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AN AUTOMATED THERMAL CONDUCTIVITY PROBE,
AND APPLICATIONS TO POWDERS *

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ABSTRACT

A thermal conductivity probe has been developed for measurements of powders and porous media from ambient temperature to 1300 K in vacuum or an inert-gas atmosphere. Automated data-acquisition and graphical analysis programs have been developed in FORTRAN for use with a laboratory mini-computer. Nonlinear data-analysis techniques are available which can account for the thermal-probe response due to contact resistance and probe thermal-mass effects. Programs have also been developed with terminal BASIC for use with a small, portable desktop computer system. Examples of the use of the probe and data acquisition and analysis system are shown for measurement of the effective thermal conductivity of ceramic and metal powders.

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INTRODUCTION

The transient thermal conductivity probe technique generally allows more rapid determination of the conductivity of materials than steady state methods. In addition, the probe method usually requires a smaller sampling volume, less measurement equipment, and is more suitable for nondestructive in situ measurements. As a result, the probe technique is often employed for use as a portable thermal conductivity measurement device for remote applications. However, there is generally a lack of portable automated instrumentation for the acquisition and immediate analysis of probe data.

In this report, we describe a thermal conductivity probe measurement system which has been used for the measurement of powdered materials. The probe designed for this work has been used from ambient temperature to 1300 K in both vacuum and inert gas atmospheres. The data acquisition and analysis system uses a desktop microcomputer, which allows the entire system to operate as a portable, stand-alone instrument. Applications of the probe system are provided which show measurements of the thermal conductivity of ceramic and metal powders.

ANALYTICAL BACKGROUND

The reader may consult additional references for analytical background in the measurement technique [1-3]. Basically, the thermal conductivity probe is based on the transient line source technique, where heat is supplied at a constant rate to the test material along the axis in a cylindrical geometry. The temperature response of the probe as a function of time is used to determine the effective thermal conductivity of the test material.

We have used two analytical methods to analyze probe data in this laboratory. An integral solution, due to Jaeger [4], describes the probe

temperature rise as a function of time through the entire time period of the probe response. This solution includes the effects of the thermal mass of the probe and sample material, thermal contact resistance at the probe-sample interface, and the finite dimensions of the probe; an infinite probe thermal conductivity is assumed. Our implementation of this solution follows the work of Koski and McVey [2], which is useful when only short time data are available.

The second solution which has been employed provides a simpler, but approximate, analytical method. It has been shown [4] that after an initial time period during which probe thermal mass and contact resistance effects are important, the probe temperature rise is linear with $\ln$ time. The slope of the temperature rise is then proportional to the probe heater input power and inversely proportional to the effective thermal conductivity of the material. When the linear temperature rise is fully developed, this so-called straight line method is readily employed as the basis for probe data analysis techniques.

EXPERIMENTAL DETAILS

A. High Temperature Constantan Probes

Thermal conductivity probes incorporated constantan heater wire for use to temperatures approaching 1300 K with minimum electrical resistance change with temperature. Such a probe is depicted in Fig. 1. The constantan heater comprises a dual chromel-constantan type E replacement thermocouple with the two junctions grounded to the stainless steel external sheath [5] (e.g., Omega Engineering, Inc., part number Dual SCXSS-040G RP). The constantan legs of the two thermocouples are used to provide a continuous constantan heater element. The heater leads are surrounded by a refractory insulation inside the 1.0 mm (0.040 in.) O.D. 304 stainless steel sheath.

Depending on the application, probe heaters of lengths from 30 to 66 cm (12 to 26 in.) have been constructed in our laboratory.

The temperature sensor consists of a type K chromel-alumel thermocouple in a 0.25 mm (0.010 in.) stainless steel sheath (e.g. Omega P/N SCASS-010G), welded to the probe heater using stainless steel tabs, as shown in the figure. These probes typically exhibit resistance of the order of 0.4 ohms per cm of length. The figure shows the temperature sensor junction attached 3.8 cm (1.5 in.) from the heater end, for use on axis of a 75 mm (3 in.) cylindrical crucible. In this application, the length to diameter ratio of the overall probe is approximately 65.

We have successfully used the high temperature constantan probes in inert gas environments to 1300 K. The probe is typically placed on the axis of ceramic cylindrical crucibles, 75 mm long x 35 mm inside diameter. Once filled, the crucible and probe are placed in a high temperature tube furnace, typically 40 cm in length and 7 cm I.D. The constantan probe extends beyond the sample material and crucible and outside of the furnace. A correction is applied which accounts for the variation in heater resistance with temperature, due to the temperature gradient along the probe [6].

B. Instrumentation

A diagram of the portable thermal conductivity probe system is shown in Fig. 2. The hardware components comprise the Hewlett-Packard HP-85 Personal Computer [5], the HP 3497A Data Acquisition/Control Unit (referred to as a DVM/scanner), an HP 6266B Constant Voltage DC Power Supply, a probe switching system of our own design, and a thermal conductivity probe. The entire system requires not more than 56 cm (22 in.) of vertical space in a standard electronics rack.

The constant voltage DC power supply provides a source of power to the resistive load selected by the probe switching system. The probe switching system provides a means of switching DC power to the probe, either manually or by an external contact closure, in this case provided by computer control from the 3497A. When the probe is switched off, power is supplied to a dummy load whose resistance is matched to the probe; this allows power supply stabilization at the appropriate load.

The HP 3497A DVM/Scanner is used for analog input, analog-to-digital conversion, and digital output. Hardware thermocouple compensation is provided by an external electronic ice point reference. The probe thermocouple voltage, probe heater voltage, and probe heater current are measured on three analog input scanner channels. Analog-to-digital conversion in the 3497A is accomplished by the 5 1/2 digit DVM, with 1 microvolt resolution. The DVM/scanner and HP-85 are connected by the HPIB interface bus. The contact closure necessary for operation of the probe switching system is provided by a digital actuator card in the 3497A.

DATA ACQUISITION AND ANALYSIS SYSTEMS

We have developed two independent data acquisition and analysis systems for use with the thermal conductivity probe measurement device. In our laboratory, a Hewlett-Packard HP 1000 minicomputer system is used for data acquisition, graphical data analysis, and nonlinear data fitting of thermal conductivity probe data. For this computer, software was developed in FORTRAN. This minicomputer has sufficient computational size and speed to allow the use of the Jaeger integral solution for data simulation and nonlinear parameter estimation techniques. Software documentation and examples are provided in Ref. [7].

For a portable, stand-alone probe system, data acquisition and analysis software was developed in BASIC for use with the system depicted in Fig. 2, incorporating the straight line analysis method. The HP-85 Personal Computer is used for data acquisition, device control, data storage and retrieval, graphical data analysis, and data presentation. The HP-85 microcomputer consists of the HP-85A Personal Computer, with keyboard, CRT display, cassette magnetic tape, an integral thermal printer, 32K memory, and an HP-IB interface (IEEE-488). Additional hardware and software documentation may be found in Ref. [6].

The software developed for the HP-85 for thermal conductivity probe data acquisition and analysis is indicated in Fig. 3. The program uses the special function keys on the HP-85 to direct the operator to the various functional sections of the program. These functional sections include data acquisition (ACQUIRE), graphical data analysis (ANALYZE), and visual readout of the instrumentation parameters (VIEW).

Once loaded and initialized, the program waits for operator selection of one of the desired functions. During this period, the system periodically samples the probe temperature in order to determine the linear baseline temperature drift of the probe. This information is available to the operator with the VIEW function once sufficient data are obtained for a least squares baseline determination. VIEW also provides the operator with system parameter information such as time, probe temperature, voltage, current, and resistance of the load (either the dummy load or the resistive probe heater).

The ACQUIRE section is used for probe data acquisition. The operator may optionally specify a file name for data storage on cassette tape following data collection. Once acquisition has begun, the system switches power from the dummy load to the probe heater, and readings of the time and probe

temperature are recorded. The power (current and voltage) applied to the probe is also recorded. Acquisition is terminated by operator intervention or after a specified run duration. The applied power is then switched back to the dummy load. The program either returns to the function selection menu or proceeds to the analysis section, depending on the software configuration.

In ANALYZE, acquired data are obtained from storage on tape or in memory for subsequent graphical data analysis using the straight line analysis method. The data are plotted on the CRT screen as temperature vs ln time. A cursor is drawn on the screen, which may be moved with the special function keys. The operator selects the linear region, a least squares fit is made to the data, and the thermal conductivity is computed.

APPLICATIONS

A. Ceramic Microspheres

A series of measurements were made on the thermal conductivity of alumina microspheres in nitrogen and argon from 25 to 650 C. The microspheres were originally supplied by General Atomic Company to Oak Ridge National Laboratory (ORNL), where the conductivity was measured in 1 atm. nitrogen using the steady-state radial heat flow technique [8]. Subsequently, samples of the microspheres were measured at AERE Harwell by Hall [9], again at 1 atm. nitrogen. McElroy at ORNL supplied our laboratory with samples as well; our results are described in this section.

The alumina (98.9% pure) microspheres ranged in diameter from 495 to 505 micrometers. Measurements were made in argon and nitrogen, for the most part at 670 Torr (1 psig, Albuquerque). The tap density of the microspheres was approximately 2.35 g/cm^3 for these measurements, or about 60% theoretical density.

The probe response (temperature rise vs $\ln$ time) for the microspheres in nitrogen at room temperature is shown in Fig. 4 as a function of gas pressure. The linear regions used in the analysis are bounded by the vertical dashed lines. As the gas pressure is reduced, the effective conductivity of the material decreases, and the probe-material contact resistance increases. The conductivity values are as follows: 0.325 W/m-K at 670 Torr nitrogen, 0.169 at 10 T, 0.066 at 1 T, and 0.031 at 0.1 T. As the contact resistance increases, the time at which the linear region begins (the inflection in the curve) is delayed, as may be observed by inspection of the arrows in the figure. The observed reduction in effective conductivity with decreasing gas pressure is typical of measurements on powder-gas systems [10], and is due to mean free path effects. Prior to the inflection point; a "false" straight line region is evident, particularly at the lower pressures.

Fig. 5 shows the results of measurements on the microspheres at this and other laboratories. The curves were determined from quadratic polynomial fits to the data points using a least squares technique. For the ORNL and Harwell fits, a limited amount of data was available (4 and 8 points, respectively). Throughout the reported range of measurement, the Harwell data are 6-7 % higher than the ORNL values. For an MgO powder, Moore, et al. [10] reported results from the ORNL apparatus to be 20% lower than previous measurements. Our data are in reasonable agreement with the Harwell data to about 400 C. At 600 C, the difference between the results is about 9 %, which suggests some systematic error at the higher temperatures between the different measurement devices. The fitted conductivity data from the probe technique is described by:

$$k \text{ (W/m-K)} = 0.297 + (5.739 \times 10^4) \times T - (1.0714 \times 10^7) \times T^2,$$

$$(\text{rms deviation} = 0.020 \text{ W/m-K})$$

for T in degrees C. This fit yields a conductivity at ambient temperature (T=25 C) of 0.311 W/m-K in 1 atm. nitrogen. For argon at 670 Torr, the the fitted conductivity values give

$$k \text{ (W/m-K)} = 0.200 + (4.80 \times 10^4) \times T - (2.073 \times 10^7) \times T^2,$$

$$(\text{rms deviation} = 0.007 \text{ W/m-K}).$$

This fit yields a conductivity at room temperature of 0.211 W/m-K.

B. Metal Powders

An example of the probe response in metal powders is shown in Fig. 6. These measurements on Te powders were done at room temperature in air. The solid curve shows the temperature rise vs ln time (in seconds), and the characteristic inflection in the curve (change in slope) near $\ln t = 4$.

Curve S represents a powder of 5 - 20 micrometer spherical Te particles; the packing fraction was 59% (41% void); the measured conductivity was 0.135 W/m-K. Curve A represents a Te powder of 1-100 micrometer irregularly-shaped particles, 50% void fraction, and a conductivity of 0.115 W/m-K. Generally, we would expect the higher density powder to have a higher conductivity, as was measured.

We have also made measurements on a powder of HP-9-4-20 stainless steel spherical particles, 10 - 150 micrometer diameter, in ambient temperature air. The void fraction was 0.34; the thermal conductivity was 0.390 W/m-K. A 50 w/o mixture of the spherical Te in the steel gave a powder with a density of 4.63 g/cm^3 and a conductivity of 0.265 W/m-K. These results are summarized in the table.

Table. Thermal Conductivity of Metal Powders by the Probe Method, Ambient Temperature, Air Atmosphere.

Material	Particle Diameter (micrometer)	Powder Density (g/cm ³)	Void Fraction	Thermal Conductivity (W/m-K)
Te, spherical	5 - 20	3.67	0.41	0.135
Stainless steel, HP-9-4-20, spherical	10 - 150	5.20	0.34	0.390
Mixture, steel + Te, 50 w/o	5 - 150	4.63	-----	0.265

SUMMARY

A thermal conductivity probe measurement system was described which uses a probe capable of operation to 1300 K and a portable, stand-alone data acquisition and analysis system. Device software was developed to allow automated acquisition and conductivity analysis of thermal probe data by the straight line method. Measurements on ceramic and metal powders were presented which demonstrate the use of the system.

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REFERENCES

1. W. D. Drotning, J. A. Koski, and P. E. Havey, "Development of a Technique for Measurement of the Thermal Conductivity of Powders at High Temperatures," SAND81-0631, Sandia National Laboratories, Albuquerque, NM, August, 1981.*
2. J. A. Koski and D. F. McVey, "Application of Parameter Estimation Techniques to Thermal Conductivity Probe Data Reduction," in Thermal Conductivity 17, edit by J. G. Hust, Proc. of the 17th Inter. Thermal Conductivity Conference, June 15-18, 1981, Gaithersburg, MD (Plenum Press, NY, 1983), p. 587.
3. A. W. Pratt, "Heat Transmission in Low Conductivity Materials," in R. P. Tye, Thermal Conductivity, Vol. 1, Academic Press, NY, 1969.
4. J. C. Jaeger, Aust. J. Phys. 9, 167 (1956).
5. Reference to a particular product or company implies neither a recommendation nor an endorsement by Sandia National Laboratories or the U. S. Department of Energy, nor a lack of suitable substitutes.
6. W. Drotning and T. Tormey, "Thermal Conductivity Probe Measurement System," SAND83-2057, Sandia National Laboratories, Albuquerque, NM (to be published).*
7. J. A. Koski, "CONFIT: A Computer Code for Thermal Conductivity Probe Data Reduction With the Use of Parameter Estimation Techniques," SAND82-0741, Sandia National Laboratories, Albuquerque, NM, May, 1982.*

8. D. L. McElroy, Oak Ridge National Laboratory, personal communication.

9. R. O. A. Hall, AERE Harwell, personal communication.

10. J. P. Moore, R. J. Dippenaar, R. O. A. Hall, and D. L. McElroy,

"Thermal Conductivity of Powders with UO_2 or ThO_2 Microspheres in Various Gases from 300 to 1300 K," ORNL/TM-8196, Oak Ridge National Laboratory, Oak Ridge, TN, June, 1982.

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CONSTANTAN PROBE

1. Construction detail of high temperature constantan probe.

DATA ACQUISITION & CONTROL SYSTEM

2. Block diagram of thermal conductivity probe measurement system.

3. Block diagram of thermal conductivity probe data acquisition and analysis software, based on the HP-85 microcomputer.

4. Probe response curves for alumina microspheres at ambient temperature at various nitrogen pressures. The linear region used in the data analysis is bounded by vertical dashed lines. The arrows mark the inflection region, where the slope of the response curve changes.

5. Thermal conductivity of ORNL alumina microspheres vs temperature in nitrogen and argon at 670 Torr. Curves represent quadratic fits to the data (this work, at SNLA, probe method, as well as ORNL and Harwell data by the radial heat flow method).

6. Thermal conductivity probe response for two Te metal powders.