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SURVEY ON THE MEASUREMENT OF THERMAL CONDUCTIVITY OF SOLIDS PRODUCED BY EVAPORATION AND CALCINATION OF SYNTHETIC FUEL REPROCESSING SOLUTIONS

H. W. Godbee
J. T. Roberts

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CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

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ABSTRACT

Simplified equations to calculate the maximum temperature rise in stored solid wastes are given. The need for thermal conductivity values of solid wastes is demonstrated using these equations. For 55-gal containers of a calcined TBP-25 waste in a granite cave the temperature would rise 694-3307°F depending on the decay prior to storage. A program is proposed for measuring the thermal conductivity of solids produced by evaporation and calcination of synthetic fuel reprocessing solutions. Thermal conductivity is defined and factors influencing the thermal conductivity of nonmetallic solids are considered. Particularly the effects of temperature and porosity on thermal conductivity are considered. Methods of measuring thermal conductivity are outlined. Criteria used in selecting an arrangement of apparatus for measuring thermal conductivity are enumerated. An apparatus for determing the thermal conductivity of solids produced by evaporation and calcination of synthetic fuel reprocessing solutions is described.

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1.0 INTRODUCTION

This report records data on thermal conductivity obtained in an extensive literature search as the first step in a program of experimental determination of thermal conductivities, as functions of temperature and porosity, of the more important types of solid wastes. The containment or storage of wastes from the processing of nuclear fuels must take into account, among other things, the dissipation of heat generated by the radioactive decay of unstable isotopes. Wastes must be confined under conditions that preclude the attainment of a temperature detrimental to the waste, its container, or the surrounding medium. Since the reduction of liquid wastes to less mobile, less corrosive, less voluminous solids before storage is a promising waste disposal scheme, a knowledge of the temperature rise to be expected in stored solid wastes is needed. The calculation of a numerical temperature within a system as a function of space and time coordinates requires a knowledge of the thermal properties of the system.

2.0 TEMPERATURE RISE IN STORED WASTES

A proposed waste disposal scheme provides for storage of solid waste in a dry cave or vault (Fig. 2.1). For satisfactory storage, the heat generated by fission product decay would have to be removed since excessive temperature rise could develop under some conditions. Among the factors influencing temperature rise in stored solid wastes is the thermal conductivity of the solid.

The need for accurate thermal conductivity values of solid wastes can best be illustrated by an example. For the case of calcined waste stored in a cave or vault, the maximum temperature rise in the waste can be estimated if the storage system is approximated by a spherical waste package in a spherical cave. Equations for the maximum temperature rise (1) in the waste from center to surface, (2) across the space between waste and cave wall, and (3) at the cave wall for this system are listed in Table 2.1. For 55-gal containers of waste in a cave cut into a granite formation, the total temperature rise is 3307°F in a waste decayed 120 days prior to storage and is 694°F in a waste decayed one year prior to storage. The largest part of the total temperature rise is due to the rise from center to surface of the waste package, which is inversely proportional to the thermal conductivity.

The "coefficient of thermal conductivity" or, more simply, the "thermal conductivity," is that property of a material which determines the temperature gradient under fixed heat flow. The definition of thermal conductivity is contained in the generalized statement of experience with heat flow,[29] generally called Fourier's law:

$$q = -kA\frac{dT}{dx} \qquad (1)$$

where the heat flow and the temperature gradient are perpendicular to the area A. The thermal conductivity depends on the chemical composition, the physical

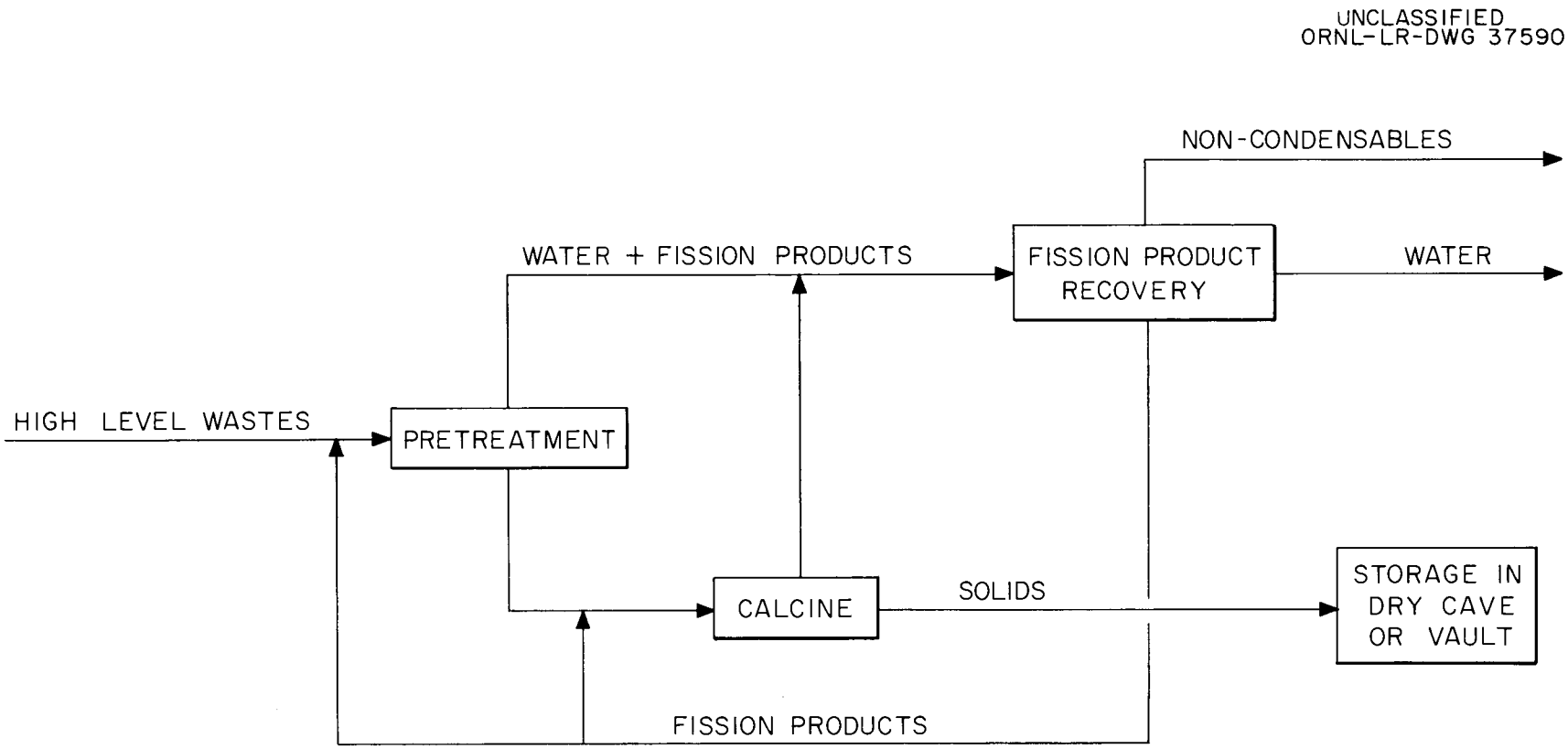

Fig. 2.1. General flow sheet for reduction of radioactive waste to solids.[13]

Table 2.1 Estimation of Temperature Rises in Stored Calcined Wastes[a]

	Temperature Rise (ΔT), °F	
	(a) Decayed 120 days prior to storage	(b) Decayed 1 year prior to storage
Temperature rise in waste from center to surface, $\Delta T_W = \frac{QR^2}{6k}$	2590	544
Temperature rise across the space between waste and cave wall, $\Delta T_C = \frac{QR}{3h}\left[1 + \left(\frac{R}{r}\right)^2\right]$	630	132
Temperature rise at cave wall, $\Delta T_C = \frac{QR^2}{3K}\left(\frac{R}{r}\right)$	87	18
Total temperature rise in waste, $\Delta T_{total} = \Delta T_W + \Delta T_S + \Delta T_C$	3307	694

[a]Properties of system: Q = (a) 1056, (b) 223 Btu/hr·ft^3 (Sec. 5.2)
R = 1.21 ft (55 gal sphere)
r = 6 ft
k = 0.1 Btu/hr·ft·°F (Sec. 5.3)
h = 0.7 Btu/hr·ft^2·°F (Sec. 5.4)
K = 1 Btu/hr·ft·°F (Sec. 5.5)

The symbols are defined in Sec. 5.1

structure, and the state of a substance (Fig. 2.2). It is not a constant for any one material, but may be a function of a number of variables. For example, factors which may influence the thermal conductivity of nonmetallic solids include:

a. Chemical composition, molecular structure in pure compounds, and impurities in solid solutions

b. Physical texture: (1) porosity, total void space and size and shape of pores with solid phase continuous, size and shape of grains with gas phase continuous; (2) presence of a vitreous or liquid phase, total amount and distribution of phase; (3) development of ceramic bond and sintering, time and temperature of firing; and (4) anisotropy and directional effects

c. Temperature

d. Pressure, stress, or strain

e. Heat flow

The thermal conductivity of most metals, at ordinary temperatures, shows a small and nearly linear decrease with increase of temperature, but a few (e.g., aluminum and platinum) show the opposite effect, as do also many alloys. The thermal conductivity of most nonmetallic materials varies considerably with temperature (Fig. 2.3). The k of many crystalline materials decreases with increasing temperature, being in general proportional to the reciprocal of the absolute temperature. Kingery[18] attributes the increase in thermal conductivity of BeO, MgO, and Al_2O_3 at high temperature to increased heat transmission by radiation. However, Jamieson and Lawson[16] show that the contribution to thermal conductivity from radiative transfer is too small to fit the observed deviations from a T^{-1} relation. Further, they suggest that the increased thermal conductivity may be explained by a flow of excitation energy (excitons). The k of an amorphous body, such as fused silica (Fig. 2.3), increases with increasing temperature, but the temperature dependence of k cannot be predicted with certainty and thermal conductivity data cannot be reliably extrapolated to higher temperatures.

Among the many factors that influence thermal conductivity, physical texture causes the greatest variation (Figs. 2.4 and 2.5).[5,7,12] There are two types of porous materials, cellular and granular. A cellular material is a two-phase system in which the solid phase is continuous and the gas phase is dispersed. A refractory in which voids (cells) are formed by the volatilization or combustion of some ingredient during firing is representative of this type of substance. A granular material is a two-phase system in which the gas phase is continuous and the solid phase is dispersed. Snow, sands, wools, and dusts are representative of this type of substance. A granular material has a lower conductivity than a cellular one of the same material and porosity. Several theoretical equations for the effect of porosity on thermal conductivity are listed in Table 2.2. A brief critical study of structural schemes assumed in deriving theoretical relations for the effect of porosity on thermal conductivity is given by Babanov.[6] Austin,[5] Waddams,[32] Barrett,[7] and Wygant and Crowley[34] found that Russell's equation yields results fitting their experimental data.

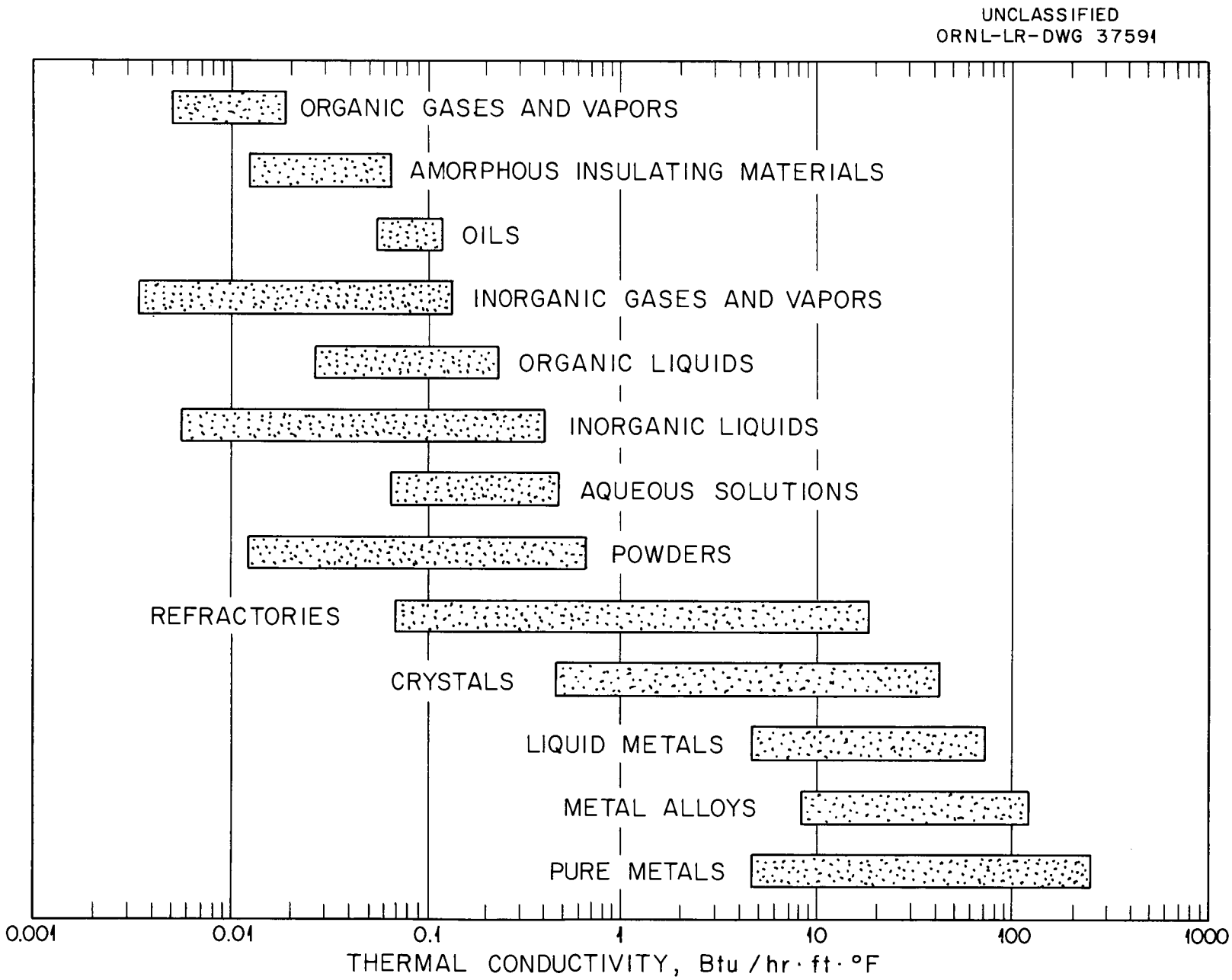

Fig. 2.2. Approximate order of magnitude of thermal conductivity for various substances at ordinary temperatures.[30]

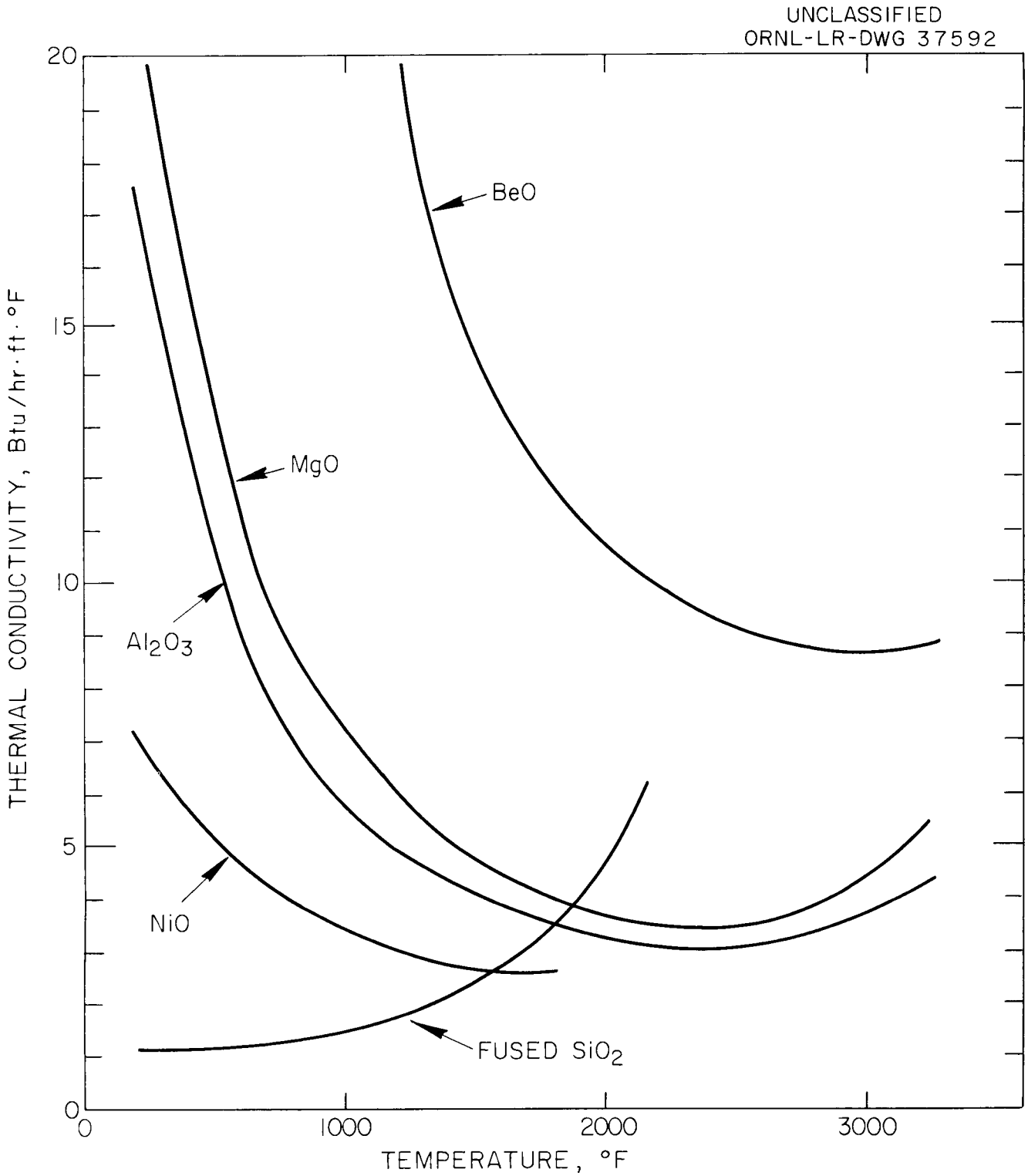

Fig. 2.3. Variation of thermal conductivity with temperature.[18]

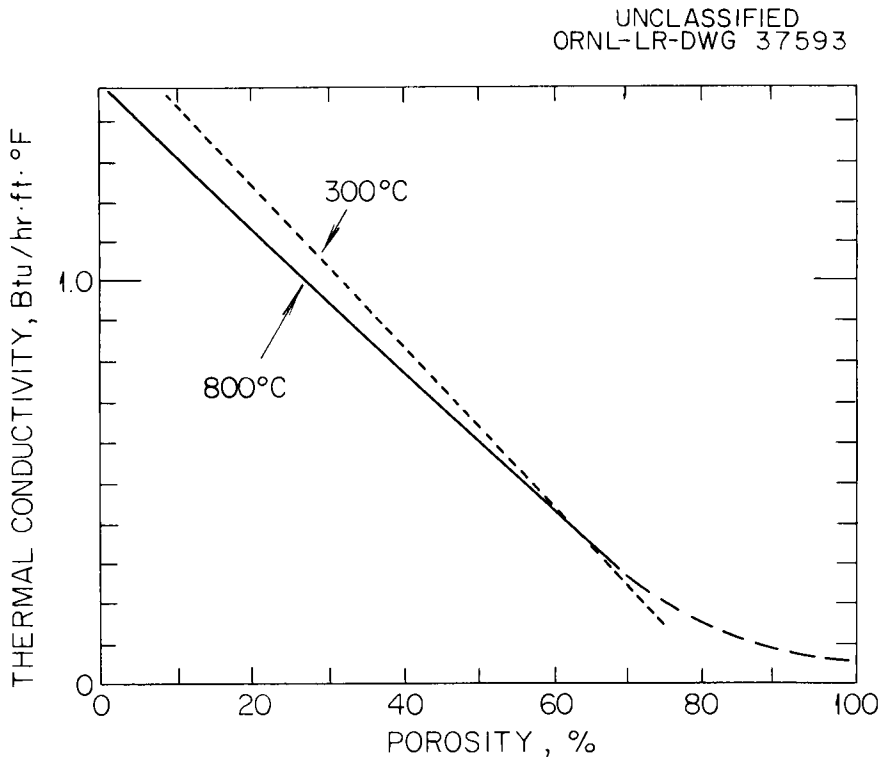

Fig. 2.4. Variation of thermal conductivity with porosity in corundum brick at constant temperature.[5]

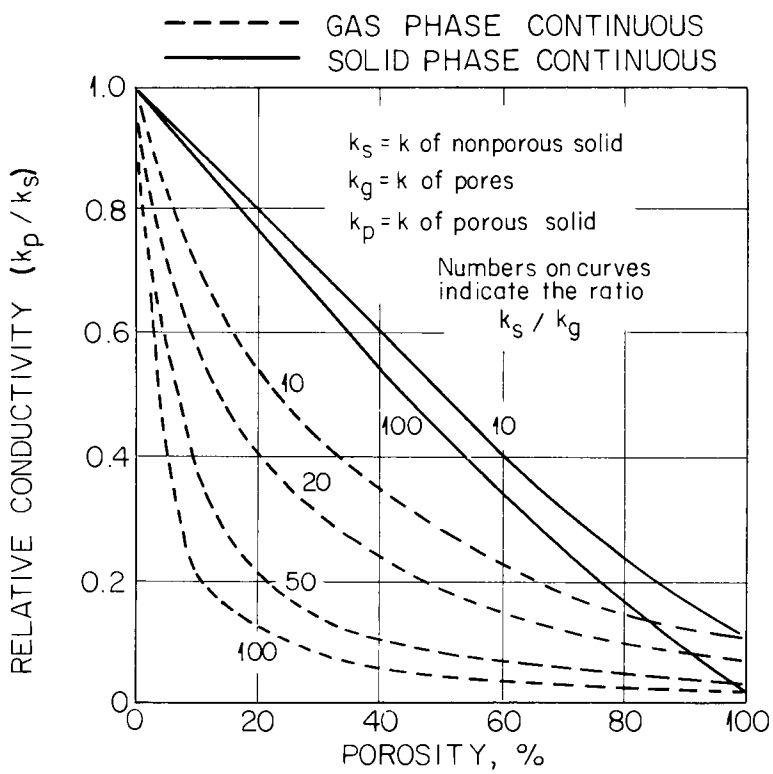

Fig. 2.5. Variation of thermal conductivity with porosity as calculated by Russell's equation.[28]

Table 2.2. Equations for Effect of Porosity on Thermal Conductivity

Reference	Equation[a] for Cellular Material (solid phase continuous, gas phase dispersed)	
Eucken[11]	$k_p = k_s \dfrac{1 + 2P\left(\dfrac{1 - k_s/k_g}{2k_s/k_g + 1}\right)}{1 - P\left(\dfrac{1 - k_s/k_g}{2k_s/k_g + 1}\right)}$	γ = shape factor ϵ = emissivity σ = radiation constant d = pore dimension P = porosity k_s = k of nonporous solid k_g = k of pores k_p = k of porous solid T = absolute temperature
Russell[28]	$k_p = k_s \dfrac{P^{2/3} + \dfrac{k_s}{k_g}(1 - P^{2/3})}{P^{2/3} - P + \dfrac{k_s}{k_g}(1 - P^{2/3} + P)}$	
Ribaud[25]	$k_p = k_s(1 - P^{2/3}) + k_g P^{1/3}$	
Loeb[22]	$k_p = k_s(1 - P) + \dfrac{P}{(1 - P) + \dfrac{k_s P}{4\gamma\epsilon\sigma d T^3}}$	

[a]Eucken's and Russell's equations above may be modified to apply to a granular material (gas phase continuous, solid phase dispersed) by replacing P by $(1 - P)$ and by interchanging k_s and k_g.

Table 3.1. Equations for Thermal Conductivity Obtained By Integrating Fourier's Law for Simple Shapes

Shape	Equation	
Plane plate		a = semiminor axis of prolate spheroid c = semifocal length of spheroidal coordinates $\overline{k}$ = mean thermal conductivity L = length of cylinder q = heat flow rate R = radius of circle, sphere, or cylinder s = side of square T = temperature x = thickness of plate 1 = inner isothermal surface 2 = outer isothermal surface
(a) square	$\overline{k} = \dfrac{q(x_2 - x_1)}{s^2(T_1 - T_2)}$	
(b) round	$\overline{k} = \dfrac{q(x_2 - x_1)}{\pi R^2(T_1 - T_2)}$	
Sphere (hollow)	$\overline{k} = \dfrac{q\left(\dfrac{1}{R_1} - \dfrac{1}{R_2}\right)}{4\pi(T_1 - T_2)}$	
Cylinder (hollow)	$\overline{k} = \dfrac{q \ln(R_2/R_1)}{2\pi L(T_1 - T_2)}$	
Prolate spheroid[a] (hollow)	$\overline{k} = \dfrac{q \ln\left(\dfrac{\sqrt{a_2^2 + c^2} - c}{\sqrt{a_2^2 + c^2} + c} \cdot \dfrac{\sqrt{a_1^2 + c^2} + c}{\sqrt{a_1^2 + c^2} - c}\right)}{8\pi c(T_1 - T_2)}$	

[a]For a derivation of the equation for a prolate spheroid see reference 2.

Factors influencing thermal conductivity are treated in more detail by Austin,[5] Jakob,[14] Wilkes,[33] Barrett,[7] Kingery and McQuarrie,[19] Francl and Kingery,[12] and Kingery.[18]

3.0 MEASUREMENT OF THERMAL CONDUCTIVITY

Thermal conductivity may be measured by either static or dynamic methods. In static methods the sample is allowed to come to steady state; i.e., temperature is a function of a space coordinate only, and the temperature at two or more positions is measured. The thermal conductivity is then determined from an integrated form of eq. 1. In dynamic methods the sample is in an unsteady state; i.e., temperature is a function of a time coordinate as well as a space coordinate, and the temperature change with time at one or more positions is measured. The thermal diffusivity, α, is then determined from an appropriate solution of the equation which underlies thermal conduction and temperature history,[15] usually referred to as Poisson's equation:*

$$\frac{dT}{d\theta} = \alpha\left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}\right) + \frac{Q}{\rho c} \qquad (2)$$

A survey of the literature on k reveals that steady-state methods of measuring k are more widely used than unsteady-state methods; however, unsteady-state methods are becoming more widely used since instruments for accurately recording rapidly changing temperatures are available. Static methods determine k directly while dynamic methods generally determine thermal diffusivity, α.** Dynamic methods that determine thermal diffusivity require a knowledge of density and specific heat before thermal conductivity can be determined. Ross[27] has recently reviewed dynamic methods of measuring k for solids. Since confidence in dynamic methods is usually based on agreement with static methods, the use of steady-state methods appears to be desirable until a fairly comprehensive background of data has been acquired. Thus, unsteady-state methods will not be considered further in this report.

In static methods for measuring thermal conductivity, test specimens take a shape for which an exact solution of eq. 1 is possible. These shapes are a plane plate (slab), a sphere, a prolate spheroid, and a cylinder. Solutions of eq. 1 for these shapes are listed in Table 3.1. In flat plate arrangements heat flows through a sample of constant cross section whose lateral surfaces are, ideally, covered with a nonconductor of heat. Since there is

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* For a derivation of this equation, refer to a standard text on heat flow such as Jakob[15] or Schneider.[31]

** In this connection, the "thermal conductivity probe" method should be mentioned. This is a dynamic method which has been developed to allow direct measurement of k. It is based on an approximate relation between k and the temperature rise in an infinite homogeneous medium caused by an infinite line heat source of constant strength.[21,23]

no perfect thermal insulator, a technique frequently used to reduce lateral heat flow to negligible proportions is the provision of heat guards (separately heated portions of the test material) whose temperature profile matches as closely as is practical the temperature profile of the test sample (Fig. 3.1). The present standard method of test for the k of refractories in the United States is of this type.[3]

To obtain adequate guarding for accurate determinations of k, quite large samples are required in this arrangement. Standard minimum dimensions recommended by the American Society for Testing Materials are given in Table 3.2.

Table 3.2 ASTM Standard Minimum Linear Surface Dimensions in Twin-plate Arrangement (Square or Round) for Thermal Conductivity Measurements

Sample Thickness, in.	Width of Main Plates, in.	Width of Guard Ring, in.
1	4	1.5
1.5	8	2.25
2	12	3
4	12	6

Radial flow through a hollow sphere or prolate spheroid is an attractive arrangement since no heat losses occur except through the electrical leads to the heater and thermocouples. But the forming of uniform heat sources and fabricating of hollow samples is frequently a difficult requirement of these shapes. Cylindrical arrangements lose heat at their ends. A technique to reduce axial heat flow in a cylinder to negligible proportions is to provide end heat guards (Fig. 3.2). A tentative method for determining the k of pipe insulation in the United States is of this type.[4] Another technique to reduce axial heat flow in a cylinder is to use a sample long compared with its diameter and to work only in the center section where the isothermal surfaces are essentially cylindrical.

Accurate measurement of thermal conductivity is difficult since it involves the determination of temperature and heat flow in a sample of known dimensions. The greatest difficulty in the measurement of k is obtaining heat flow in the sample such that the relation between A and x coincides exactly with that assumed in integrating eq. 1. However, thermal conductivity can be measured with equivalent results by various arrangements and by different investigators. This fact is indicated in the report by Robinson and Watson[26] on the determination of the k of cork by 17 different laboratories using linear flow arrangements. It is further indicated by the results of Adams[1] and of Kingery[17] on the determination of k for aluminum oxide using radial flow in a hollow prolate spheroid, sphere, and cylinder.

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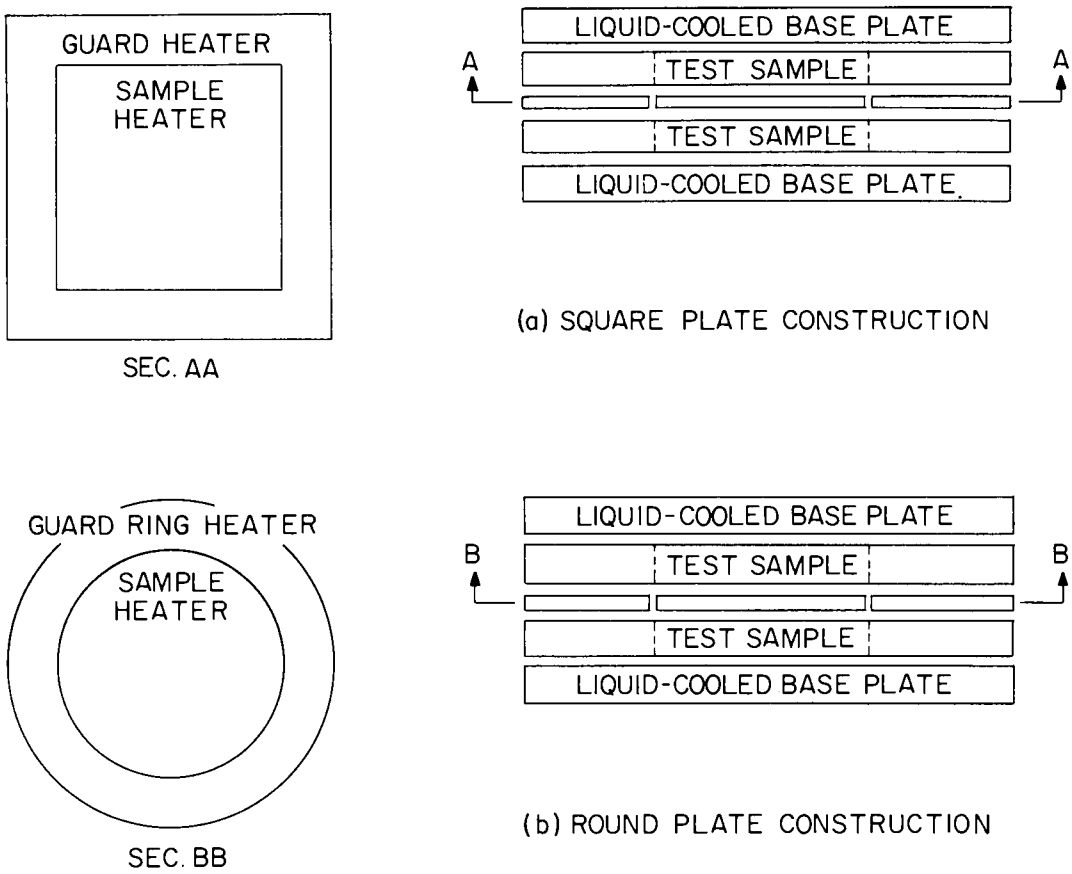

Fig. 3.1. Schematic diagrams of linear flow in flat plate apparatus for determining thermal conductivity illustrating heat guards.

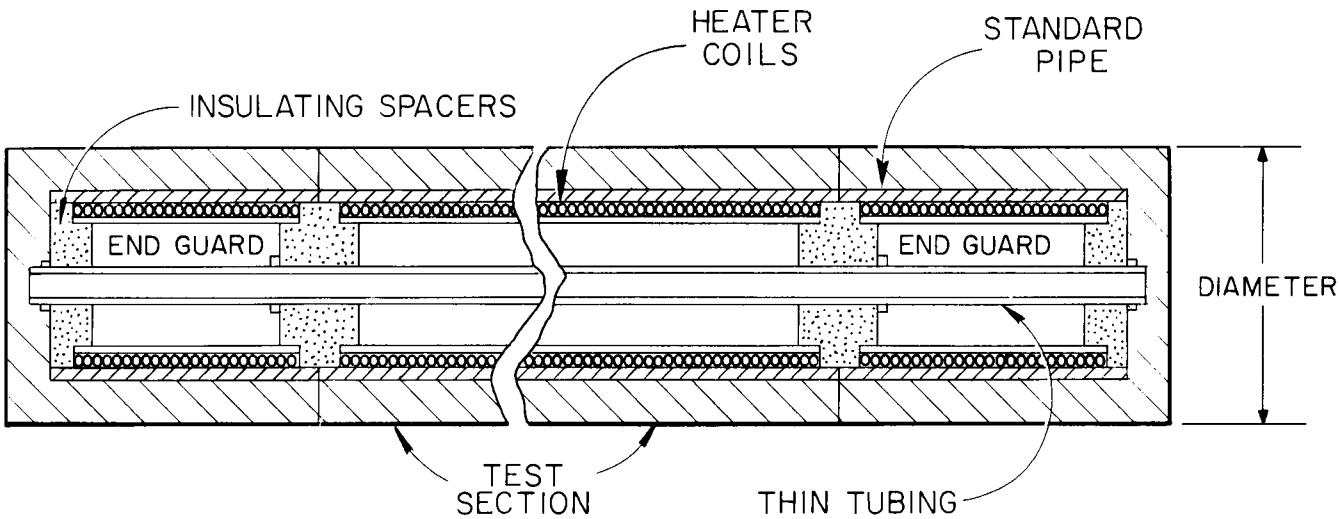

Fig. 3.2. Schematic diagram of radial flow in hollow-cylinder apparatus for determining thermal conductivity, illustrating end guard coils.

Recent surveys of the literature on the measurement of the thermal conductivity of solids have been published by Powell,[24] Cooper,[9] and Ross.[27]

Choice of Apparatus for Measuring Thermal Conductivity. As indicated above, thermal conductivity can be measured by various arrangements of apparatus. Each arrangement has certain limitations and the choice of one over another is governed by physical structure, temperature, and conductivity.

For example, powders, single crystals, pipe covering, refractory bricks, etc. require an arrangement of apparatus adapted to the material. The solids formed when acidic or neutralized wastes are calcined to about 750°C can be described as porous crystalline aggregates in which the solid phase is the continuous phase. Attempts to remove these solids from the container in which they are formed result in their crumbling. This crumbling causes the cellular material to become more like a granular material and suggests that the k of solid wastes should be determined in the container in which they are calcined.

Measurement of the thermal conductivity of copper at -250°C and at +250°C requires entirely different apparatus. Current Oak Ridge National Laboratory practice is to calcine at about 750°C; however, the temperature can be raised above 1000°C with present equipment. Higher temperatures may be advantageous, not only because the chemical stability of the product is better, but also because some sintering may improve the thermal properties of the product.

Pure metals, good conductors, require relatively simple apparatus since heat leakage will be small; but, refractories, poor conductors, require more elaborate equipment since care must be taken to reduce heat leakage. Since the solids formed by calcination of wastes to date have been mainly porous mixtures of metal oxides, they should be classified as poor conductors. Consequently, care must be taken to reduce heat losses in the measurement of the k of these solids.

From a consideration of the factors listed above, as well as the advantages and limitations of the various arrangements outlined, radial flow in a hollow-cylinder arrangement of apparatus has been chosen to measure the thermal conductivity of various wastes as a function of temperature and porosity. To reduce heat flow out the ends of the cylinder, a sample long compared with its diameter* will be used (Figs. 3.3 and 3.4).

The thermal conductivity is calculated by integrating eq. 1 for a cylinder of length L between the limits of radius R_1 at temperature T_1 and radius R_2 at temperatures T_2 to give

$$\bar{k} = \frac{q}{2\pi L(T_1-T_2)} \ln\left(\frac{R_2}{R_1}\right) \qquad (3)$$

* Waddams[32] has shown that the length/diameter ratio must be at least 4 to ensure essentially cylindrical isotherms at the center of a finite cylinder.

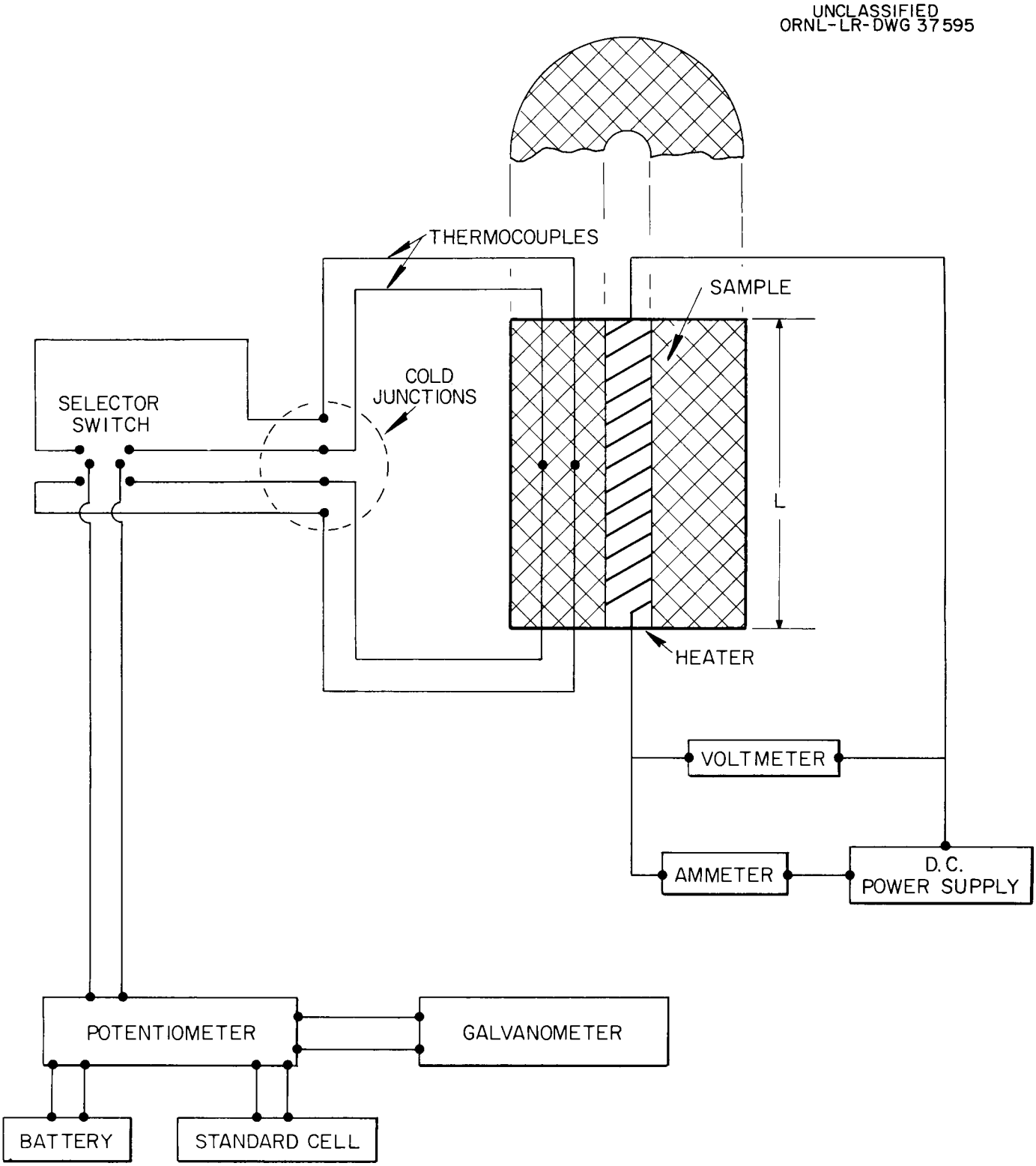

Fig. 3.3. Schematic of the arrangement of apparatus to measure the thermal conductivity of various solid wastes. L = length of hollow cylinder taken as test zone.

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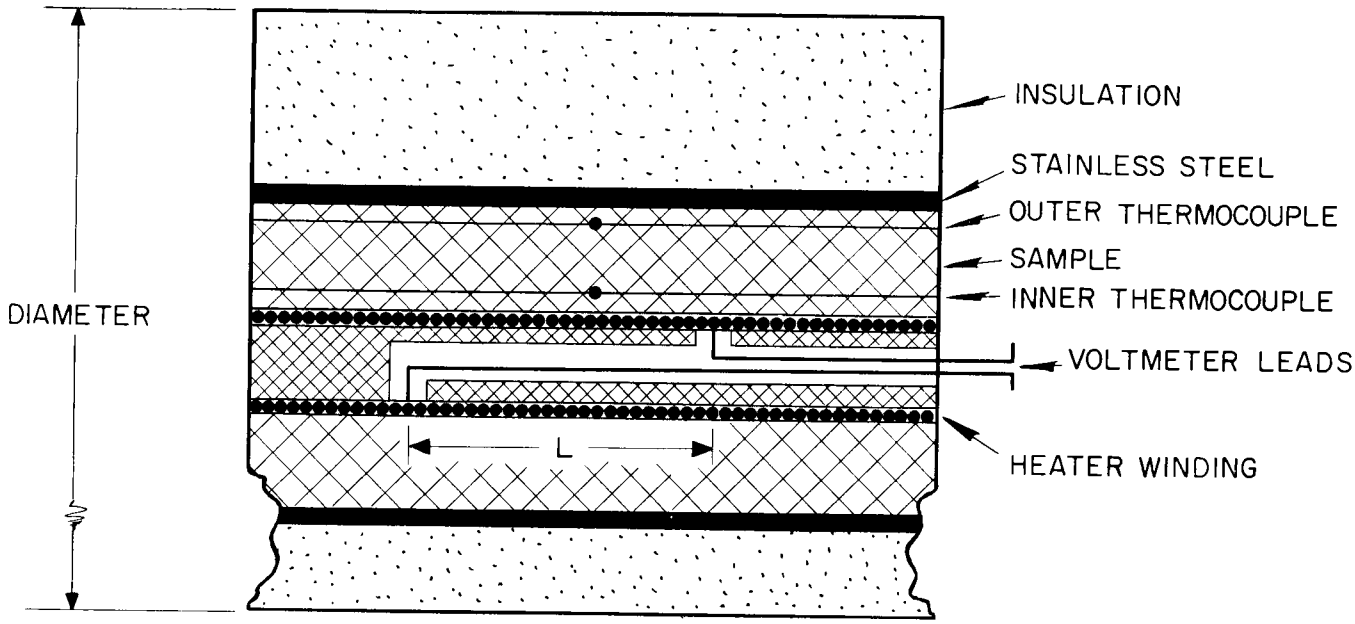

Fig. 3.4. Cross section of test specimen to measure the thermal conductivity of various solid wastes. L = length of test zone.

where $\bar{k}$ is the mean value of k over the temperature range T_1 to T_2. The mean thermal conductivity is defined by

$$\bar{k} = \frac{\int_{T_1}^{T_2} k \, dT}{T_2 - T_1} \tag{4}$$

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5.0 APPENDIX

5.1 Notations

Symbol	Quantity	Units
A	Area	ft^2
c	Specific heat	$Btu/lb \cdot {}^oF$
h	Surface conductance, defined by $h = q/A(T_{surface} - T_{gas})$	$Btu/hr \cdot ft^2 \cdot {}^oF$
k	Thermal conductivity	$Btu/hr \cdot ft \cdot {}^oF$
$\bar{k}$	Mean thermal conductivity over a temperature range	$Btu/hr \cdot ft \cdot {}^oF$
K	Thermal conductivity of cave or vault	$Btu/hr \cdot ft \cdot {}^oF$
q	Heat flow rate	Btu/hr
L	Length	ft
P	Porosity	
Q	Volumetric heat generation rate	$Btu/hr \cdot ft^3$
r	Radius of cave or vault	ft
R	Radius of waste package	ft
t	Decay (shutdown) time	sec
T	Temperature	${}^oF$, ${}^oR$
x,y,z	Space coordinates	ft
ΔT	Temperature difference	${}^oF$
ΔT_W	Temperature rise from center to surface of waste package	${}^oF$
ΔT_S	Temperature rise across space between waste package and wall of cave or vault	${}^oF$
ΔT_C	Temperature rise at the cave or vault wall	${}^oF$
V_s	Volume of solid phase	ml
V_p	Volume of porous body	ml

Notations (Cont'd)

Symbol	Quantity	Units
α	Thermal diffusivity, defined by $\alpha = k/c\rho$	ft^2/hr
θ	Time	hr
ρ	Density	lb/ft^3
ρ_s	Density of void-free solid (theoretical density)	g/ml
ρ_p	Density of porous body (apparent density)	g/ml
σ_f	Thermal neutron fission cross section	cm^2
τ	Irradiation time	sec
ϕ	Thermal neutron flux	$neutrons/cm^2 \cdot sec$

5.2 Calculation of the Volumetric Heat Generation Rate of a Calcined TBP-25 Waste

Assuming that a Materials Testing Reactor (MTR) fuel element is irradiated at a flux of 2×10^{14} neutrons/cm^2·sec to 35% burnup, that 0.11 gal of waste is formed per gram of uranium-235 processed,[20] and that the waste is reduced to 24% of its original volume on drying and calcining,[10] the volumetric heat generation rate of the calcined waste can be calculated from the relation

$$\% \text{ burnup} = 100\ (\phi \tau \sigma_f) \qquad (5)$$

and Blomeke's[8] curves for total fission product power as a function of irradiation time and shutdown time.

First, the irradiation time of the above fuel element is calculated from relation 5:

$$35 = 100\ (2 \times 10^{14})\ (\tau)\ (580 \times 10^{-24})$$

$$\tau = 3 \times 10^6 \text{ sec}$$

Second, for a decay time of 120 days (1×10^7 sec), the power as read from Blomeke's curve for $\phi = 1 \times 10^{14}$, $\tau = 3 \times 10^6$, and $t = 1 \times 10^7$ is doubled (since ϕ is 2×10^{14} in this case) to give a total fission product power of $2(2.1 \times 10^{-22})$ watt per original atom of uranium-235.

Third, using the values given above for volume of waste per original atom of uranium-235 and for volume reduction on calcining, the volumetric heat generation rate of the calcined waste is calculated to be 41.3 watts/gal or 1056 Btu/hr·ft^3.

Similar calculations for a decay time of one year ($t = 3 \times 10^7$ sec) gives a volumetric heat generation rate for the calcined waste of 8.73 watts/gal or 223 Btu/hr·ft^3.

5.3 Estimation of Thermal Conductivity of Calcined TBP-25 Waste

Waste from the TBP-25 process for recovering uranium from uranium-aluminum alloy fuel elements is primarily aluminum nitrate plus fission products. Assuming that a calcined TBP-25 waste is principally alumina (theoretical density 3.97 g/ml), that the density of the calcined product is 0.367 g/ml,[10] and that the data for corundum (native alumina) in Fig. 2.4 is representative of the alumina obtained in calcination, k_p at 800°C is estimated as follows:

First, the porosity of the waste is calculated from the density measurements above using the relation

$$P = 1 - \frac{V_s}{V_p} \approx 1 - \frac{\rho_p}{\rho_s} \qquad (6)$$

to give a porosity of 91%.

Second, the curve for corundum at 800°C in Fig. 2.4 is extrapolated to the thermal conductivity of air at 800°C (0.041 Btu/hr·ft·°F).

Third, the thermal conductivity at 91% porosity on the extrapolated curve is estimated to be 0.092 Btu/hr·ft·°F.

A value of 0.134 Btu/hr·ft·°F is obtained for k_p at 800°C by extrapolating the curve for corundum at 800°C to 0% porosity (k_s = 1.52 Btu/hr·ft·°F) and then using Russell's equation (Table 2.2) for a cellular material.

5.4 Order-of-Magnitude Values of Surface Conductance

Surface conductance depends on many factors, such as shape, orientation, dimensions, and condition of the surface; properties of the fluid; velocity of the fluid past the surface; and whether or not the fluid is changing phase. The following values are given in the literature:

	h, Btu/hr·ft^2·^{o}F
Air, natural convection	0.02 - 8
Polished surface, small ΔT	
Vertical	1.3
Horizontal (facing up)	1.7
Blackened surface, small ΔT	
Vertical	1.8
Horizontal (facing up)	2.5
Air, forced convection	2-50
Oil, flowing	10-300
Water, flowing	50-3000
Water, boiling	300-9000
Steam, condensing	1000-20000

5.5 Thermal Conductivities of Some Rocks, Soils, and Building Materials

Material	T, ^{o}F	K, Btu/hr·ft·^{o}F
Calcium carbonate, natural	86	1.3
Concrete, cinder mix	--	0.5
Concrete, sand, and gravel (average)	--	0.75
Concrete for dams	--	1.4
Dolomite	122	1.0
Granite	--	1.0 - 2.3
Limestone	--	0.5 - 0.9
Marble	--	1.2 - 1.7
Rock salt	122	3.5
Sand, medium fine quartz, dry	68	0.15
Soil, loosely packed clay	--	0.2
Heavily packed clay	--	0.8
Wet subsoil	--	1.3

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