, the thermal properties of the target material are crucial to the engineering designs under consideration. The interplay of density, grain size, thermal conductivity, and diffusion properties of the uranium carbide must be well understood before a successful target can be built.

We present here recent investigations of uranium carbide samples involving the thermochemical reduction starting from the oxide [2], as well as samples made from UC₂ with various binding agents. Thin sample pellets have been produced for measurements of thermal conductivity using the method of electron bombardment. The experimental apparatus consists of a 10 kV vertical electron beam source, which is used to heat the samples while the thermal radiation is observed using a two-color optical pyrometer. Initial measurements of the physical and thermal properties of pellets prepared from uranium carbide using various binder materials are now being undertaken and the results compared with the earlier work by De Coninck on uranium dicarbide at high temperatures [3].

The thermal conductivity of an UC_x pressed disk was first investigated. The sample dimensions, its mass and density, are given in Table I. The thermal conductivity measurement is based on the proportionality of the heat flow and temperature gradient (Fourier's law). For a thermally insulated slab with no internal heat sources the steady-state rate of heat flow (q) into one side of the slab equals the one out of the other side that is:

$$q/A = -k \Delta T / \Delta x \quad (\text{Eq. 1})$$

where A = cross-section of the slab,
 ΔT = temperature difference between the faces
of the slab,
 Δx = thickness of the slab,
 k = thermal conductivity,

as schematically represented in Fig. 1.

II. EXPERIMENTAL METHOD

The sample was heated on one side by a vertical electron beam gun or “mortar” source manufactured by Veeco, GMBH. It has been installed within our NRC 3117 evaporator, which is mainly employed for the high temperature reduction/distillation of rare-earth isotopes to produce metal targets [4]. The product of the supplied voltage times the current gives the power delivered by the electron beam. The sample was mounted on top of a 15 cm. glass tube where the spot size is approximately the size of the target as shown in Fig. 2. The sample was supported by a molybdenum mesh (95% transmission) that was mounted central to the beam axis using a tantalum metal ring. This whole assembly was then positioned on top of the glass tube, as shown in Fig. 3. The electron energy was 12.27 keV, as measured using a high-voltage probe.

After achieving thermal equilibrium, the temperature of the two faces of the sample (Fig. 1) was measured with the aid of a two-color pyrometer [5]. The Model R-35C15 pyrometer (IRCON, Inc., 7300 N. Natchez Ave., Niles, IL 60714 USA) was first set up to measure the temperature of the bottom face of the sample then re-positioned above to view the top face. For a complete measurement, the experiment consisted of the sample being irradiated twice.

II.A Thermal Conductivity Analysis

The thermal conductivity of the UC_x sample was calculated using the following equations:

$$\text{Power/sample area (Watts/cm}^2\text{)} = [\text{Current (mA)} \times 12.27 \text{ (kV)}] / 0.712 \text{ (cm}^2\text{)} \quad (\text{Eq. 2})$$

$$\text{Emissivity } (\epsilon) = [\text{Power/sample area}] / \sigma [(T_{\text{cold}}^4 - T_o^4) + (T_{\text{hot}}^4 - T_o^4) + (2\Delta x/D)(T_{\text{avg}}^4 - T_o^4)] \quad (\text{Eq. 3})$$

$$P_{\text{cold}} = [\epsilon \sigma (T_{\text{cold}}^4 - T_o^4)] = k \Delta T / \Delta x \quad (\text{Eq. 4})$$

where k - thermal conductivity (W/cm K);
 Δx - thickness of the slab;
 D - sample diameter;

T_{hot} - temperature of the side of the sample that is being irradiated;
 T_{cold} - temperature of the side of the sample not heated by the electron beam;
 P_{cold} - power per unit area out of the cold face of the sample;
 T_o - room temperature = 20 °C;
 $T_{\text{avg}} = (T_{\text{hot}} + T_{\text{cold}}) / 2$;
 ΔT - temperature difference between the cold and hot faces of the sample.

After the determination of T_{cold} and T_{hot} , the emissivity and the thermal conductivity of the sample were calculated using Equations 3 and 4 respectively. The thermal conductivity was evaluated at T_{avg} and plotted over the range of temperatures measured.

III. SAMPLE PREPARATION

III.A Reduction from the Oxide

The preparation of the uranium carbide pellets starts with the reduction of the oxide with an excess of graphite [6]. The equation is given as follows:



Depleted uranium oxide and powdered graphite were mixed in a proportion of 79% to 21% and reduced by heating under vacuum, the details having been previously presented in Ref.2. As given in Table I, the densities for these samples are quite low, approximately one quarter of that for solid UC_2 .

III.B Sample Stoichiometry

The phase of interest normally associated with an excess of carbon is designated “ UC_2 ” although this phase is almost always found to be sub-stoichiometric. From the literature [7], the composition lies in the range $UC_{1.75-1.90}$ with the commonest form being $UC_{1.8}$. Hence, the stoichiometry of the pellets prepared in this fashion is designated UC_x , where x can approach 2. From Table I, it can be seen that in some instances there occurred incomplete reduction of the oxide in the compound. From the chemical equation, complete reduction corresponds to approximately 16 % weight loss. A microscopic analysis in Fig. 4 shows areas where the uranium oxide was not converted to the carbide. For each sample, a detailed chemical analysis would have to be performed to determine the actual composition.

III.C Uranium Carbide Samples

In addition to the unknown composition of the UC_x , problems arising with the samples due to brittleness prompted the use of uranium carbide powder directly, as there was some material available. Initial attempts with pressing pellets from the UC_2 powder alone were not successful, resulting in the need for binding agents, the simplest of which being the addition of carbon powder. Other binders are presently being explored.

The sample disks for the thermal conductivity measurement were prepared in a laboratory hood by first weighing 0.32 g of UC_2 powder (-60 mesh) (CERAC, Inc., P.O. Box 1178, Milwaukee, WI 53201 USA). As an aid in holding the pellet together, 40 mg of carbon is added (ratio of 8:1) in the form of high-purity synthetic graphite powder (-200 mesh) (Alfa Aesar, 30 Bond St., Ward Hill, MA 01835 USA). The material is then transferred to a 10 ml glass beaker where two drops of albumin (separated egg-white) are added as a binder and mixed thoroughly. The use of egg white as a binder in ISOL type targets is a common practice employed at TRIUMF [8]. The approximate weight of the albumin is 15 mg. Next, the mixture is poured into the 10 mm compaction die (International Crystal Laboratories, 11 Erie St., Garfield, NJ 07026 USA) and pressed to 5 tons (10,000 psi) using a Model B laboratory press (Carver, Inc., 1569 Morris St., Wabash, IN 46992 USA). As can be seen in Table II, the sample disks produced by this method are on the order of 1 mm in thickness. The densities of 5-6 g/cc are still about only 50% of those obtained for pellets produced as reactor fuel elements [9].

Since uranium carbide is no longer commercially available, we have begun in-house preparation of this material by the method of arc melting of uranium metal in the presence of an excess of solid carbon [10]. Sample preparation using various carbon and/or graphite materials is being investigated. As grain size will prove to play an important role in ultimate densities and thermal conductivities achieved, this prepared material can be further characterized using fine-mesh sieves. The desired release of the fission products produced under neutron irradiation also needs to be explored as a function of grain size. Research into this aspect of our UC_2 samples as a step toward real RIA targets is just now beginning.

III.D Sintering

Since it is known that the physical properties of these pressed powder samples vary depending on their thermal history, investigations are underway to explore and quantify these effects. Changes in density, thermal conductivity and the microscopic structure of the sample

pellets have been determined for three sintering temperatures, 1100, 1500 and 2000° C. The sample disks were placed in a tantalum crucible and heated in a Type CH-10 crucible heater (R.D. Mathis Co., P.O. Box 92916, Long Beach, CA 90809 USA) within a vacuum evaporator under a nominal pressure of a few 10^{-6} Torr. Currents of 300-400 A were applied as in a standard thermal evaporation with the temperature monitored using an optical pyrometer (Leeds & Northrup Co., 3 Fountain Ave. Ellwood City, PA 16117 USA). For the final sintering temperatures of 1100° C and 1500° C, the heat was slowly brought up over a 30-minute duration. The oven was then held at the sintering temperature for 30 minutes and then slowly ramped down again over a 30-minute time span. For the 2000° C temperature, the samples were first sintered to 1500° C as above. To reach the higher sintering temperature, the pellets were then heated directly with the electron beam set-up used for the thermal conductivity measurements. Again, the beam power was slowly increased, held at 2000° C for a few minutes, and decreased slowly, the whole process taking approximately 30-40 minutes. Although an increase in density with sintering temperature has previously been reported [7], no change was observed for the sintered samples vs. the unsintered samples.

IV. INITIAL RESULTS AND DISCUSSION

The thermal conductivity obtained for “reduced” UC_x samples was disappointingly low (typically 1-2 W/m-K), a value that renders this material unacceptable for high power target designs.

In Fig. 5 we plot, in the upper graph, the thermal conductivity, k in W/cm-K over the temperature range of 1700° to 2400° K for three samples. In the lower graph is plotted initial data for three UC_2 samples. At twice the density, these samples showed improved thermal conductivities of 2-15 W/m-K, making this material quite attractive for use in the RIA target. The calculated emissivity, ϵ , was determined to be 0.7-0.8 which is higher than the value of 0.42 given by Grossmann [11]. In addition, the thermal conductivity is plotted over the same temperature range for a “ UC_2 Nukem melt” sample (Ref. 3) with 99.5% theoretical density. As can be seen, there is still some improvement in thermal conductivity to be attained.

In Fig. 6 we display a plot for UC_2 Sample II, which shows the influence of sintering. The measurement was performed twice, first for an unsintered sample and then again after sintering. Although overall, the thermal conductivity is rather low ($k \sim 1.5$ W/m-K), there was a marked improvement after sintering. In the lower plot the thermal conductivity is given over the temperature range

1800-2400° K for one of the recent samples using carbon and albumin as a binder. The thermal conductivity measured was quite good from 4 to about 10 W/m-K and the emissivity ranged from 0.4-0.5. More samples have been prepared and further measurements are underway.

V. CONCLUSION

In conclusion, a method for measuring the thermal conductivity of small samples of porous materials has been demonstrated. The method will be refined and checked for accuracy and overall degree of reproducibility. The results for porous UC_x samples prepared by the ISOLDE prescription give densities on the order of 3 g/cc and thermal conductivities of 1-2 W/m-K. Densities and thermal conductivities for UC₂ samples show improved properties (k = 2-12 W/m-K) and have great promise for use in the high-power applications needed for RIA targets.

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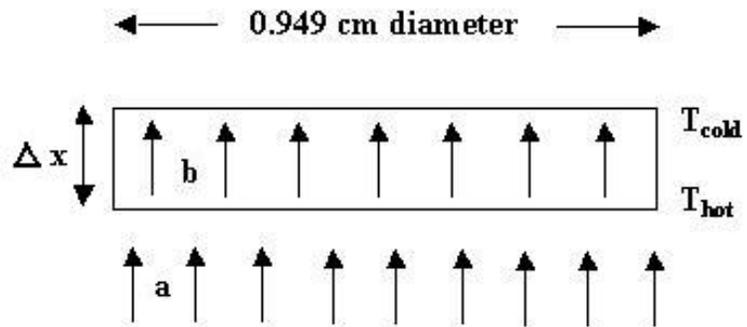
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Table I: UC_x sample pellets reduced from the oxide.

Sample	Orig. Weight (g)	Final Weight (g)	% Change	Diameter (mm)	Thickness (mm)	Density (g/cc)
2	0.324	0.2858	11.79			
3	0.3169	0.2601	17.92	9.7	1.56	2.26
4	0.3185	0.2733	14.19	9.62	1.32	2.85
5	0.3248	0.2779	14.44	9.67	1.39	2.72
6	0.3207	0.2716	15.31	9.68	1.3	2.84
7	0.3137	<i>broke</i>	-	-	-	-
8	0.3258	0.2759	15.32	9.61	1.4	2.72
9	0.3104	0.2562	17.46	9.33	1.36	2.76
10	0.3231	0.277	14.27	9.67	1.29	2.93
11	0.3205	0.2794	12.82	9.68	1.25	3.04
12	0.3216	0.278	13.56	9.65	1.27	2.99
A	0.3797	0.3412	10.14	9.71	1.53	3.01
B	0.3772	0.3476	7.85	9.66	1.51	3.14
C	0.3791	0.3449	9.02	9.76	1.54	2.99
D	0.3675	0.3307	10.01	9.68	1.53	2.94
E	0.3845	0.3523	8.37	9.68	1.5	3.19
F	0.3845	0.3526	8.30	9.69	1.62	2.95
G	0.3615	<i>broke</i>	-	-	-	-
H	0.3919	0.3612	7.83	9.68	1.56	3.15
I	0.3776	0.3521	6.75	9.68	1.45	3.3
J	0.3149	0.2993	4.95	9.64	1.24	3.3

Table II: Sample Pellets of Uranium Carbide (10 mm dia.)

Sample	Sintering Temp. (°C)	Weight (g)	Thickness (mm)	Diameter (mm)	Density (g/cc)
1		0.6392	1.59	9.68	5.47
2		0.6369	1.60	9.79	5.29
3		0.6336	1.61	9.81	5.21
I (broken)					
II		0.3662	1.02	9.59	4.97
III		0.3259	0.97	9.46	4.78
IV		0.3582	0.95	9.74	5.06
UC0130A		0.277	0.67	9.51	5.82
UC0130B		0.301	0.88	9.42	4.91
UC0130C		0.294	0.71	9.62	5.70
UC/C0130A		0.402	1.05	9.58	5.31
UC/C0130B		0.322	0.85	9.55	5.29
UC/C0130C		0.306	0.95	9.56	4.49
UC/0128A		0.531	0.78	12.90	5.21
UC/0128B		0.509	0.80	12.98	4.81
UC/C/ACB02/21 A	1000	0.4394	1.04	10.10	5.28
B	1500	0.5070	1.26	10.18	4.95
C	2000	0.5136	1.21	10.15	5.25
UC/C/ACB05/07 A	2000	0.6555	1.49	10.20	5.39
B	2000	0.6745	1.43	10.19	5.79



- a) Electron beam heats lower side of the sample
- b) Heat flux through the sample = heat radiated from upper side

Fig. 1: Schematic representation of the thermal analysis process.



Fig. 2: Photograph of the sample above the mortar source.

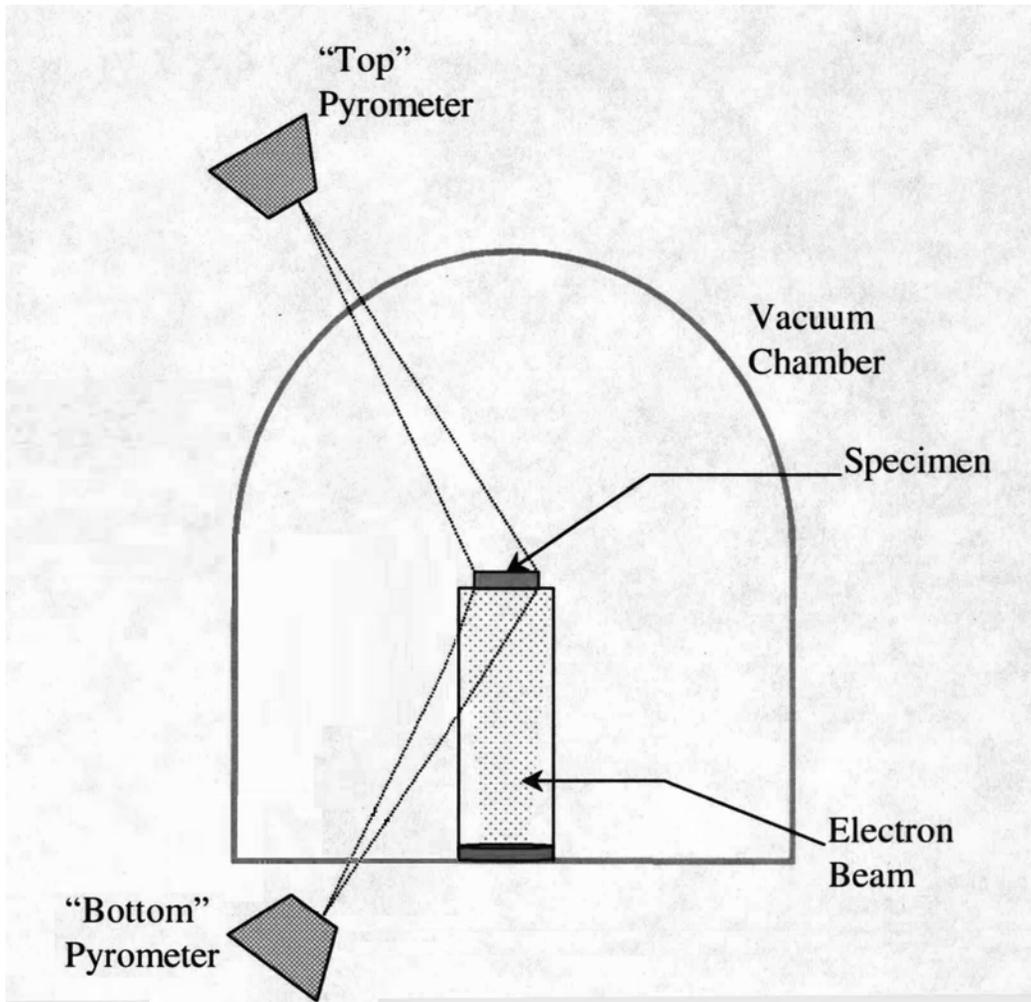


Fig. 3: Schematic of the experimental setup.

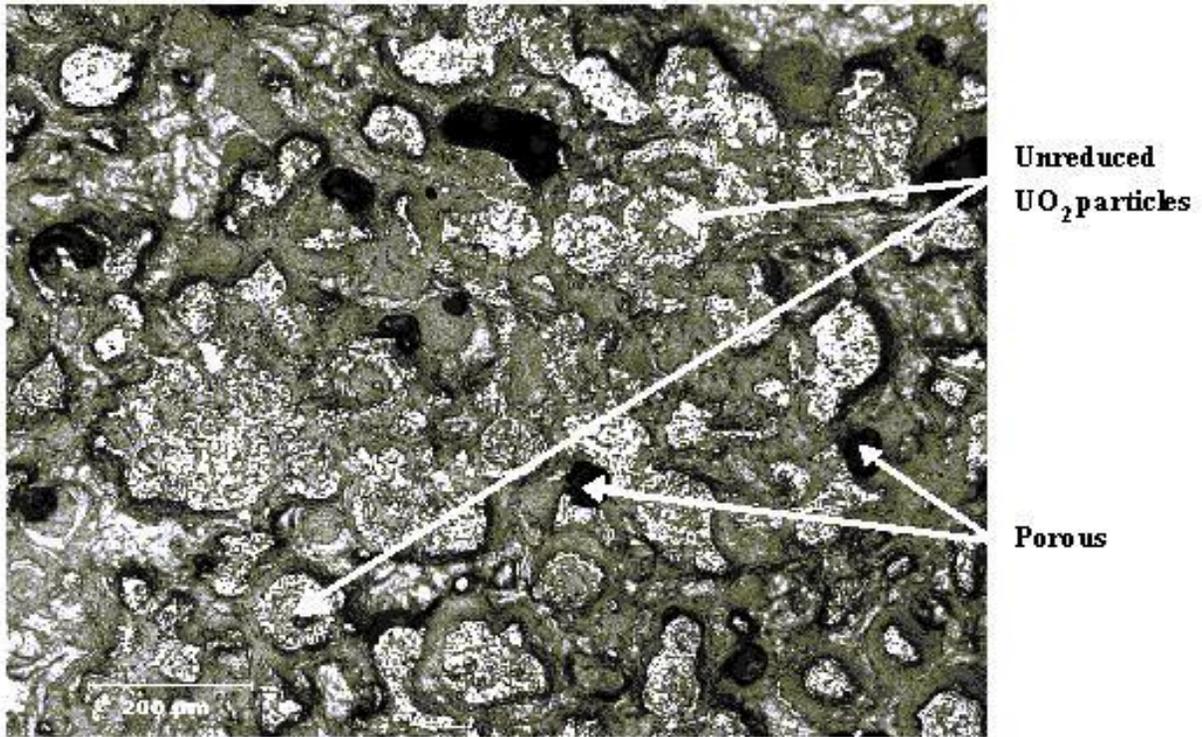


Fig. 4: Microstructure of uranium carbide prepared from UO_2 by reduction, magnification 100x.
The indicating arrows show unreduced UO_2 particles.

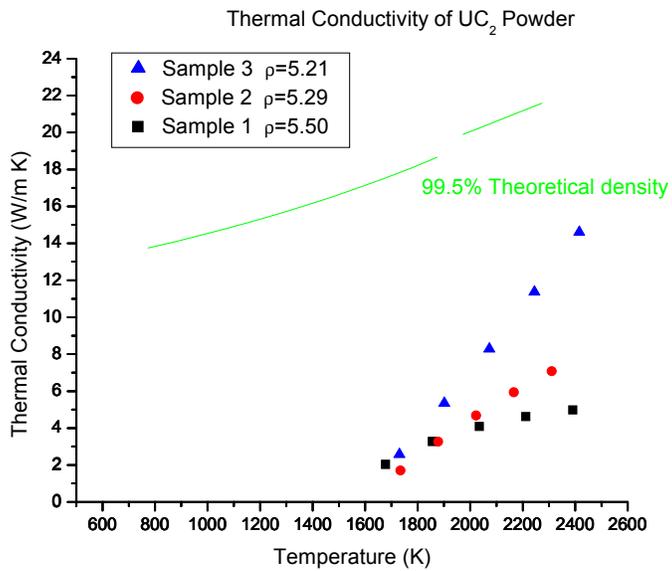
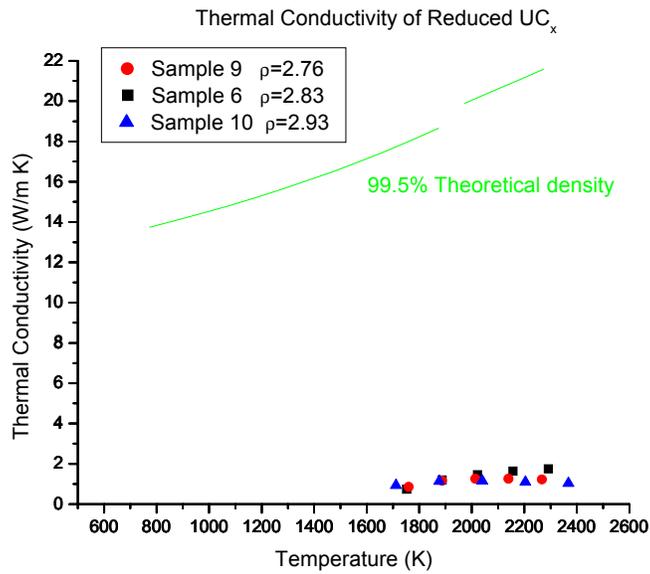


Fig. 5: Initial thermal conductivities obtained for reduced UC_x (top) and UC₂/C (bottom) samples as a function of temperature. Also shown is the thermal conductivity for a “UC₂ Nukem melt” sample (Ref. 3) of 99.5% theoretical density.

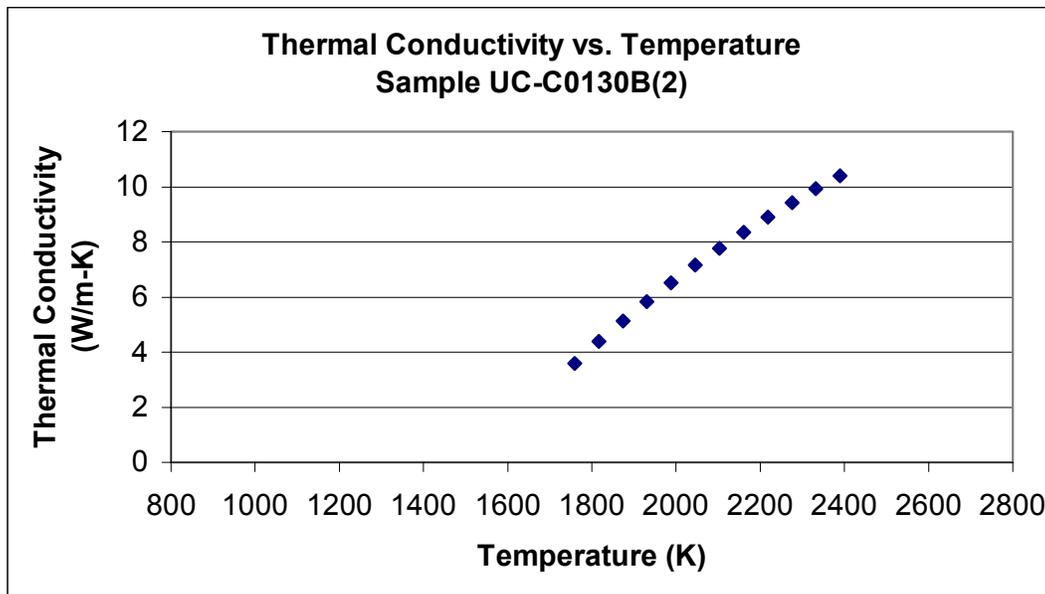
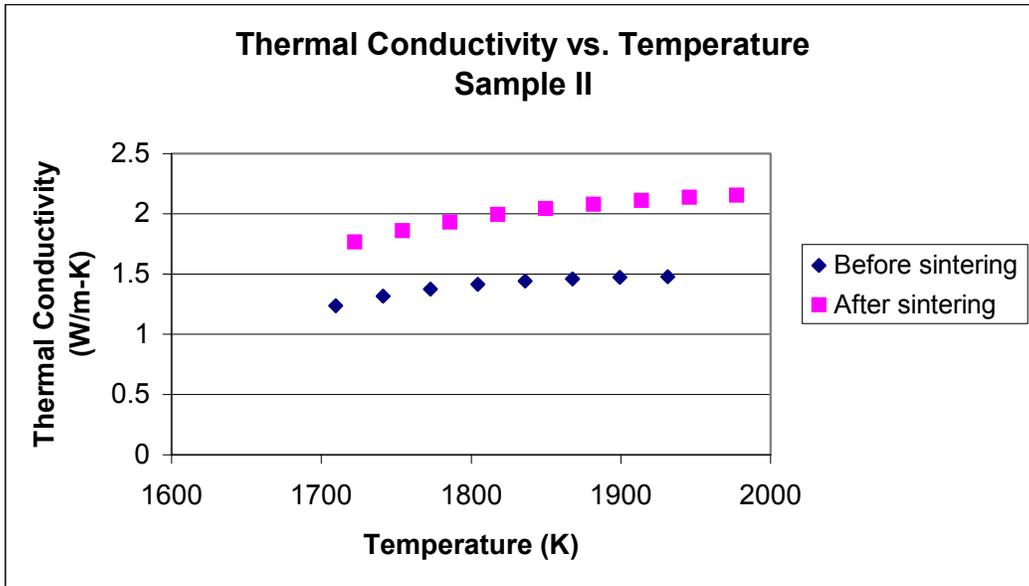


Fig. 6: Thermal conductivity of two UC₂ sample pellets showing the effects of sintering.