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THE THERMAL CONDUCTIVITY OF URANIUM

by

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A Monograph

METALLURGY DIVISION

April 1957

Operated by The University of Chicago
under
Contract W-31-109-eng-38

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FOREWORD

The literature on uranium metallurgy is relatively young in years, dating back to the initiation of the Manhattan Project in 1940. During the short span of the 16 years that have passed, there has accumulated a rather impressive body of technical information on the physical and metallurgical properties of this formerly relatively obscure metal. The information is contained largely in a variety of "progress reports," technical notes, memoranda, and notebooks of the various AEC-associated laboratories.

To the scientific researcher in this field, and especially to the newcomer, both scientist and engineer, the use of this material presents some difficult problems. To make a thorough literature search of a given subject he must examine a large number of monthly, quarterly, or annual progress reports, and cull from each the items pertinent to his subject. Having done so, more often than not he is apt to be confronted with a sheaf of data which are at times contradictory, incomplete, inaccurate, or misleading. This is generally inherent in the nature of "progress reports," particularly those which have not been subjected to open scientific scrutiny. In the early days of the Manhattan Project (1940-46) urgency was the overriding consideration. Many determinations and measurements of properties were made with the sole objective to provide in as short a time as possible a "reasonable" answer to some immediate engineering problems. Such data are bound to lack in accuracy and precision of method. Another factor to consider in this connection is the chemical purity of the metal that was used in the early work: it was not until about 1952 that high-purity material became available to permit a re-evaluation of some of the properties.

To put the information into more usable form, and to provide the research worker with a critical evaluation of the present status of our knowledge of this field, the Metallurgy Division at ANL has undertaken the preparation of a series of monographs on various subjects in the metallurgy of uranium and its alloys. The present monograph on "Thermal Conductivity" is the first in this series. The initial effort is confined to the physical metallurgy; it is hoped to expand the series later to include subjects on technology, e.g., fabrication, melting and casting, cladding, etc. The monographs are being prepared by various staff members who have actively participated in research activities in their respective subjects. Each monograph will contain a complete bibliography of all the references that had been searched and considered in the preparation of the monograph. The bibliography for this monograph includes references up to July 1, 1955. All later references will be included in future revisions of the monograph.

The authors of the monographs would appreciate receiving from readers and scientific workers who have contributed to the literature in this field, comments and suggestions on references and additional data that might have been inadvertently left out.

F. Foote
H. H. Chiswik

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THE THERMAL CONDUCTIVITY OF URANIUM

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GENERAL

To provide a background for the data presented, some introductory theory and a general survey of apparatus are given in the nature of auxiliary information, and no attempt has been made to cover the details of either of them completely. A detailed treatment of methods of measurement is given by Seibel.⁽⁸⁶⁾

Little or no interest was attached to the thermal conductivity of uranium prior to the work done under the auspices of the Manhattan Project. With the early work directed at producing controlled nuclear fission, a need was soon felt to know how rapidly heat could be removed from the uranium employed as a reactor fuel. Since that time many separate agencies engaged in atomic energy have performed their own particular tests, and, while a great deal of disconnected information has been published, there exists no single source which could embrace all this work, sum up results, and point out suitable frontiers for new workers in this field. This monograph is an attempt to provide this service relating to work up to the present time.

Engineering tests which were designed to give over-all heat transfer figures under special conditions of geometry, such as uranium slug measurements, have been excluded as not being directly applicable to the measurement of a precise physical quantity. While useful and even indispensable under the special conditions of the individual project, these programs have been classed as non-contributory for the purposes outlined herein.

Special interest and attention has been directed to the manner in which thermal conductivity varies with temperature. In addition to the work at elevated temperatures, recent investigations in the low Kelvin range, which are especially sensitive to purity, have been included.

The relation between electrical and thermal conductivity might have been more aptly treated space-wise under the heading of the monograph on electrical conductivity.⁽⁹⁴⁾ However, thermal testing methods are relatively difficult and the consequent desire to employ converted electrical data made inclusion of these relations under this heading seem logical and useful.

NOTES ON THEORY

Heat is a form of energy characterized by and consisting of random atomic, molecular, or electronic agitations. This energy is transferred in accordance with the laws of thermodynamics from the hot to the cooler regions of a substance. The mechanism of its transfer has received attention by a number of theoretical investigators since the year 1900.

A critical review of the "electron theories of matter" up to about 1930 has been presented by Hume-Rothery.⁽¹⁾ The earliest theory on thermal conductivity was advanced by Drude⁽²⁾ in 1900, who postulated the existence of a gas made up of electrons free to move about in metals and subject to the classical laws applying to an ideal gas. The simplifying assumptions forming the basis of this theory, although representing a striking advance at the time, were not suited to explain certain relationships to electrical conductivity, and, in particular, the phenomenon of superconductivity. There followed a long list of modifications of the original theory by different workers.

Thomson⁽³⁾ suggested a system of polarized electrons in combination with a small number of free electrons.

Wien⁽⁴⁾ and Grüneisen⁽⁵⁾ conceived the electrons as being limited to guided paths between the atoms, a view which was changed by Bridgman⁽⁶⁾ to mean guided paths through the atoms themselves.

Hall⁽⁷⁾ amended this concept to conceive of the electrons as consisting of two groups: one large group bound to the atoms, with energies independent of the temperature, and another small group of free electrons with energies proportional to the temperature.

Lindemann⁽⁸⁾ advanced another theory that the electrons did not move independently of one another, but moved through the material in a sort of lattice formation, and thus attempted to explain thermal transfer with all its attendant phenomena.

A great many of the obvious difficulties with all the classical attempts to explain heat transfer were met with the parallel development of the Heisenberg quantum mechanics and the Schroedinger wave mechanics. The concept of a continuous spectrum of energy was discarded in favor of one made up of tiny discrete pulses of energy behaving in accordance with statistical laws involving a probability function of energy transfer. All the classical physical theories were revised in the light of this new concept.

The first theory of thermal conduction in the light of this revision was by Sommerfeld,⁽⁹⁾ who returned to the idea of Drude of a free electron gas. In 1929, Houston⁽¹⁰⁾ again modified the theory in accordance with the

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existence of guided paths between atoms. Block⁽¹¹⁾ introduced the possibility of free electrons in motion in a periodic field of force corresponding to that supplied by the atomic spacing.

The band theory, expanded by Brillouin,⁽¹²⁾ served to explain the difference between electrical conductors and insulators (see Seitz⁽¹³⁾). Recent theoretical contributors have been Mott and Jones⁽²⁵⁾ and Makinson.⁽¹⁴⁾ A recent review has also been made by Kittel.⁽¹⁵⁾

The present status of the theory of heat transfer is that two types of mechanisms are involved. The first of these is electronic, or metallic, conduction. In this type, excitation of the outermost electrons of each atom proceeds from one region to another. The second means of transfer is directly from one atom to its neighbors, called lattice, atomic, or non-metallic conduction. Total conduction may be of one type or the other, or a combination of both in parallel. In cases where both types exist, they may also be interdependent.⁽¹⁴⁾ The resulting mathematics has become quite formidable and there is some doubt existing as to the feasibility of treating the mechanisms separately in view of lack of experimental control of the variables involved.

The degree to which electronic conduction is able to operate is a function of the average probable distance between collisions of electrons with other particles, generally atoms. This in turn depends on such things as regularity of the lattice, amount of thermal agitation, presence of foreign atoms, presence of unrelieved stresses, and other irregularities. This distance is called the "mean free path" of an electron through the material, and the electronic component of the thermal conductivity is a direct function of this parameter.

In lattice conduction, each atom in a crystal transmits part of its thermal energy to its neighbor in the colder directions, and a wave thereby travels to the cold end of the material. These waves are periodic, but are not of exact harmonic character. For this reason, waves following adjacent paths interfere with one another and with the electrons in what is referred to as "anharmonic coupling." The most probable distance between these centers of interference and between them and the conduction electrons is another type of "mean free path" which governs the lattice component of the conductivity of the material. By analogy with the term "photon" which applies to a quantum of electronic excitation, the energy quantum traveling along an atomic lattice is termed a "phonon."⁽¹⁵⁾ The phonon mean free path has a separate and distinct numerical value from the photon mean free path.

Pure metals conduct heat essentially by electronic conduction. Impure metals, alloys, disordered systems, and strained metals conduct heat by a combination of electronic and atomic means. Ionic crystals, non-metals, insulators and glasses are lattice conductors.

The degree by which the atomic lattice departs from a regularized pattern is some measure of the importance of lattice heat conduction.

Unlike electrical conductivity, in which a net transfer of electrons takes place from one end of a metallic rod to the other, thermal conductivity involves only the transfer of energy, from one electron to another, or to a neighboring atom. Most of the electrons in a metal are pictured as tightly bound to the central atomic cores, leaving a few loosely bound ones in the outer shells. These, upon receiving quanta of energy, jump to neighboring outer shells of other atoms, preferably those having unoccupied bands. The electrons themselves eventually return to their former equilibrium positions under lower quantum excitation, and so nothing but energy and momentum is transferred for the thermal case.

The thermal conductivity as a mathematical function of mean free paths of electrons and phonons is complex, especially as regards the anharmonic coupling, and complete evaluation has not been possible. Until this particular phase of solid state physics catches up with the experimental phase, only generalizations and qualitative conclusions are in order.

Since thermal agitation is one element which bears on the mean free path of both photons and phonons, the mean free path can be controlled by changing the temperature. The effect of raising the temperature is to decrease the mean free path of the electrons in the metal. Therefore, the effect of the other variables which modify heat conduction, such as purity, internal strains and the like, can be better studied at low temperatures, in which range the effect of thermal vibrations is less of a limiting factor. The only effect of variations in purity at high temperatures are slight differences in the slope and elevation of the curve of conductivity versus temperature.

Normally, as a metal is heated from the theoretical zero, a maximum of conductivity is reached within about 30°K, usually followed by a gradually decreasing conductivity up to high temperatures. Exceptions have been noted for aluminum,⁽¹⁾ for vanadium,⁽¹⁶⁾ and for uranium, which have been reported to have a slight positive slope for this curve. In each case, charges can be made that the samples were less than 100% pure, but the case for uranium seems to be better established, as will be shown in succeeding pages.

As a metal is progressively alloyed and its crystal structure thereby made more and more irregular, the slope of the conductivity-temperature curve generally becomes less negative, passes through the horizontal, and becomes increasingly positive in slope. At the same time the whole curve is displaced to a lower position.

If a metal is composed of grains which are anisotropic, it is to be expected that the conductivity will be dependent upon grain orientation, since the mean free path is dependent on the lattice constants, at least in

certain temperature ranges. That it is so dependent has been reported for various non-cubic metals by Hume-Rothery⁽¹⁾ and Bridgman,⁽¹⁹⁾ although more recent data between the temperatures of 84 to 241°C⁽¹⁷⁾ and for -40°C⁽¹⁸⁾ have indicated no important differences in the conductivity of uranium parallel and perpendicular, respectively, to the rolling direction. What differences do occur may be traceable to structure rather than texture⁽¹⁸⁾ in this case.

MEASUREMENT OF THERMAL CONDUCTIVITY

Thermal conductivity, by definition, applies to the transfer of heat within a material while all points of that material are at a steady temperature. It is an equilibrium property. Other types of tests, in which points of the specimen are not at steady temperatures, and in which observations are made of time-temperature relations at specific locations, do not measure thermal conductivity directly. The specific heat of the substance must also enter all but strictly static tests.

Transient type tests are described in the section entitled "Dynamic Methods," and they actually measure the "thermal diffusivity," which is a time-space-temperature relation. From these it is only possible to calculate the thermal conductivity by the simultaneous consideration of two other variables, the specific heat and the density. These methods have their own advantages under certain conditions.

Steady State Methods

The basic equation defining K, the thermal conductivity, is

$$K = \frac{Qd}{A t (T_2 - T_1)} \quad (1)$$

where Q = quantity of heat, having the dimensions of work, or energy

d = length of specimen, or distance between reference thermocouples

A = area of constant cross section

t = duration of heat flow

$T_2 - T_1$ = temperature difference between reference thermocouples.

If all points of the specimen are in thermal equilibrium, K is defined as the thermal conductivity, a property which varies from one material to another. It is a function of temperature, pressure, and the constitution of the material. It is generally considered to be independent of the temperature gradient existing at a particular point. The thermal conductivity calculated from the above basic equation is really an integrated value over the

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temperature range represented by $T_2 - T_1$. For small values of this difference, linearity of the relation is assumed, and the thermal conductivity is generally referred to the "mean temperature" of the measurement:

$$T_{\text{mean}} = \frac{T_2 - T_1}{2} .$$

The thermal conductivity of a substance, therefore, is defined as the amount of heat carried through one centimeter of the material, per square centimeter, per second, per unit temperature difference.

Units of thermal conductivity are expressed in the c.g.s. system as calories per second-centimeter-degree centigrade, but there are a number of other units in common use. The several units and their respective conversion factors appear in Table 1, together with related information for thermal calculations. The c.g.s. system is used in this monograph.

Thermal conductivity (or resistivity) could be measured in similar fashion to its electrical counterpart except for the fact that thermal insulators are not nearly as good as electrical insulators. Heat leaks quickly reduce the accuracy of poorly designed equipment, although methods are basically similar.

Absolute methods in thermal techniques correspond to ammeter-voltmeter methods in electricity, and there are thermal methods of comparison type analogous to Wheatstone bridge methods.

Absolute Methods

In the absolute method, a current of heat is made to flow along a specimen, as shown in Figure 1, usually a round bar, by the application of heat at one end and cooling at the other. Thermocouples placed at intervals along the bar measure the temperature gradient. As heat would radiate laterally from the bar and escape measurement, steps are taken to conserve this heat so that essentially all the applied heat will travel through the bar. These precautions vary from the simple use of insulation around the bar to elaborate shielding, or guarding, schemes which are designed to neutralize the radiation loss by exactly matching the temperature along the bar. This may take the form of a tube concentric with the specimen which is heated by an independent means.

When the temperatures in the whole assembly have been brought to a steady state, a process which usually takes several hours, the conductivity can be calculated from the power being supplied to the bar. Under such conditions, if E is the voltage and I the current supplied to the main heater, the factor Q/t in equation (1) will be, under unity power factor, $0.239 EI$ for c.g.s. units. A and d are constants measured on the specimen, and $T_2 - T_1$ is taken from the thermocouple readings.

Modifications of the method are legion and include such expedients as measuring the heat expelled at the cold end of the bar by a calorimeter and the use of odd specimen shapes with modification of heater arrangement. Apparatus designed for high temperature work in a vacuum have been described by Davey and Danielson⁽²⁰⁾ and by Moss.⁽²²⁾ An ingenious method employing resistance heating of the specimen itself has been described by Kohlrausch.⁽⁹¹⁾

For low temperature operation, different designs are required, although the principle is the same as shown in Figure 1. For the cold sink some kind of refrigerant is used, such as liquid nitrogen or helium. A very small heater coil is used at the hot end, and thermocouples, which are relatively insensitive at low temperatures, are replaced by gas thermometers. Difficulties from radiated heat losses are less than at elevated temperatures, however, and a thermal jacket surrounding the entire apparatus is usually sufficient to prevent large errors from this source. Rosenberg⁽²³⁾ has described a low-temperature thermal conductivity apparatus in some detail.

Comparative Methods

The comparative method of measuring conductivity has become widely used in recent years. In this method, an unknown sample is measured relative to another which has a known conductivity. Most of the schemes proposed for this type of measurement are modifications of the apparatus described by Shelton and Van Dusen⁽²⁴⁾ in 1934. This has been used chiefly at elevated temperatures and Figure 2 shows the essentials of the method.

A type adaptable to remote control operation has been described by Weeks and Seifert.⁽²¹⁾ Two identically shaped specimens are placed in series, one of which is a material with a known conductivity. A hot source and cold sink are provided as in Figure 2. The thermal shield is a composite of two materials, A and B, of lower and higher thermal conductivity, respectively. A central auxiliary heater is provided on the shield, which is used to equalize any difference in the temperature gradients on the guard with those on the specimen and the standard, respectively.

When equilibrium is established, the conductivities of the two samples at their respective mean temperatures are inversely proportional to their temperature gradients, as given by the thermocouple readings. Best accuracy results when the conductivities of specimen and standard roughly match.

This system possesses an advantage that the proportionality is independent of any heat loss from the source, a factor which would appear as an error in the absolute method.

A disadvantage is that any error already appearing in the calibrated standard will appear as an additional error in the determination of the conductivity of the unknown sample.

Technique and Margin of Error

The technique of attaching specimens to sources and sinks is quite important in any type test. All flat surfaces across which heat is intended to flow, as from source to specimen and specimen to sink, should preferably be welded, brazed, or soldered. If such is not feasible, good results are sometimes had by a process of lapping the mating surfaces or by inserting a soft copper or aluminum foil under pressure between the surfaces. Liquid bonds, such as molten tin, are also sometimes used, but in that case it is necessary to know that the liquid actually wets the surfaces of the mating pieces.

Another important experimental technique is the proper attachment of thermocouples. The bead may be spot welded to the surface of the metal, but alloying with the parent material may alter the calibration. The couple may be inserted into a hole drilled preferably past the center of the sample. Another widely used method of attachment is to make a saw cut in the specimen, insert the bead from the end of the cut, and pean the surrounding metal over the bead using a center punch. The bead may be further secured by pressing under an arbor press. The leads to the couple should then be wrapped for several turns around the specimen and withdrawn as nearly as possible through an isothermal region. By this expedient, any electrical insulation failure on the couple wires will result in a negligible error in reading potential.

The error to be expected in any steady state thermal conductivity test is comparatively large, as most physical tests go. It has been stated by Jakob⁽²⁶⁾ that the most careful techniques result in about 5% uncertainty, in spite of the claims of many observers to better accuracy. It must be admitted that apparatus used in thermal conductivity tests has its full measure of caprices and results should be judged accordingly.

Dynamic Methods

Although possessed with new interest, dynamic methods date back to 1861 to a method described by Angstrom.⁽²⁷⁾

These methods measure directly the thermal diffusivity of a material, which is defined as the thermal conductivity per heat capacity of a unit volume. Given portions of a sample in this case are not at constant temperature and the heat distribution function will depend on the specific heat, or how rapidly the material can gain and lose temperature.

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Thermal diffusivity k is defined as

$$k = \frac{K}{c \cdot \rho} \quad , \quad (2)$$

where K is the thermal conductivity, c is the specific heat, and ρ the density. The unit in the c.g.s. system is square centimeters per second. A derivation of diffusivity relations has been given by Johnson.⁽²⁸⁾

In practice the method of measurement consists of heating the specimen uniformly to some temperature and then impressing a variable harmonic temperature on one end of the bar while the other remains at constant temperature. Observations of the temperature fluctuations, usually made with a recording potentiometer, are made either of (1) the phase lag (or velocity), (2) the amplitude decrement, or (3) both simultaneously.

According to Sidles and Danielson,^(29, 30) the advantages of the dynamic method are the following:

- "1. Good thermal contacts between the sample and the heat source and between the sample and the heat sink are not important factors in evaluating the reliability of the measurements.
- "2. Insulating or guarding the sample to prevent heat losses is not necessary.
- "3. The thermal diffusivity is determined at a fixed temperature rather than as an average value over a large temperature range.
- "4. Sample dimensions and shape are such that only a small amount of material is required and no difficult fabrication problems are encountered."

However, Chao and El-Hifni⁽³¹⁾ have evaluated a heat loss factor for their experiments which may not be negligible under certain conditions. This factor arises from considerations of heat lost by the sample during the high temperature portion of the cycle which are uncompensated for by the heat gains during the low temperature portion of the cycle.

If both velocity and amplitude decrement are used simultaneously the expression for the diffusivity is given^(29, 30) as

$$k = Lv/2 \ln q \quad ,$$

where L is the reference distance, v the velocity, and " $\ln q$ " the natural logarithm of q , the amplitude decrement between the two reference points. Complete theoretical development and description of apparatus have been given by both pairs of investigators.^(29, 30, 31)

Certain other less refined methods involving time-space-temperature measurements are suitable for engineering evaluation, but as physical tests deserve only passing mention. They are useful only as a basis of screening acceptable materials while avoiding the time-consuming tests of higher precision.

The diffusivity data are seldom desired for themselves alone, and conversion to conductivity at any specific temperature may be made by the equation

$$K = k \cdot c \cdot \rho \quad ,$$

assuming that the specific heat and density are known at that temperature.

THERMAL CONDUCTIVITY OF URANIUM

For purposes of analysis, the entire body of thermal conductivity data on uranium has been dissected into four parts as follows:

- A. The preliminary Chicago data
- B. The pre-1948 elevated temperature data
- C. The post-1948 elevated temperature data
- D. Recent low temperature data

Groups B and C show a divergent character between 100 and 300°C. In fact, basic disagreements exist among all four groups.

The Preliminary Chicago Data

The first recorded observations of thermal conductivity were reported by Snyder and Kamin⁽³²⁾ in 1942, who stated later⁽³⁴⁾ that no guard was used on the specimen. On this early material, a value of 0.057* was measured for Westinghouse cast uranium and 0.037 for sintered powder obtained from Metal Hydrides, Inc. for temperatures 25-100°C. A report by Hilberry⁽³³⁾ commented that direct measurement of thermal conductivity gives a value 1/16 that of copper compared to 1/20 predicted by the electrical conductivity. Plott and Raeth later reported⁽³⁵⁾ 0.046-0.048 and 0.042-0.044 for the same materials, respectively. In another report⁽³⁶⁾ their summary also included Snyder and Kamin's data. These same values were subsequently requested five times in later reports: Creutz,⁽³⁷⁾ Mulliken and Lane,⁽³⁸⁾ Hirsch,⁽³⁹⁾ Simon,⁽⁴⁰⁾ and Directorate of Tube

*All units are c.g.s.

Alloys.⁽⁴¹⁾ Thus, the ten previous references⁽³²⁻⁴¹⁾ are based on two original observations^(32,35) averaging 0.057 and 0.047 for cast uranium, respectively.

Although the first of these figures is well within the present day scatter band, the fact that impure metal was used without thermal shielding suggests that this early work, which admittedly faced many difficulties, should now be discounted. However, the range of data has been shown in Figure 3 for comparison.

It is worthy of comment that Simon,⁽⁴⁰⁾ even at this early stage, was able to estimate a value of 0.07 at 25°C for very pure metal on the basis of theory, a value very close to present day measurements.

The Pre-1948 Elevated Temperature Data

The data on thermal conductivity from 1943 to 1947, inclusive, are shown on Figure 4. These are distinct from the post-1948 data (in solid lines in Figure 5), described in the next section, to illustrate the different curvature of the two groups. This curvature has been noted by Katz and Rabinowitch,⁽⁸⁰⁾ Franklin and Turkel,⁽⁸¹⁾ and in the Reactor Engineering Handbook.⁽⁸²⁾

The cause of this diverse behavior must be blamed either on chance, different testing procedure, or different material.

In 1943, four independent runs are indicated in reports published by Kratz and Raeth,⁽⁴²⁾ Kratz,^(43, 44, 54) and Creutz.⁽⁴⁵⁾ Although the data diverged at other temperatures, results at 200°C were all in the narrow range 0.064-0.065. One curve even exhibited a cusp.⁽⁴⁴⁾ Kratz and Raeth did their work on extruded one-inch rod,* but Creutz states only that his material was of "commercial purity." Kratz states that a thermal shield was used in his apparatus, with a comparative system employing a brass standard. The conductivity of the standard was estimated rather than measured, and he stressed the accuracy of only the slope of his curves.

In 1944 Kratz^(46, 47) gave room temperature data which showed quite high conductivity for the period, near 0.070 at 100°C, extrapolated. The only comment on material was that it had "been heated in a vacuum for several days." In the same year, the National Physical Laboratory (England) was credited⁽⁴¹⁾ with the figure of 0.06 for 20 to 100°C.

In 1945, Raeth^(48, 50, 51) confirmed Kratz's 1944 high conductivity results in a series of tests, presumably on the same apparatus with an estimated standard, varying the heat treatment. Discrepancies between

*Extrusion temperature unknown, but believed to be gamma extruded.

supposedly identical tests,(48, 51) however, cast doubt upon the whole series, as far as absolute values are concerned. The same observers, however, were reporting(49) tests with low and intermediate conductivities with purity as a variable. Unfortunately the purer material was the one with the low conductivity, so no dependable facts were secured, although Lucks(52) at Battelle was able to confirm one of their curves. Robinson and Sterling(53) of the Bureau of Standards were able to confirm the other one of Kratz and Raeth's results, thus completing the confusion for the year 1945.

In 1947, Grenell(55) confirmed the data of Lucks,(52) using Lucks' equipment, apparently.

To summarize, in the group of results from 1943 to 1947, no positive correlation can be achieved between conductivity and purity, heat treatment, or preferred grain orientation, although it is apparent from the average values that the group as a whole had better uranium to work with than did the 1942 Chicago group. The variations in conductivity, which appear to lie in three discrete sub-groups in Figures 4 and 5, seem to reflect similar results for similar equipment because of the timing and location. It may be that systematic errors associated with the particular equipment were responsible for the scatter in reported results. The common feature of the whole group of curves, that of a tendency toward concavity downward, might just possibly be explained by inadequate vacua and progressive gaseous contamination of the uranium at the higher temperatures.

The Post-1948 Elevated Temperature Data

The data in this group have been furnished by Babbitt, et al,(56) Sidles and Danielson(57, 30) (from diffusivity data), and Deem and Nelson,(59,62) and have been carried up to higher temperatures than those previously done (Figure 4). An interesting complement to the above work, which was on bars measured in the rolling direction, are the experiments by Droher and others(18, 58, 60, 61, 63) on material measured in the radial direction of a bar (Figure 5) and by Weeks(83) on high purity uranium.

This group, shown in Figure 5, occupies essentially the same scatter band as the previous group, but differs from it in that the curves of the later group are concave upward in the comparable 100-300°C region. It is not clear why this later group of curves feature a positive inflection unless there are some uncompensated radiation losses not present in the earlier group. This seems somewhat unlikely with equipment which has been specially designed for higher temperatures.

The Canadian work opened a controversy on stability of the thermal conductivity in the alpha phase. Babbitt, et al(56) reported a continuously increasing thermal conductivity as more tests were run and specimens were reheated a number of times. Their last reported curve (Figure 6) is the

highest conductivity ever reported for uranium. They raised the question as to whether it was possible to stabilize the conductivity of uranium in the alpha region. Their measurements were made in the rolling direction of a gamma-extruded bar, 2.5 centimeters in diameter and 8 centimeters long. Their analysis shows the presence of 260 ppm silicon, 188 ppm iron, along with amounts of chromium, nickel, manganese, copper, silver, and cobalt aggregating 87 ppm.

The Canadian results are not confirmed by Droher and others^(18, 58, 60, 61, 63) of North American Aviation, although the preponderance of their measurements were made in the radial direction of bars. Even though a slight shift was noted between alpha-rolled and beta-quenched material, all further indications were for stability of the latter, through five repeated excursions of temperature.

Additional tests were made on strips cut both parallel and perpendicular to the rolling direction. The conductivity of these strips was measured as a function of repeated annealing at temperatures to 1000°C, and no significant dependence of the conductivity on annealing temperature was noted for either direction. These tests were carried out at a mean temperature of -40°C, however. The literature carries no earlier comment on transverse conductivity except two by Kratz (1943-4),^(85, 84) in which latter reference he states, "there is some indication that the thermal conductivity may be higher in the radial direction than in the direction of extrusion."

The positive inflection of the curves for radial thermal conductivity is very slight as compared to the longitudinal data shown in Figure 6. The scatter band is lower for the radial measurements. Whether either of these effects, or both, are dependent on preferred crystal orientation or on the presence of elongated foreign phases or materials, would be indicated only by further investigation in which these variables are separated. On the basis of Droher's tests on strips at -40°C, however, the effects of crystal anisotropy appeared to be non-existent.

As to any discontinuities in the thermal conductivity versus temperature curve at 660°C, the alpha-beta transition temperature, Sidles⁽⁵⁷⁾ has noted such a discontinuity in the diffusivity curve for uranium, but also notes an equal and opposite change in the specific heat. (See also supporting quarterly report references 69 through 78 for Iowa State College.) The inference is that no discontinuity should occur in the conductivity curve at this point. Droher⁽⁵⁸⁾ remarks, in confirmation, about samples having part of their core in alpha and part in beta, and others having part in beta and part in gamma, "there was no significant variation in the thermal conductivity of bi-phase uranium from the thermal conductivity curve." A slight variation noted by Droher and Domingo⁽⁶¹⁾ is claimed by the investigators to be a possible statistical variation. At the writing of this

monograph, Erikson and Hålg⁽⁹²⁾ report a linear curve from 0 to 800°C, with no discontinuities (see Figure 4). At any rate, continuity cannot be regarded as established beyond a reasonable doubt. There are definite discontinuities in the electrical conductivity at the transition temperatures.

It appears that the post-1948 data have provoked rather than answered questions. Some of them are as follows: (1) Is the thermal conductivity really stable? (2) Is the conductivity-temperature curve really concave upward? (3) Are there discontinuities at the transition temperatures? (4) What are the roles of purity, structure, and texture? (5) What is responsible for the width of the scatter band, material differences or unavoidable experimental factors?

Recent Low Temperature Data

Low temperature data on uranium have been reported by the Los Alamos Scientific Laboratory,^(64, 65) Tyler, Wilson and Wolga⁽⁶⁶⁾ of Knolls Atomic Power Laboratory, and Rosenberg^(23, 79, 90) of the University of Oxford. These results are shown together in Figure 7.

The same scatter is evident as in the previous group, Rosenberg's data being about 43 per cent higher than the Knoll's data at 20°K. In a region where purity should have large consideration, no chemical analysis are available. Following are the stated material types:

Los Alamos:	Alpha rolled, heat treated at 745°C for six hours in helium. Slowly cooled to 540°C, then rapidly cooled to room temperature.
Knolls:	Normal uranium. Heated 10 minutes in lead bath. Water quenched to 50°C.
Oxford:	Unannealed rod from AERE. Purity not specified but "believed to be very high."

The effect of structure and of preferred orientation may again represent the variation of results, as many different forms are possible.

One anomaly is the apparent discontinuity of the scatters of the low temperature group with groups B and C. If the data are apparatus-sensitive, it would be interesting to be able to carry each method over the room temperature dividing line. The best match between single observers appears to be the Knolls cryogenic curve⁽⁶⁶⁾ and the Battelle high temperature curve⁽⁵⁹⁾ in which one is an approximate extension of the other.

With the close agreement of thermal and converted electrical data, most observers are convinced that thermal conduction in uranium is largely

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electronic. Rosenberg,⁽²³⁾ however, is inclined to accept some lattice conductivity as low as 60°K, and Bennett⁽⁹³⁾ appears to draw the same conclusion.

The unresolved practical questions here are as follows: (1) What is responsible for the large scatter among different observers? (2) What experimental factor causes an apparent room temperature discontinuity?

DATA CONVERTED FROM ELECTRICAL CONDUCTIVITY

Use is frequently made of relations with electrical conductivity for pure metals, that is, those whose thermal conductivity would be expected to be largely electronic in character.

In 1853, Wiedemann and Franz⁽⁸⁷⁾ first observed this relation, which states that at ordinary temperatures the ratio of the thermal and electrical conductivities is approximately constant for all pure metals, and is about 14×10^{-7} at 0°C. Lorentz⁽⁸⁸⁾ later observed a temperature dependence of this ratio and suggested the form

$$L = \frac{K}{CT} = \frac{KP}{T},$$

where K is the thermal conductivity, C the electrical conductivity, P the electrical resistivity, and T the Kelvin temperature. L is thus correctly defined as the "Lorentz Constant," although it is also frequently called the Wiedemann-Franz ratio, a term which should be reserved for the older formula. The commonly accepted value of the constant is 5.8×10^{-9} .

The constancy of this "constant" is far from universal.⁽¹⁾ Silver, copper and gold obey the relation at high temperatures but not at low temperatures. Beryllium and beta-manganese show large deviations (see Seitz,⁽¹³⁾ pp. 176-8).

Based on electrical resistivity data presented in the monograph on electrical conductivity⁽⁹⁴⁾ and an assumed constant 5.8×10^{-9} , a calculated thermal conductivity curve versus temperature is given in Figures 4 and 5 along with the experimental curves. It is to be noted that the curve lies within the experimental scatter band below 500°C, deviating slightly below it at higher temperatures. This could be construed as further indication of uncompensated radiation losses reflected in the experimental data.

Since experimental scatter is so much less on the electrical than on the thermal results, it is felt that considerable weight should be attached to the calculated curve as representing the pure state, even considering the assumed Lorentz constant. It is to be emphasized, however, that such

calculation reflects only the electronic component of thermal transfer, and would be in error by the amount of any lattice component, together with the amount of any interaction between the two.

Early attempts^(40, 89) to calculate the thermal conductivity of uranium by this method were handicapped by poor material. The Lorentz constant for uranium was recently calculated⁽⁶⁶⁾ for low temperatures as follows:

<u>Temperature (°K)</u>	<u>Lorentz Constant (c.g.s.)</u>
20	5.67×10^{-9}
60	5.96
100	6.65
140	6.96
180	6.96
220	6.82
260	6.60
300	6.45

The authors state that this is in agreement with the work of Mendelssohn and Rosenberg⁽⁹⁰⁾ for below 20°K. The values show signs of approaching the conventional 5.8×10^{-9} value at higher temperatures.

CONCLUDING REMARKS

In Figure 8 is shown a curve which is a composite of what appear to be the best available values to date of the thermal conductivity of uranium from absolute zero to 900°C. This curve approximates Rosenberg's data⁽⁹⁰⁾ in the low Kelvin range, a general coverage of the Knolls⁽⁶⁶⁾ and Los Alamos⁽⁶⁵⁾ data up to room temperature, and Battelle's data⁽⁵⁹⁾ above room temperature. This is understood to apply to commercial purity uranium, beta annealed, along the rolling direction.

The curve is subject, however, to serious difficiencies and uncertainties as cited below.

1. The reason is not known for the apparent discontinuity in the scatter bands of Figure 3 at room temperature. This may be attributed to some hitherto unsuspected factor of technique in one or the other types of equipment.

2. Although theory calls for large differences between conductivities in different directions of highly oriented uranium, such wide divergence has not been reported. It may be that the effects of structure, such as elongated stringers of foreign phases, compensate for the effects of texture to some extent.

3. Although it would seem that Eriksen and Hålg⁽⁹²⁾ have proved beyond reasonable doubt that the curve is continuous over the transition temperatures, at 600 and 770°C, additional confirmation is desirable.

4. Additional confirmation as to the thermal stability of beta-treated uranium with repeated heating in the alpha range would be desirable.

5. The effect of trace impurities as a function of temperature has not been assessed in a satisfactory manner.

6. Reproducibility of testing still appears to be less than desirable as determined from one piece of equipment to another, as shown by the considerable width of the scatter bands. This may be due in part to differences in material being tested, but such view is not supported by an examination of electrical resistivity data.

A better understanding of the thermal conductivity of uranium appears to await filling of some of the above named gaps.

Table 1

THERMAL UNITS AND CONVERSION FACTORS

$$1 \text{ calorie} = 0.00397 \text{ BTU}$$

$$1 \text{ watt} = 3.41 \frac{\text{BTU}}{\text{hr}} = 0.239 \frac{\text{cal}}{\text{sec}}$$

$$1 \text{ BTU} = 252 \text{ cal} = 17.58 \frac{\text{watts}}{\text{min}}$$

$$1 \frac{\text{BTU}}{\text{hr}} = 0.084 \frac{\text{cal}}{\text{sec}} = 0.293 \text{ watts}$$

$$1 \frac{\text{cal cm.}}{\text{cm.}^2 \text{ sec. } ^\circ\text{C}} = 241.9 \frac{\text{BTU ft.}}{\text{ft.}^2 \text{ hr. } ^\circ\text{F}} = 4.183 \frac{\text{watt cm.}}{\text{cm.}^2 \text{ } ^\circ\text{C}}$$

$$0.00413 \frac{\text{cal cm.}}{\text{cm.}^2 \text{ sec. } ^\circ\text{C}} = 1 \frac{\text{BTU ft.}}{\text{ft.}^2 \text{ hr. } ^\circ\text{F}} = 0.0173 \frac{\text{watt cm.}}{\text{cm.}^2 \text{ } ^\circ\text{C}}$$

$$0.239 \frac{\text{cal cm.}}{\text{cm.}^2 \text{ sec. } ^\circ\text{C}} = \frac{\text{BTU ft.}}{\text{ft.}^2 \text{ hr. } ^\circ\text{F}} = 1 \frac{\text{watt cm.}}{\text{cm.}^2 \text{ } ^\circ\text{C}}$$

$$1 \frac{\text{BTU ft.}}{\text{ft.}^2 \text{ hr. } ^\circ\text{F}} = 12 \frac{\text{BTU in.}^*}{\text{ft.}^2 \text{ hr. } ^\circ\text{F}}$$

*This unit is in common use in the insulation trade.

FIG.1 ABSOLUTE METHOD FOR THERMAL CONDUCTIVITY TEST

FIG. 2 COMPARATIVE METHOD FOR THERMAL CONDUCTIVITY TEST

FIG. 3. SCATTER BANDS FOR DATA GROUPS

FIG. 4. THERMAL CONDUCTIVITY VS TEMPERATURE DATA FOR URANIUM AT ELEVATED TEMPERATURES

FIG. 5. THERMAL CONDUCTIVITY VS TEMPERATURE DATA FOR URANIUM AT ELEVATED TEMPERATURES

FIG. 6. THERMAL CONDUCTIVITY ALONG AND ACROSS ROLLING DIRECTION

FIG. 7. THERMAL CONDUCTIVITY VS TEMPERATURE DATA FOR URANIUM AT LOW TEMPERATURES

FIG. 8 THERMAL CONDUCTIVITY OF URANIUM

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