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A New Method for the Evaluation of Thermal Conductivity and Thermal Diffusivity from Transient Hot Strip Measurements[1]

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The standard straight-line fit to data of a transient hot strip (THS) experiment to determine the thermal conductivity λ and thermal diffusivity a suffers from two major drawbacks: First, due to the statistical nature of the estimation procedure, there is no relation between the uncertainty of the measured value on one hand and the transport properties obtained on the other. Second, in order to account for the heat capacity of the strip and outer boundary conditions, two intervals of the plot must be rejected before analyzing it. So far, these intervals are selected arbitrarily. We now treat the THS working equation as a function of the four parameters concerned, λ, a, U_0 (initial voltage), and t_0 (time delay). Chi-square fittings, following the Levenberg–Marquardt algorithm, are performed separately for several overlapping time intervals of the entire plot to find λ and a with minimal standard deviation. In the course of subsequent iterations an individual weighting factor is applied to each point to account for systematic errors. This procedure yields the "best" values of λ and a along with their individual errors, comprising the systematic and the statistical errors. Experimental results on Pyrex glass 7740 were taken to verify the new procedure.

KEY WORDS: Pyrex; thermal conductivity; thermal diffusivity; transient hot strip.

1. INTRODUCTION

Compared with steady state methods of measurement, the transient hot strip (THS) method developed by Gustafsson et al. [1] to determine the thermal conductivity λ and the thermal diffusivity a of solids offers a

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number of advantages. Among these are short measurement times and a simple experimental setup. The complex evaluation method causes, however, considerable difficulties as far as the determination of the uncertainties of the measurands a and λ is concerned.

2. THEORY

In an ideal THS experiment, a current of sufficient intensity flows through an infinitely long and thin metal strip, which is completely surrounded by the dielectric to be measured. The entire electric power $P = \text{const}$ is transformed into Joule heat, which diffuses freely in the specimen. The temperature increase with time $\Delta T(t)$ of the strip is a measure of the thermal conductivity λ and the thermal diffusivity a of the material. It leads to an increase in the electrical resistance of the strip, which can be measured as voltage drop $U(t)$ along the strip [1]:

$$U(t) = U_0 + \frac{\alpha I U_0^2}{4 l \lambda \sqrt{\pi}} f(\tau) = U_0 + k f(\tau) \tag{1}$$

where

$$f(\tau) = \tau \operatorname{erf}\left(\frac{1}{\tau}\right) - \frac{\tau^2}{(4\pi)^{1/2}}\left[1 - \exp\left(-\frac{1}{\tau^2}\right)\right] - \frac{1}{(4\pi)^{1/2}} \operatorname{Ei}\left(-\frac{1}{\tau^2}\right)$$

and

$$\tau = \frac{[a(t - t_0)]^{1/2}}{d}$$

U_0 is the measurement voltage at $t = 0$, α is the temperature coefficient of the electrical resistance of the strip, τ is the dimensionless time (square root of the Fourier number), t_0 is the time delay before the power is released in the strip, and l and d are the half-length and width, respectively, of the strip. The error function and the exponential integral are denoted by $\operatorname{erf}(\cdot)$ and $\operatorname{Ei}(\cdot)$, respectively. Equation (1) cannot be solved analytically with respect to the two measurands, λ and a. In an iteration process, the four unknown parameters U_0, k, a, and t_0 must therefore be fitted to the linear, measured curve $U_i(f(\tau_i))$ so that the relevant correlation coefficient reaches a maximum. This termination condition results in the squares of the deviations (χ) between measured and calculated values becoming a minimum [2]:

$$\chi = \sum \left(\frac{U_i - U(t_i)}{\sigma_i}\right)^2 \rightarrow \min \tag{2}$$

where σ_i is the standard deviation of the value U_i and describes the weight of this value.

For the method of analysis used so far [1], initially the number of the parameters to be fitted is reduced from four to one, i.e., to the quantity a, before iteration is carried out. The simplifying assumption here is that there is a uniform distribution of the standard deviation to the individual measurement values U_i which means a uniform weighting of these data, $1/\sigma_i = 1$. Furthermore, it is assumed that the function $f(\tau)$ can be estimated to be $f(\tau) \approx \tau - \tau^2/(4\pi)^{1/2}$ for $\tau < 0.7$, and the set of data reduced to this range can be described by the square polynomial $U = U_0 + A(t + t_0)^{1/2} + B(t + t_0)$. This term leads directly to the time delay t_0 [3–5]. Finally, the two other parameters, U_0 and k, can be excluded analytically so that only the quantity a must be fitted.

It is an essential drawback of this method that in the condition $\tau(a) < 0.7$ the quantity searched, a, is considered to be known. In addition, it can be easily demonstrated that the distribution of the standard deviation of U_i is in fact nonuniform. After all, the systematic measurement errors by which all measured values are affected are not taken into consideration in the evaluation. An improved method must not, therefore, limit the range covered by the experiment, $[\tau(a)]$, from the very outset; this makes, however, the simultaneous fitting of all four parameters necessary. The systematic measurement errors should then be included in the analysis.

The iterative search for the parameters U_0, k, a, and t_0 is conducted according to the Levenberg–Marquardt algorithm [2]. This method consists in finding such increments of the model parameters b_i ($b_1 = U_0$, $b_2 = k$, $b_3 = a$, $b_4 = t_0$) which correspond to a "movement" along the gradient of the sum χ towards the minimum. These increments Δb_i are the solution of the system of equations

$$\sum_{l=1}^{4} \alpha'_{kl}\,\Delta b_l = \beta_k, \qquad k = 1,\dots, 4 \tag{3}$$

with

$$\alpha'_{kl} \equiv \begin{cases} \alpha_{kl}(1+\varphi), & k = 1 \\ \alpha_{kl}, & k \neq 1 \end{cases}$$

$$\alpha_{kl} = \sum_{i=1}^{N} \frac{1}{\sigma_i^2}\left[\frac{\partial U(\vec{b}, t_i)}{\partial b_k}\,\frac{\partial U(\vec{b}, t_i)}{\partial b_l}\right] \tag{4}$$

$$\beta_k = \sum_{i=1}^{N} \frac{U_i - U(\vec{b}, t_i)}{\sigma_i^2}\,\frac{\partial U(\vec{b}, t_i)}{\partial b_k}$$

N is the number of measured points, $\vec{b} \equiv (b_1, b_2, b_3, b_4)$, φ is a measure of the "distance" from the minimum searched. φ disappears when the minimum is approximated. The individual derivatives covered by Eq. (4) are the following:

$$\frac{\partial U}{\partial b_1} \equiv \frac{\partial U}{\partial U_0} = 1$$

$$\frac{\partial U}{\partial b_2} \equiv \frac{\partial U}{\partial k} = f(\tau)$$

$$\frac{\partial U}{\partial b_3} \equiv \frac{\partial U}{\partial a} = k\,\frac{\partial f(\tau)}{\partial a} = \frac{k\tau}{2a}\left[\operatorname{erf}\left(\frac{1}{\tau}\right) + \frac{\tau}{\sqrt{\pi}}\exp\left(-\frac{1}{\tau^2}\right) - \frac{\tau}{\sqrt{\pi}}\right]$$

$$\frac{\partial U}{\partial b_4} \equiv \frac{\partial U}{\partial t_0} = \frac{k\tau}{2(t+t_0)}\left[\operatorname{erf}\left(\frac{1}{\tau}\right) + \frac{\tau}{\sqrt{\pi}}\exp\left(-\frac{1}{\tau^2}\right) - \frac{\tau}{\sqrt{\pi}}\right]$$

The procedure proper starts with the assignment of values to U_0, the first voltage value measured, and to the material properties a and λ searched. Furthermore, t_0 is set equal to zero. The procedure is then continued as follows:

1. Calculation of $\chi_0 = \chi(U_0, k, a, t_0)$.
2. Calculation of β_k and α'_{kl}; solving of the system of equations (3) and estimation of the sum $\chi_1 = \chi(U_0 + \Delta U_0,\ k + \Delta k,\ a + \Delta a,\ t_0 + \Delta t_0)$.
3. If $\chi_1 \geqslant \chi_0$, φ is multiplied by 10.
4. If $\chi_1 < \chi_0$, φ is divided by 10; assignment of the relevant values $\vec{b} + \Delta\vec{b}$ to the parameters $\vec{b}$ and setting χ_0 equal to χ_1.
5. Return to step 2.

The calculation is discontinued when the increments Δb_i are sufficiently small.

Systematic measurement errors can now be accounted for without any problem in Eq. (4) as the standard deviation σ_i of the measured values. The new evaluation method takes the following components into account: the instability of the current source and the measurement error of the voltmeter, the heat capacity of the strip, the variation of the power during constant-current operation, and a possible heating of the specimen surfaces if the experiment is carried out incorrectly. Because of these disturbing effects the voltage deviations are separately calculated for each point of time, summed up, and then used in Eq. (4) as the value of the standard deviation. Systematic deviations caused by the thermal contact resistance between the specimen and the strip and by end effects due to heavy

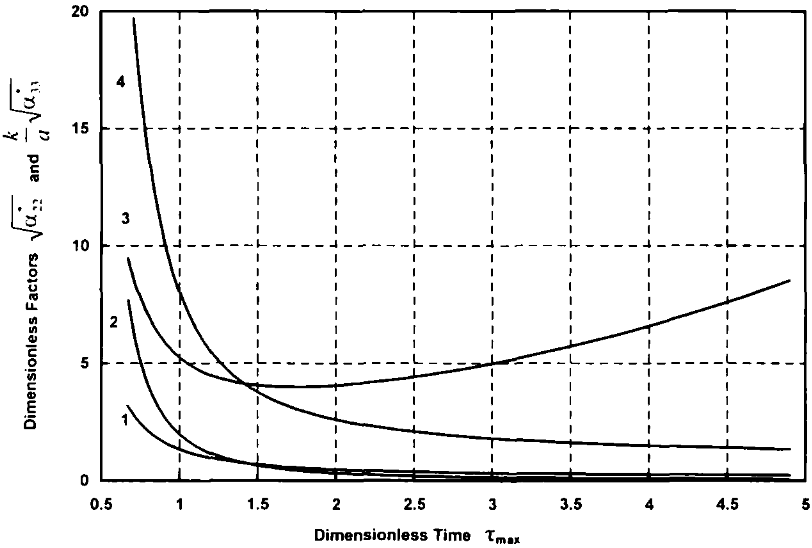

Fig. 1. Dimensionless factors of the uncertainties $(\alpha^*_{22})^{1/2}$ (for λ, curves 1 and 3) and $(k/a)(\alpha^*_{33})^{1/2}$ (for a, curves 2 and 4) versus upper dimensionless time limit τ_{max} calculated for two cases: $N = 1000$ (curves 1 and 2) and $N = 500\tau^2_{max}$ (curves 3 and 4).

electrical leads, which were shown in Refs. 1, 3, and 6 to be negligibly small, are not taken into account in the evaluation.

The influence of random measurement errors on the uncertainty of measurement of λ and a has not yet been analyzed in detail in the literature. However, first investigations carried out at the PTB [6] suggest that even smallest differences (0.007%) between two model curves $U_1(t)$ and $U_2(t)$ obtained by calculation may lead to differences of 20% for a and of 6% for λ. The confidence interval of the parameter b_k is given by

$$\delta b_k = [F(P, j)]^{1/2} \left(\frac{\chi_{min}}{N-j}\right)^{1/2} (\alpha^*_{kk})^{1/2}$$

where $F(P, j)$ is a function of the confidence level (P) and of the parameters number (j), α^*_{kk} being the corresponding element of the inverted matrix $[\alpha]$ $([\alpha^*] \equiv [\alpha]^{-1})$. According to Ref. 2 for $P = 95.4\%$ and $j = 4$, $F(95.5\%, 4) = 9.70$. At the confidence level of 95.4%, the following are valid for the confidence intervals of the thermal conductivity and the thermal diffusivity:

$$\frac{\delta\lambda}{\lambda} = \frac{\delta k}{k} = \left[\frac{9.7\chi_{min}}{k^2(N-4)}\right]^{1/2} (\alpha^*_{22})^{1/2}, \qquad \frac{\delta a}{a} = \left[\frac{9.7\chi_{min}}{k^2(N-4)}\right]^{1/2} \frac{k}{a} (\alpha^*_{33})^{1/2} \quad (5)$$

As the two dimensionless quantities $(\alpha_{22}^*)^{1/2}$ and $(k/a)(\alpha_{33}^*)^{1/2}$ usually differ, the individual uncertainties of a and λ must differ as well. Figure 1 shows these quantities separately, as functions of the upper limit of the ranges $[0, \tau_{max}]$, each time calculated for two complementary cases:

1. *Ideal case:* The distance between the real-time reference points $t_{i+1} - t_1$ is constant for all τ, whereas the number of measurement points changes according to $N = 500\tau^2$: *curves 3 and 4.*

2. *Real case:* The number of measurement points ($N = 1000$) is constant, whereas the distance between the real-time reference points $t_{i+1} - t_i$ varies with τ: *curves 1 and 2.*

Curves 3 and 4: In the ideal case of an infinitely extended sample and constant power output from the strip, the measurement errors of both a and λ decrease with increasing Fourier number, i.e., with the duration of the experiment.

Curves 1 and 2: In the practical case of the finitely extended sample, the duration of the experiment and thus the number of the measured values are limited because of the fixed outer boundary conditions. For an upper limit between $\tau = 1.5$ and $\tau = 2$, the measurement error of a is smallest; that of λ decreases strictly monotonically with τ.

From an examination of Fig. 1 it can be seen that the uncertainty of the measurand λ is in fact always smaller than that of a.

In the course of the experiment, the initial deviations of the real system from the model increase even more with time, due, for example, to the increased electric power input into the strip in constant-current operation and the finite size of the sample body. Per definition, given by Eq. (5), the respective confidence intervals $\delta\lambda/\lambda$ and $\delta a/a$ of the measurands first decrease with increasing τ, reach a minimum for certain τ_{max} values, and then increase again. Within the scope of the new evaluation method, the position of the minima is calculated and the measured curve evaluated in the range which furnishes the smallest standard deviations for the two measurands.

3. RESULTS

Specimens of Pyrex glass 7740 prepared by le Société Corning France (Sovirel) were used for a first experimental verification of the above evaluation method. This material has been studied widely [5–7, 10–17] and is especially useful for qualification measurements. The thermal conductivity and the thermal diffusivity of this particular Pyrex glass were measured in the temperature interval between 191 and 473 K. The temperature coefficient of the electrical resistance of the nickel strip used was calibrated in this temperature range.

The measurement time Δt was 35 s for each measurement; 500 measurement points were recorded each time. The evaluation of the measurement series furnished an optimum measurement time $\Delta t_{opt} = 30$ s (for a minimum value of the confidence interval), i.e., a value somewhat smaller than the actual measurement time. Measurement times shorter than 30 s lead to a greater uncertainty of the results, whereas longer measurement times (measurements were carried out up to $\Delta t = 105$ s) neither increase nor decrease the confidence interval. The uncertainties were estimated at 3.2% for λ and at 17% for a (confidence level: 95%). The repeatability was checked in nine measurements of the same kind which yielded a repeatability of a single measurement value of 1.3% for λ and of 9% for a.

The measured thermal conductivity $\lambda(T)$ of Pyrex and its literature values are shown in Fig. 2. The THS data agree with the reference values with a some deviation from them at the high temperatures.

In the temperature range indicated, the thermal diffusivity values decrease from 0.52 (at 191 K) to 0.41 $mm^2 \cdot s^{-1}$ (at 423 K) and are approximately 25% smaller then the reference data [12, 13, 16, 17].

The deviation of both measured values, the thermal conductivity and thermal diffusivity, from THS data given in Ref. 6 is smaller than 1%.

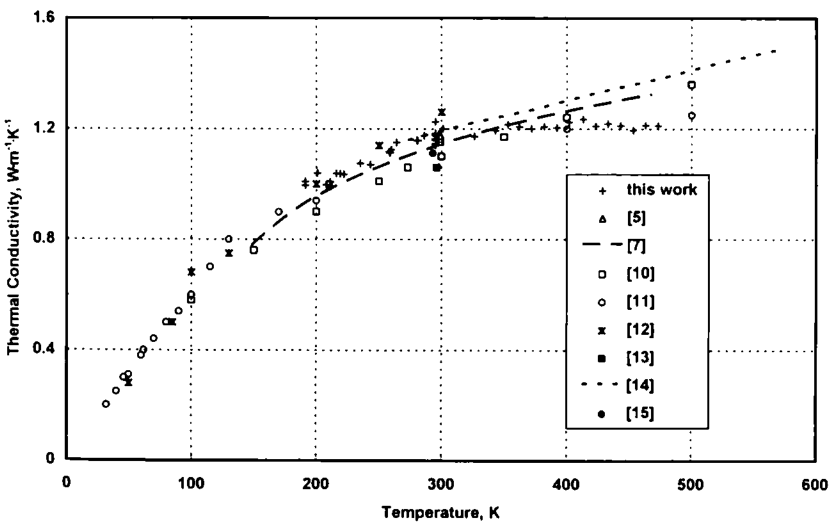

Fig. 2. Temperature dependence of the thermal conductivity of Pyrex glass 7740.

4. CONCLUSIONS

The Levenberg–Marquardt algorithm has been used for the first time to evaluate THS measurement curves, which allows all four unknown parameters of the mathematical model of the THS method to be fitted simultaneously. This means that, in addition to the measurands λ and a, the parameters "initial voltage" and "time correction" are also fitted. So far, the two last quantities could only be estimated. Another essential advantage of the new procedure is that the uncertainty of the two measurands can now be determined analytically. It is shown in this context that the quantity λ is always affected by a smaller uncertainty than that of a. The evaluation of various intervals of the same measurement curve previously have furnished values for a and λ which deviate substantially. The newly developed method excludes this error and instead selects that interval for evaluation which guarantees minimum uncertainties for a and λ.

The results obtained both theoretically and by experiment indicate that the values of the measurands a and λ are very sensitive to even the smallest random measurement errors (0.007%). This effect involves considerable difficulties with regard to an improvement of the uncertainty of measurement by the THS method of, at present, 3.2% for λ and 17% for a. It is therefore to be assumed that the uncertainty of measurement of 1% stated in the literature for the thermal conductivity and, especially, for the thermal diffusivity will hardly bear a closer scrutiny.

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