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Temperature dependence measurements of thermal parameters of solid samples by the relaxation method

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Abstract—A generalization of the relaxation method [Budke and Dieška, *Acta Phys. Slov.* **35**, 27–39 (1985)] has been elaborated. While an application of the relaxation method requires the sample holder temperature to be constant in time, this generalized version admits a polynomial change of the sample holder temperature. Such a temperature course is a very common and realistic one at temperature dependence measurements of thermal parameters.

1. INTRODUCTION

NON-STATIONARY methods for measuring the thermal parameters (thermal conductivity, λ, and thermal diffusivity, a) of solid samples have been described in refs. [1–6]. They have involved a point, a line or a plane source.

Non-stationary methods, based on the recording of a transient effect of heat transfer, are used mainly for the investigation of thermal properties of materials at continuously changing conditions of an experiment. In contrast to stationary methods they enable the realization of a measurement in a time substantially shorter than the time characteristic for the change in conditions of an experiment. These must be mostly taken into consideration either through the changes of the temperature of a sample holder or the change of thermal parameters of the measured sample in the course of time (as in the case of kinetic effect investigations [7]).

In this paper attention is focused on the applicability of the so called 'relaxation method' [6] at varying conditions of an experiment, for a non-constant temperature variation of the sample holder.

2. RELAXATION METHOD

The relaxation method [6] is based on the recording of the time dependence of the temperature of a plane heat source, which is in ideal contact with one side of the measured plane-parallel sample (Fig. 1).

The sample holder enforces the temperature $T(x_0, t) = \text{const.}$ (where t is time, T is temperature and x_0 is sample thickness) on that side of the sample, which is opposite the plane of the heat source. In practice a symmetrical arrangement is used with two samples and with the heat source between them. The heat source is assumed to be an ideal heat conductor with finite heat capacity per unit area C. The power per unit area, $q(t)$, does not depend on the coordinates and is a step function of time.

Heat losses from the source and the lateral walls of the (finite) plane-parallel sample (otherwise arbitrarily shaped) are neglected. A detailed discussion of this assumption is presented in ref. [6]. The temperature changes occurring during one measurement are supposed to be so small, that the thermal parameters of the sample (thermal conductivity, λ, and thermal diffusivity, a) are constant.

Such a model is described by the one-dimensional heat conduction equation

$$\frac{\partial T(x,t)}{\partial t} = a\frac{\partial^2 T(x,t)}{\partial x^2}, \tag{1}$$

where $a = \lambda/c_p\rho$ is thermal diffusivity (c_p is the specific heat at the constant pressure and ρ is the mass density of the sample). The corresponding boundary conditions are:

$$C\frac{\partial T(0,t)}{\partial t} = q(t) + \lambda\frac{\partial T(0,t)}{\partial x}, \tag{2}$$

where

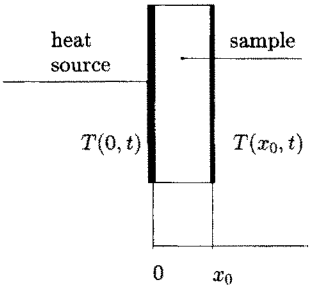

FIG. 1. The measured plane-parallel sample with the plane heat source.

NOMENCLATURE

a	thermal diffusivity
c_p	specific heat of sample at constant pressure
C	heat source heat capacity per unit area
q_0	heat source power per unit area
t	time
T	temperature difference between the heat source and the sample holder
$\bar{u}(x,p)$	Laplace transformation of the function $u(x,t)$
$v_n/n!$	coefficient of a polynomial
x	coordinate in the direction orthogonal to the plane-parallel sample
x_0	thickness of sample.

Greek symbols

α	increase rate of sample holder temperature
θ	unit jump function
λ	thermal conductivity
μ_n	roots of characteristic function
ρ	mass density of sample
τ_n	time constant.

$$q(t) = q_0\theta(t) \tag{3}$$

and

$$T(x_0,t) = 0; \tag{4}$$

$\theta(t)$ is the unit step function. The initial condition is

$$T(x,0) = 0. \tag{5}$$

It can be shown that for such a model the temperature difference $T(t)$ between the heat source and the sample holder is

$$T(t) \equiv T(0,t) = \frac{q_0x_0}{\lambda}\left[1-\sum_{n=1}^{\infty} A_n\exp\left(-\frac{t}{\tau_n}\right)\right] \tag{6}$$

with

$$A_n = \frac{2}{1+y+y^2\mu_n}\cdot\frac{1}{\mu_n^2}, \quad \tau_n = \frac{x_0^2}{a\mu_n^2}, \tag{7}$$

where μ_n are the roots of the characteristic equation

$$\cot\mu = y\mu, \quad y \equiv \frac{Ca}{\lambda x_0}. \tag{8}$$

For time larger than the largest time constant τ_n, i.e. $t > \tau_1$, equation (6) indicates

$$T(t) \cong T_{\max}\left[1-A_1\exp\left(-\frac{t}{\tau_1}\right)\right], \tag{9}$$

where $T_{\max}$ denotes

$$T_{\max} = \frac{q_0x_0}{\lambda}. \tag{10}$$

By fitting the parameters of formula (9) to the experimental values one can find $T_{\max}$ and τ_1 and from them the required parameters λ and a (see details in ref. [6]).

3. THE RELAXATION METHOD AT VARYING TEMPERATURE CONDITIONS

For many experimental conditions the sample holder temperature may vary with time. This implies that an additional value of temperature is superimposed on the temperature field given by equations (1), (2), (4) and (5). This contribution, which will be denoted by $u(x,t)$, must satisfy the heat conduction equation (1), the boundary condition (2) without the heat source ($q_0 = 0$) and the initial condition (5). Instead of boundary condition (4) it is now required that

$$u(x_0,t) = \sum_{n=0}^{N} v_n\frac{t^n}{n!}. \tag{11}$$

Relation (11) means that the sample holder temperature is described by a polynomial of the Nth degree with the known coefficients v_n to one side of the sample.

Applying Laplace transformation to equations (1), (2) and (11) the following are obtained:

$$p\bar{u}(x,p) = a\frac{\partial^2\bar{u}(x,p)}{\partial x^2}, \tag{12}$$

$$Cp\bar{u}(0,p) = \lambda\frac{\partial\bar{u}(0,p)}{\partial x}, \tag{13}$$

$$\bar{u}(x_0,p) = \sum_{n=0}^{N} v_n p^{-(n+1)}, \tag{14}$$

where $\bar{u}(x,p) = \int_0^\infty e^{-pt}u(x,t)\,dt$ is the Laplace transformation of the function $u(x,t)$.

The solution of this boundary problem can be written in the form

$$\bar{u}(x,p) = \bar{u}(x_0,p) \times \frac{\left(Cp+\lambda\sqrt{\frac{p}{a}}\right)\exp\left(x\sqrt{\frac{p}{a}}\right)-\left(Cp-\lambda\sqrt{\frac{p}{a}}\right)\exp\left(-x\sqrt{\frac{p}{a}}\right)}{\left(Cp+\lambda\sqrt{\frac{p}{a}}\right)\exp\left(x_0\sqrt{\frac{p}{a}}\right)-\left(Cp-\lambda\sqrt{\frac{p}{a}}\right)\exp\left(-x_0\sqrt{\frac{p}{a}}\right)}. \tag{15}$$

The Laplace transformation of the difference

$$\Delta\bar{u}(p) = \bar{u}(0,p) - \bar{u}(x_0,p), \qquad (16)$$

has the form

$$\Delta\bar{u}(p) = \bar{u}(x_0,p)[\bar{g}(p) - 1], \qquad (17)$$

where

$$\bar{g}(p) = \frac{1}{yx_0\sqrt{\frac{p}{a}}\sinh\left(x_0\sqrt{\frac{p}{a}}\right) + \cosh\left(x_0\sqrt{\frac{p}{a}}\right)}. \qquad (18)$$

The inverse transformation of the relation (17) can be written as a convolution

$$\Delta u(t) = \int_0^t u(x_0,\tau)\, g(t-\tau)\,\mathrm{d}\tau - u(x_0,\tau), \qquad (19)$$

where

$$g(t) = \sum_{n=1}^{\infty} G_n \exp\left(-\frac{t}{\tau_n}\right) \qquad (20)$$

and

$$G_n = \frac{2a}{x_0^2}\cdot\frac{\mu_n\sqrt{(1+y^2\mu_n^2)}}{1+y+y^2\mu_n^2}. \qquad (21)$$

Time constants τ_n are defined identically as in (7) and coefficients μ_n are again the roots of the characteristic equation (8). Relation (19) can be, after some calculations, expressed in the form

$$\Delta u(t) = \sum_{k=0}^{N-1} a_k t^k + \sum_{n=1}^{\infty} B_n \exp\left(-\frac{t}{\tau_n}\right), \qquad (22)$$

where a_k, S_n and B_n are given by:

$$a_k = \frac{1}{k!}\sum_{m=1}^{N-k} (-1)^m v_{k+m} S_m \qquad (23)$$

$$S_m = \sum_{n=1}^{\infty} G_n \tau_n^{m+1} = \frac{(-1)^m}{m!}\lim_{p\to 0}\frac{\partial^m}{\partial p^m}\bar{g}(p) \qquad (24)$$

and

$$B_n = G_n \sum_{m=1}^{N} (-1)^{m+1} v_m \tau_n^{m+1}. \qquad (25)$$

The coefficients G_n are defined in equation (21).

4. DISCUSSION

By measuring the transient temperature difference between the heat source and the sample holder the superposition of the influence of the heat source (6) and of the change of the sample holder temperature (22) can be obtained. These contributions cannot be separated during the measurement and, in general, they can be of the same order of magnitude.

From the form of solution (22) an important general property can be seen. If the sample holder temperature changes in time as a polynomial of the Nth degree and the heat source is inactive, the temperature difference between the two sides of the plane-parallel sample is a polynomial of the $(N-1)$th degree.

The relation

$$T(t) \cong \sum_{k=0}^{N-1} a_k t^k + T_{\max} + (B_1 - T_{\max}A_1)\exp\left(-\frac{t}{\tau_1}\right) \qquad (26)$$

is now the analogue to the approximate relation (9). Relation (26) was obtained by summing the relations (6) and (22) for the time $t > \tau_1$.

From the experimentally obtained dependence of $T(t)$, $T_{\max}$ and τ_1 must be identified in order to determine λ and a, respectively. To do so, the coefficients a_k must be fixed in another way. Relation (23) shows the need of knowledge of coefficients v_m (obtainable from the measurement of the time dependence of the sample holder temperature) and the knowledge of parameters S_m in (24). Consider the first four of the S_m:

$$S_0 = 1$$

$$S_1 = \frac{x_0^2}{a}\left(y + \frac{1}{2}\right)$$

$$S_2 = \frac{x_0^4}{a^2}\left(y^2 + \frac{5}{6}y + \frac{5}{24}\right)$$

$$S_3 = \frac{x_0^6}{a^3}\left(y^3 + \frac{7}{6}y^2 + \frac{61}{720}\right). \qquad (27)$$

From the relations (27) it is evident that S_m, and consequently a_k, depend on yet unknown quantities λ and a. This fact causes some complications which can be avoided in one of the two ways:

1. An iteration procedure to fit the expression on the right-hand side of equation (26) to the experimental values, $T(t)$, i.e. the λ and a, used in iterations, must be identical with those of calculated ones. When calculating the right-hand side of equation (26) the definition of a_k is used, which needs the experimental values of v_n. v_n is known from an independent measurement of the sample holder temperature.
2. For identification of individual terms in equation (26) one can employ the following experimental procedure: quantity $T(t)$ is measured in time intervals

—when the heat source is active and
—when it is inactive and all exponential transient effects have died away.

The second group of measurements enables the first term on the right-hand side of equation (26) to be identified. From measurements, when the heat source is active, it is subsequently possible to determine values of $T_{\max}$ and τ_1.

Such a sequence of computations can be used only if the sample holder temperature is a smooth function of time. In practice this is the most common case and the choice of $N-1 = 2$ is precise enough.

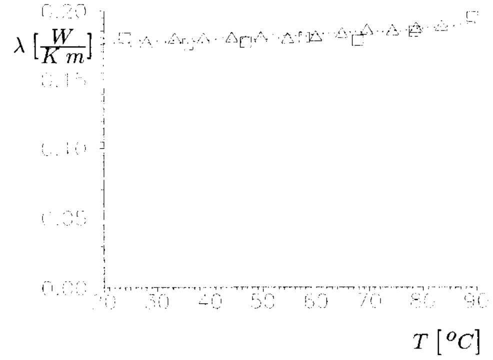

FIG. 2. Temperature dependences of thermal conductivity λ measured at different rates of increase of sample holder temperature α. Dots denote $\alpha = 0.0018$ K s^{-1}, △, $\alpha = 0.0078$ K s^{-1}, □, $\alpha = 0.018$ K s^{-1}.

To illustrate our statements temperature dependence measurements of an epoxide bitumen thermal conductivity in a temperature interval of 20–90°C are considered. This material is characterized by a slight dependence $\lambda(T)$. The time constant τ_1 of our samples was $\tau_1 \simeq 50$ s.

Measurements are presented for the temperature dependences of thermal conductivity of samples at three different rates of increase of the sample holder temperature: $\alpha_1 = 0.0018$ K s^{-1}, $\alpha_2 = 0.0078$ K s^{-1} and $\alpha_3 = 0.018$ K s^{-1}. From Fig. 2 it is evident that the values of thermal conductivity are nearly independent of αs. When realizing that during one measuring period ($\simeq 10 \times \tau_1$) the temperature of sample holder has changed by about 0.9, 3.9 and 9 K, respectively, then the observed fact that λ does not depend on α justifies the use of the proposed method.

5. CONCLUSION

A generalization of the relaxation method presented in ref. [6] has been elaborated. In this case the temperature of the surroundings need not be constant but it can change in time as a polynomial of the Nth degree. From the experimental point of view this method shortens the necessary time of usually lengthy temperature dependence measurements of thermal parameters and improves accuracy.

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