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A COMPUTER-CONTROLLED APPARATUS FOR THERMAL CONDUCTIVITY MEASUREMENT BY THE TRANSIENT HOT WIRE METHOD

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Abstract

The aim of this paper is to review the transient hot wire method for measurement of thermal conductivity, which is based on the measurement of temporal history of the temperature rise caused by linear heat source (hot wire) embedded in a test material. If a current is passed through the wire, the rise in temperature will be dependent, among other factors, on the thermal conductivity of the medium, surrounding the wire. Here the mathematical basis, as well as main modifications of the hot wire method – cross technique, resistance modifications with potential and compensated lead methods; hot wire probe method and parallel wire technique, are described and discussed. A fully automated computer-controlled transient hot wire apparatus is presented and tested, which allows measurement of thermal conductivity of solid, powder and granular materials at high temperatures.

Keywords: conductivity, probe method, thermal, transient hot wire method

Introduction

The technological development, that has occurred over past decade, has generated an urgent need for information on the thermophysical properties of matter over a wide range of specific environmental conditions – at elevated temperatures, high pressures, under high electric and magnetic fields, etc. Thermal conductivity is one of such properties. The amount of available data is very poor and even when data exist, comparison between data from different laboratories, and/or obtained by using different techniques shows huge scattering. This property is one of the most difficult to deal with owing to several problems, that arise not only because of deviations from the mathematical model (which is the basis of the experimental method), but also because of the material reactivity and temperature measurement in aggressive environments.

Several original methods for measurement of thermal conductivity, have appeared and have been analyzed in literature so far [1]. One of the most popular is the transient hot wire method [2, 3], which has become established as the primary method for accurate determination of thermal conductivity of liquids and

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gases. The method has been also successfully applied on measurement of solids–ceramics, rocks, glass and various domains such as porous materials, fine granular materials, soils, biological materials, etc. The method has been given standard status in many countries [4].

Principle of the hot wire method

The hot wire method is a transient dynamic technique based on the measurement of the temperature rise in a defined distance from a linear heat source embedded in the test material. If the heat source is assumed to have a constant and uniform output along the length of test piece, the thermal conductivity can be derived directly from the resulting change in the temperature over a known time interval [5].

The ideal mathematical model is based on the assumption of the hot wire to be an ideal, infinite thin and long line heat source, which is in an infinite surrounding from homogeneous and isotropic material with constant initial temperature T_o. If q is the constant quantity of heat production per unit time and per unit length of the heating wire (W m^{-1}), initiated at time $t = 0$, the radial heat flow around the wire occurs, and the temperature rise $\Delta T(r, t)$ at radial position r from the heat source conforms to the equation [6]

$$\Delta T(r,t) = T(r,t) - T_o = -\frac{q}{4\pi k} Ei\left(-\frac{r^2}{4at}\right). \tag{1}$$

Here k is the thermal conductivity (W m^{-1} K^{-1}), a thermal diffusivity (m^2 s^{-1}), ($a = k/\rho c_p$, with ρ is the density (kg m^{-3}) and c_p the isobaric heat capacity (J kg^{-1} K^{-1})) of the test material and $Ei(x)$ is exponential integral

$$-Ei(-x) = \int_x^\infty \frac{e^{-u}}{u}\, du \tag{2}$$

If the expression $r^2/4at < 1$ is fulfilled, i.e. for a sufficiently long time t and a small distance r, then using of the first term approximation of the exponential integral $Ei(x)$ one gets from Eq. (1) the simplified formula

$$\Delta T(r,t) = \frac{q}{4\pi k} \ln \frac{4at}{r^2 C} \tag{3}$$

where $C = \exp(\gamma)$, $\gamma = 0{,}5772157..$ is the Euler's constant. Thus the measurement of temperature rise $\Delta T(r, t)$ as a function of time may be employed to determine of the thermal conductivity k, calculating of the slope K of the linear

portion of temperature rise $\Delta T(r, t)$ *vs.* natural logarithm of the time ln$t$ evolution from

$$k = \frac{q}{4\pi K} \tag{4}$$

Equation (3) is valid for a wire of infinite length embedded in a sample of infinite dimensions above a certain minimum time t_{min}. The minimum time depends on the diameter of the wire, how it is embedded in the sample, and on thermal properties of both materials – heating wire and sample. However, since every laboratory sample necessarily has finite dimensions and since the heating caused by the heating wire will be perceivable after some time on the sample surface, the time range within which the temperature rise *vs.* time corresponds to Eq. (3) is also limited. There exists a time t_{max} which, when exceeded, the heat transfer, or the heat accumulation at the radial sample surface results in strong deviations from the relationship given by Eq. (3) (Fig. 1).

The maximal suitable time t_{max} can be defined as the time, in which temperature rise at radial position r_s (r_s is the specimen radius, or the mean distance between heating wire and radial sample surface in the case of noncircular cylindrical specimens) is equal to 1% of the temperature rise in the measured position r (in the case of direct measurement of temperature rise of hot wire r is equal to wire radius r_w). The time t_{max} can be detected also experimentally, as the time of the onset of a convex curvature of the linear dependence of $\Delta T(r, t)$ *vs.* ln$t$. As compared with the error of slope, produced by heat transfer from the radial sample surface, the error of slope due an axial heat flux, or heat accumulation on the axial sample surface at the time t_{max}, is mostly negligibly small under the conditions considered. At specific experimental conditions can be used

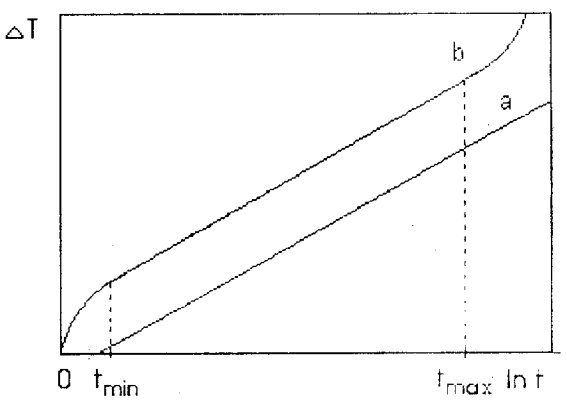

Fig. 1 Temperature rise *vs.* natural logarithm of time dependence (a – theoretical curve corresponding to Eq. (3), b – typical experimental curve)

the theory of eliminating of this influence, taken from the papers, dealing with in more detail [7–9].

If a measurement according to the principle described is to be useful, therefore it has to be necessary to calculate, or at least estimate, the times t_{min} and t_{max}, in order to define the minimum sample required, the corresponding dimensions of the furnace chamber, the heating wire diameter, and the manner of embedding of the wire in the sample.

Theoretically, on the basis mentioned above, thermal diffusivity may be obtained from the ordinate intercept of the least squares fit. In fact, because of the relatively high dependence of this parameter on satisfying of the ideal experimental conditions assumed, the method has been world-wide used mostly only for thermal conductivity determination.

Experimental techniques

Main experimental methods based on this mathematical model can be classified as follows:

In the **standard (cross) technique** between two equally sized specimens a wire cross is embedded in ground grooves [10]. The cross consists of a heating wire and the legs of a thermocouple, which acts as the temperature sensor. The hot spot of the thermocouple is in direct contact with the heating wire. The cold junction is put on the reference place. The main advantage of this arrangement is relatively good sensitivity and possibility of measurement in wide range of temperature. Problems of local temperature measurement can arise if nonhomogeneous materials have to be measured. If the hot wire is heated with DC (as usual), errors can occur, which are caused by an asymmetrical arrangement (difficult to avoid) of both thermocouple legs and the hot wire. This results a decrease, or increase, in thermoelectric voltage, depending on polarity. This effect can be diminished either by reversing of the polarity of DC voltage and taking as the correct value the mean of the original and reversed measurement, or either by using a relatively thick hot wire. When using of an AC source the problem mentioned does not occur.

In the **resistance technique** the heating wire acts also as the temperature sensor. Here the temperature is measured by the change in resistance caused by the heating-up of the hot wire. An advantage is that the mean temperature of the wire is measured along its total, or particular length. The measurement of the mean temperature of the wire eliminates the influence of local non-homogeneities of measured material. The problem of this technique is, that the theory considers an infinite line source in an infinite medium, while in practice one takes a finite length of wire in the sample of finite length and attempts to realize the

boundary conditions, that both wire and sample are infinite. There are in principle two different ways to eliminate this problem. In the potential lead method a four-terminal measurement of resistance is employed, simple by measuring of the voltage drop across a known length of platinum wire – directly with a suitable high sensitive microvoltmeter. In this case a single wire is used and the terminal effects are avoid by using of two potential leads at suitable distance from the ends of the wire. These leads consist of a wire even thinner than that used as a linear source heater, in order the temperature rise of the hot wire is said to be very little influenced by this connection [11]. The compensated lead method uses two wires (samples) of different lengths. To substract the end effects they are incorporated in the arms of a bridge. The out-of balance voltage of the bridge during a transient run is recorded with a digital microvoltmeter [12], or a computer controlled automatic bridge can used [13]. Despite of the difficulty encountered in measurement of the temperature through the resistance variation (sensitivity to temperature differences is more times less than by using of the thermocouples), compensated lead method is very useful for measurement of thermal conductivity of fluids and gases.

In the measurement of electrically conducting materials is necessary to insulate the heating wire and thermocouple wires, or potential leads, respectively. This is possible either by making of a nonconducting coating on these wires, or to enclose the heater and temperature sensor in a thin sheath or needle, which is inserted into the test material, respectively. Second one is so called probe modification of the hot wire method [14], which has been intended basically also for measurement of nonrigid materials, mainly for powders, granular, or fibrous materials, etc. Electrical insulation of the wires creates a thermal barrier between the heating wire, temperature sensor and measured material. Also finite heat capacity and thermal conductivity of heater, mainly in the case of using the probe method, results in difficulties in obtaining a linear section on the temperature rise *vs.* natural logarithm of time curve, because the satisfying of the restriction $r^2/4at << 1$ increases the time t_{min}. The way how to eliminate this problem is either in increasing of the time of measurement (t_{max}), joined with necessary enlarging of the sample dimensions, or in using of any more suitable mathematical model. The most frequently used expression for the temperature rise of the probe (non-ideal heating wire) [1], received as a solution of composite-cylinder model, which considers probe thermal properties and dimensions and thermal contact resistance between the probe and measured medium, can be written as

$$\Delta T(t) = \frac{q}{4\pi k}\left(\ln t + A + \frac{B \ln t + C}{t}\right) \tag{5}$$

where A, B and C are constants [14, 15].

Hot wire **parallel technique** is based on the measurement of temperature rise evolution in a defined distance from heating wire (about 15–20 mm). Since the measurement of temperature is performed by a thermocouple or thermistor, placed separately–parallel to the hot wire, there are no disturbances at the start of measurement and the time t_{min}, from which the calculation is possible, decreases. This allows to increase the conductivity measurement limit from 2 W m^{-1} K^{-1} for standard hot wire techniques up to 25 W m^{-1} K^{-1} in parallel technique.

Measuring system

The computer controlled hot wire apparatus was developed, which allows the determination of thermal conductivity of solids, powders and granular materials. Three measuring techniques are there available: standard cross wire technique, resistance potential lead method and the probe modification of hot wire method.

In cross wire technique as the linear heat source acts the platinum wire 0.35 mm in diameter (Heraeus), or kanthal wire 0.4 mm in diameter (Bulten Kanthal AB), used depending on the temperature range of measurement. The temperature rise *vs.* time evolution is measured by the spot welded K type thermocouple, made from Ni–NiCr wires (Heraeus) 0.2 mm in diameter, or S type – PtRh 90/10% 0.1 mm (Heraeus). The hot spot of the thermocouple is in direct contact with the heating wire and is placed in the center of sample. The reference – cold junction is immersed in Dewar cup at 0°C. In resistance technique the platinum wire 0.35 mm in diameter as the heating wire and also as the temperature sensor is used. Potential leads consist of platinum wires 0.1 mm in diameter, fixed to the heating wire at about 1.5 cm from the end of the sample. The probe method is based on the very simple cylindrical probe original construction, which consist of a heating wire and the temperature sensor, both placed in a ceramic microcapillar (Degussa) 12 cm long and 2 mm in diameter. Heating wire is located in the first hole and temperature sensor – spot welded thermocouple is in the second one, in order the measurement point is being near the center of the probe (Fig. 2). The wires used are the same as described in cross technique.

If an solid material has to be measured, the sample has usually the shape of a cylinder 10 cm in length and with diameter depending on thermal properties

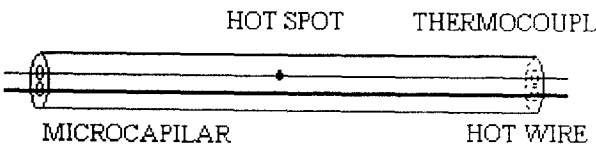

Fig. 2 The schematic diagram of the probe

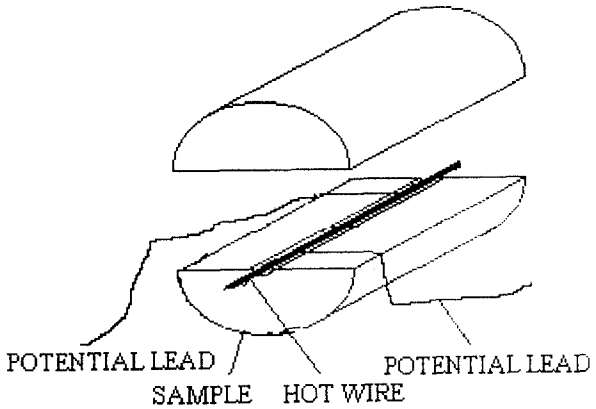

Fig. 3 Sample and wires arrangement in resistance technique (potential lead method)

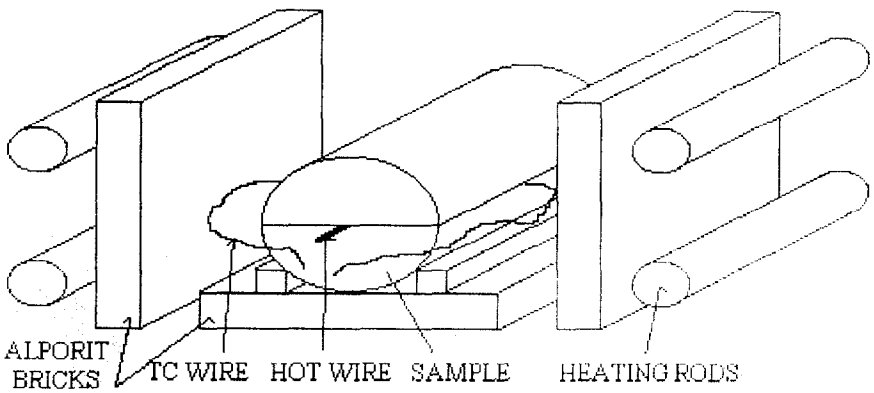

Fig. 4 The schematic diagram of the furnace with sample

of measured material (4–10 cm), cut into two parts along the axes. Sample is placed in the furnace in horizontal position, so that the bottom half one carrying the grooves for wires (or probe) is placed in first. Then are the wires put in position (Fig. 3). To ensure good embedding loose finely ground test material before placing the upper half cylinder. Here the use a cylindrical specimen rather than two bricks is preferred, because the decrease of a time required to receive a thermal equilibrium. Powders and granular materials are held in an iron container, with appropriate dimensions, so that wires or probe is crossing through the center of the container, perpendicular to the surface. The sample is heated by electro-resistance furnace, where are used as heating elements four silite rods and as the thermal insulator alporite (Fig. 4). This arrangement allows measurement on air, or in reducing environment, under atmospheric pressure, in the temperature range from room temperature up to 1200°C.

Figure 5 shows a block diagram of the instrumentation. The current flowing through the heating wire is produced by the stabilized regulated direct current supply Z-YE-2T-X (Mesit) with unit of remote control JDR-1 (Mesit). The setting of the optimal current depends mainly on sample thermal properties and dimensions and is chosen in order the temperature rise to be between 5–15°C. Instrumentation for measurement of temperature is also relatively standard. Digital microvoltmeter M1T 330 (Metra) with lockin pre-amplifier Z-35 (Metra) and compensator is used for serial measurements of transient *emf* of the thermocouple, or transient voltage corresponding to temperature rise. For the furnace temperature regulation a proportional feedback temperature controller was incorporated into this apparatus. The temperature control consists by first on reading of the actual temperature of the sample (furnace) via thermocouple located at (near) the sample surface. Then by using of the heater power controller, based on thyristor-regulated AC [17], the appropriate power flowing into the furnace is set on. All data acquisition and instrument control are performed using the PC (AT-486), via the IEEE-488 interface, despite of the heater power controller, based on programmable counter 8253 joined directly with the PC bus.

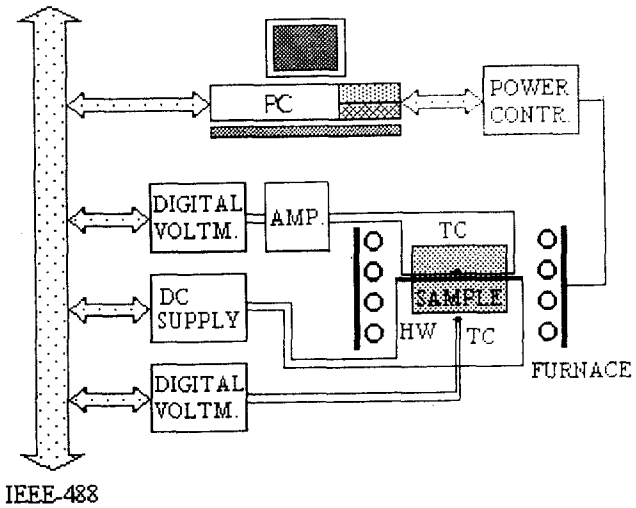

Fig. 5 Block diagram of instrumentation arrangement

For this apparatus a software package was written, which allows device control, data acquisition and data analysis. Measurement cycle begins with a temperature control routine, which was written to control the initial temperature of the sample. This routine is based on a discrete sampling proportional-integral (PI) algorithm. After a time, required to receive a thermal equilibrium in the sample at desired temperature (necessary time is usually between 40–120 min),

a data acquisition task performs the data acquisition. When executed, it first switches on the DC current flowing through hot wire, and then routine reads the transient voltage from the temperature sensor. Because the relatively long time of measurement (in the case of large samples 100–800 s) the data acquisition sections contains temperature control routine, running on background, too. The data acquisition rate, controlled with the use of PC's system counter, depends on the time, elapsed from the start of measurement, and decreases from 20 Hz to 0.5 Hz. Before saving of the data on the disk (for later use) or analyzing, the change in temperature is calculated for each experimental point, by using of an appropriated temperature *vs.* thermocouple *emf*, or *vs.* platinum resistance formula. The data reduction consists in performing a least-squares fit of the recorded temperature *vs.* time data in accordance with Eqs (3), (5) and then the thermal conductivity is determined from Eq. (4). The times t_{min} and t_{max}, calculated with respect of the theory mentioned above, can be set manually, or find by an interactive searching of the linear part of the curve ΔT *vs.* ln$t$ with the use of zoom routine.

Here the recorded experimental data are compared with their fit in various screen scale. A routine for automatic searching of a suitable time interval taken into account is also available. It is based either on an algorithm, which uses first and second order time derivatives for detecting of the beginning of curvatures of experimental curve [18], or on an another iterative procedure based on residual dispersion [19], respectively. The received values of thermal conductivity are then printed or saved (in ASCII form) for using them with any another soft-

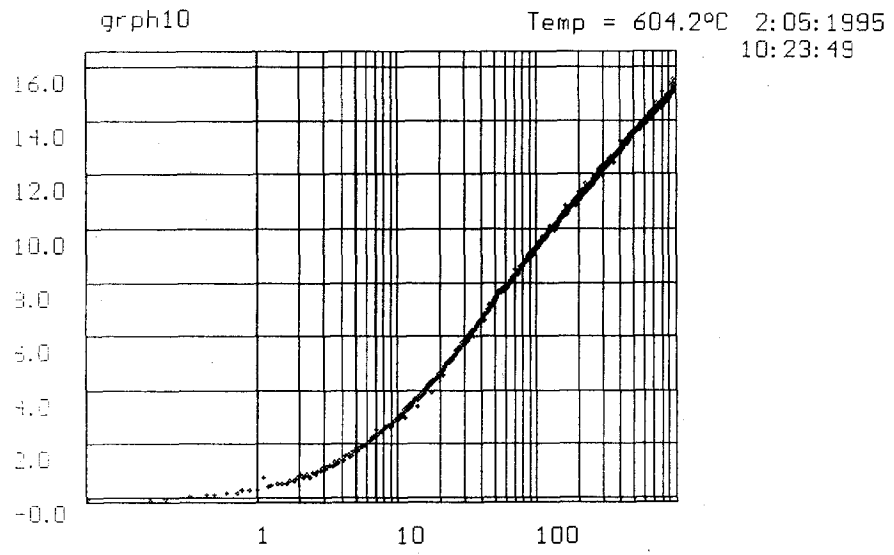

Fig. 6 Typical experimental temperature rise *vs.* natural logarithm of time dependence in probe technique

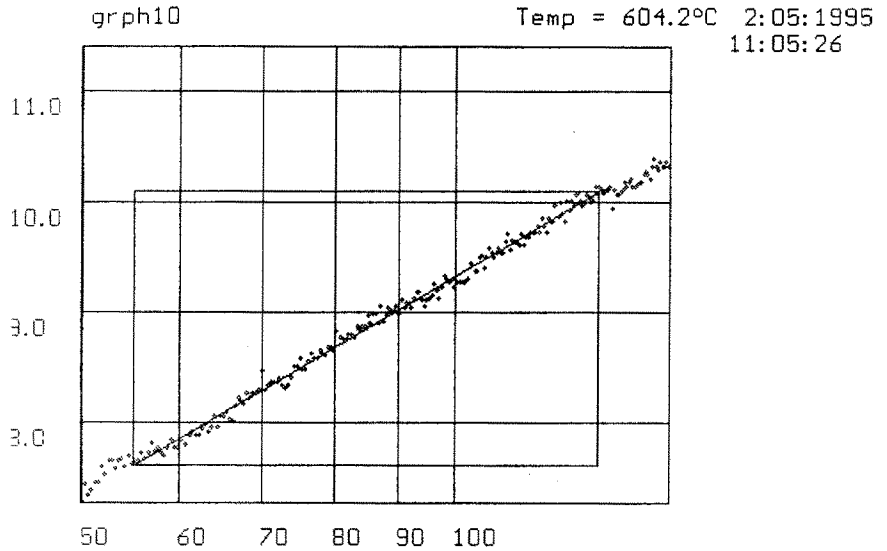

Fig. 7 Deviations of measured temperature rise data points from the fitted line between a time interval from t_{min} to t_{max}

ware. After the data are saved and/or analyzed, the described measurement cycle starts again at the next desired temperature, defined before. This enables the experimental procedure and analysis to be conducted in fully automatic way. All hot wire software package routines are written in Borland C^{++} programming language.

In order to demonstrate that the instrument described above operates in accordance with the theoretical model described, a number measurement of thermal conductivity has been performed on various samples, made from various materials. On Fig. 6 is presented a typical temperature rise *vs.* natural logarithm time, measured on graphite sand with the transient hot wire probe technique. Figure 7 shows a plot of a set of experimental points $[T(t),t]$ for this data in another scale, made by zoom routine, in order to demonstrate agreement with the calculated linear least squares fit.

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