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Thermal Diffusivity of Solids with a Low Expansion Coefficient: A Dilatometric Technique

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A dilatometric method is presented, suitable to obtain both thermal diffusivity and conductivity of low-conducting solids with a low expansion coefficient. The repeatibility of the measurements of thermal conductivity is 3%, whereas that for diffusivity is 5%. Data for fused silica at room temperature are given, consistent with those reported in the literature. Since the method is based on detecting the thermal expansion of a copper disk in thermal contact with the specimen, its range of applicability is linked to the sensitivity by which the dilation of copper can be measured: no difficulty arises between liquid nitrogen and 1000°C.

KEY WORDS: low-conducting materials; low-expanding materials; thermal conductivity; thermal diffusivity.

1. INTRODUCTION

The accuracy of thermal diffusivity measurements is increased by reducing the uncontrolled heat exchanges between specimen and environment. Satisfactory results in this direction can be achieved by a new method [1, 2] which has recently been developed for the determination of thermal diffusivity of low-conducting solids with a high thermal expansion coefficient and is based on the analysis of the expansion of the specimen in contact with a heat source. In the present paper, we describe the extension of the method to low-conducting, low-expanding materials.

The experimental apparatus used for this kind of solids is shown in Fig. 1. The specimen S is inside a hollow cylinder G of the same material, which acts as a thermal guard. This system is put on a copper disk C_1

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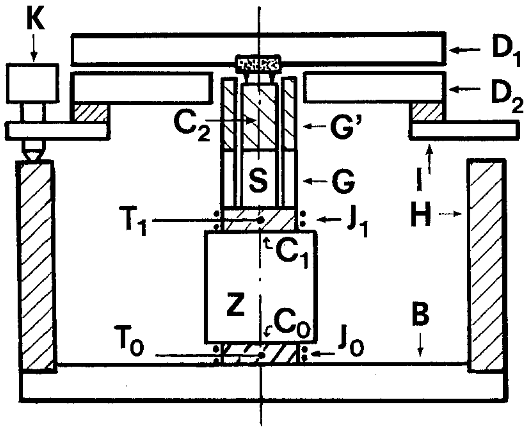

Fig. 1. Schematic view of the experimental apparatus.

surrounded by an insulated electric wire J_1, which is connected to a current generator. The Joule heat engendred in this way by J_1 is rapidly transferred to C_1 (where the temperature at any time can be considered as independent of the space coordinates, owing to the high conductivity of copper) and then slowly to S, which is characterized by a low thermal diffusivity. In this way C_1 works as a heat source for the specimen. A thermocouple T_1 inserted into C_1 is used to record the time behavior of the temperature of the heat source. On the upper base of S we put a second copper disk C_2, surrounded by the corresponding copper guard G′, which lies on G. The principle of the method consists of recording, as a function of time, the thermal expansion of C_2 as a consequence of the transfer of heat from C_1 to C_2 through the specimen S. The analysis of this experimental function is sufficient to provide both the thermal diffusivity and the thermal conductivity of S. The thermal expansion is easily detected by means of a capacitor having one plate represented by the fused silica disk D_1 which is supported by C_2. The contact between C_2 and D_1 consists of a fused silica tripod. The contact area between D_1 and the three pins at 120° of the tripod is negligibly small, so that the heat lost by conduction trhough the pins is correspondingly negligible. Also, the heat lost by radiation through the upper face of C_2 is negligible if the experiment is performed in vacuum and the copper surface is well polished. The second plate of the capacitor is the annular fused silica disk D_2. The two facing surfaces of D_1 and D_2, as well as their lateral borders, are coated by a conducting film of tin oxide. The distance between D_1 and D_2 can be adjusted by means of three leveling screws K pressing on a hollow cylinder of fused silica, H, supporting D_2 and fixed to the base B.

In order to know the correspondence between the change of gap between D_1 and D_2 and the capacitive signal, one has to perform a calibration [2] of this signal. For this purpose, the disk C_1 is put on a cylinder of Zerodur Z (glass ceramic of very low thermal expansion coefficient, $0.5 \times 10^{-7}\,K^{-1}$), and this in turn on the copper disk C_0, lying on the fused silica base B. Having switched on the current in the electric wire J_0 surrounding C_0, one simultaneously records the temperature change $\theta^{(0)}$ of this disk with respect to the environment (by means of thermocouple T_0) and the time behavior of the capacitive signal. In the calibration experiment, such a signal is unambiguously due to the known thermal expansion of C_0, namely, $\beta_{Cu} h_0 \theta^{(0)}$, where h_0 is the thickness of C_0 and β_{Cu} is the thermal expansion coefficient of copper. In fact, owing to the low thermal diffusivity of Zerodur, the height of Z (3.0 cm) is sufficient to prevent heat from reaching the copper disk C_1 during the time of measurement. Consequently, the experiment consists of two steps: in the first step, one calibrates the capacitor by heating the disk C_0; in the second step, one uses the heater C_1, and from the analysis of the capacitive signal both the thermal diffusivity and the thermal conductivity of the specimen are deduced.

By the equipment shown in Fig. 1, one achieves the important result of detecting a thermal field without perturbing the field itself. In fact the detector is represented in this case by the capacitor, which has no appreciable heat exchange with the underlying system $S-C_2$. From this point of view, the present experimental arrangement represents a considerable improvement with respect to the sandwich described in Ref. 3, where we essentially used the system C_1-S-C_2 with a thermocouple inserted into C_2. Though small, the presence of the thermocouple could introduce an uncontrolled source (or sink) of heat in the measuring system. In the present case, it has to be emphasized that the thermocouple inserted into C_1 does not introduce any uncontrolled heat source, since the heat gained or lost through the thermocouple wires (or through the resistive coil J_1) is included into the heat source itself.

2. MATHEMATICAL FRAMEWORK

The problem is unidimensional since the base of the specimen is uniformly heated and heat exchanges through the lateral surface are minimized by the thermal guard. Let k and k' be the thermal conductivities of the specimen and of copper, respectively, α and α' the corresponding thermal diffusivities, and b and d the length of the specimen and of C_2. Let us introduce a coordinate system (z) with the origin at the center of the lower base of the specimen and a system (z') with the origin at the lower base of

C_2. If the entire time interval of measurement is subdivided into N parts of equal width τ, the heat exchanged between specimen and copper disks can be considered constant in each time interval $t_{m+1}-t_m=\tau$, provided τ is chosen sufficiently small. We directly take from Ref. 3 the temperature fields $\theta_{m+1}(z)$ and $\theta'_{m+1}(z')$, referring to the specimen and to the copper disk C_2, respectively, at the time t_{m+1}. Owing to the boundary conditions imposed on the heat diffusion equation, their expressions vanish at $t=0$, so that the above fields actually represent the temperature changes with respect to the environment. The fields are given by the expressions (valid for $m \geqslant 1$)

$$\begin{aligned}\theta_{m+1}(z) = {} & (Q_m - XQ'_m)(\alpha t_{m+1} + \tfrac{1}{2}z^2)/b^2 - Q_m z/b + Q_0 M_z(t_{m+1}) \\ & - \sum_{p=1}^{m} [\alpha t_p/b^2 + \tfrac{1}{2}G_z(t_{m+1}-t_p)](Q_p - XQ'_p - Q_{p-1} + XQ'_{p-1}) \\ & + \sum_{p=1}^{m} M_z(t_{m+1}-t_p)(Q_p - Q_{p-1}) - \tfrac{1}{2}(Q_0 - XQ'_0)\, G_z(t_{m+1}) \end{aligned} \tag{1}$$

and

$$\begin{aligned}\theta'_{m+1}(z') = {} & Q'_m[\alpha' t_{m+1} + \tfrac{1}{2}(z'-d)^2]/d^2 - \tfrac{1}{2}Q'_0 R_{z'}(t_{m+1}) \\ & - \sum_{p=1}^{m} (Q'_p - Q'_{p-1})[\alpha' t_p/d^2 + \tfrac{1}{2}R_{z'}(t_{m+1}-t_p)] \end{aligned} \tag{2}$$

and by the expressions (valid for $m=0$)

$$\begin{aligned}\theta_1(z) = {} & (Q_0 - XQ'_0)(\alpha t_1 + \tfrac{1}{2}z^2)/b^2 - Q_0 z/b \\ & - \tfrac{1}{2}(Q_0 - Q'_0)\, G_z(t_1) + Q_0 M_z(t_1) \end{aligned} \tag{3}$$

and

$$\theta'_1(z') = Q'_0[\alpha' t_1 + \tfrac{1}{2}(z'-d)^2]/d^2 - \tfrac{1}{2}Q'_0 R_{z'}(t_1) \tag{4}$$

where X is the ratio $(k'b)/(kd)$. $G_z(t)$, $M_z(t)$, and $R_{z'}(t)$ are convenient functions obtained by imposing time continuity of the solutions and explicitly given in the mentioned paper [3]. Q_m and Q'_m are proportional to the heat fluxes at the specimen contacts with C_1 and C_2, respectively: more precisely,

$$\theta^{(1)}_{m+1} - \theta_{m+1}(0) = Q_m k/(bH) \tag{5}$$

$$\theta_{m+1}(b) - \theta'_{m+1}(0) = Q'_m k'/(Hd) \tag{6}$$

where $\theta^{(1)}$ is the temperature change with respect to the environment as recorded by the thermocouple T_1 inserted in C_1, and H is a parameter depending on the nature of the thermal contact between specimen and copper [4]. As shown in Refs. 1 and 2, when the specimen is low-conducting and the contact is established through a thin layer of a conducting glue, the parameter $\gamma = k/bH$ can be considered equal to zero. Inserting the previous expressions for $\theta_{m+1}(z)$ and $\theta'_{m+1}(z')$ into the boundary conditions given in Eqs. (5) and (6), one obtains a system of equations in Q_m and Q'_m which can be solved through a numerical procedure once a trial set of values (α, X) has been fixed. As in Ref. 2, we obtain the thermal expansion of the specimen and of the copper disk C_2 by integrating the temperature fields over z. In this way, we arrive at the following expressions for the dilation of the specimen and of copper:

$$d^{S}_{m+1} = \beta_S \Bigg[(Q_m - XQ'_m)(\alpha t_{m+1} + b^2/6)/b - (Q_0 - XQ'_0)b/6 + \tfrac{1}{2}b(Q_0 - Q_m) - \sum_{p=1}^{m} (\alpha t_p + b^2/6)(Q_p - XQ'_p - Q_{p-1} + XQ'_{p-1})/b + \tfrac{1}{2}b \cdot \sum_{p=1}^{m} (Q_p - Q_{p-1}) \Bigg] \tag{7}$$

$$d^{C}_{m+1} = \beta_{Cu} \Bigg[Q'_m(\alpha' t_{m+1} + d^2/6)/d - Q'_0 d/6 - \sum_{p=1}^{m} (Q'_p - Q'_{p-1})(\alpha' t_p + d^2/6)/d \Bigg] \tag{8}$$

for $m \geqslant 1$,

$$d^{S}_1 = \beta_S [(Q_0 - XQ'_0)\, \alpha t_1/b] \tag{9}$$

$$d^{C}_1 = \beta_{Cu} [Q'_0(\alpha' t_1/d)] \tag{10}$$

for $m = 0$, where β_S and β_{Cu} are the corresponding thermal expansion coefficients.

The capacitive cell D_1, D_2 detects the sum of the dilations of the specimen and of the copper disks C_1 and C_2, namely,

$$d_i = d^{S}_i + d^{C_2}_i + \beta_{Cu} h_1 \theta^{(1)}(t_i) \tag{11}$$

where h_1 is the thickness of copper disk C_1 and $\theta^{(1)}(t_i)$ is its temperature change as measured by thermocouple T_1 at time t_i.

Of course, the calculated value of d_i depends on the choice of the pair (α, X); the value of α' is assumed to be known and equal to $0.93\ \mathrm{cm}^2 \cdot \mathrm{s}^{-1}$ [5]. The values of β_{Cu} and β_{s} are assumed to be equal to 0.165×10^{-4} and $0.627 \times 10^{-6}\ \mathrm{K}^{-1}$, respectively [6]. Let us impose the best fit of d_i to the experimental change of gap $(d_i)_{\mathrm{exp}}$ by minimizing the expression

$$\delta = \sum_{i=1}^{N} [d_i - (d_i)_{\mathrm{exp}}]^2 \tag{12}$$

where i runs over all the times through which the whole time interval of measurement has been subdivided. This is easily done by exploring the behavior of δ in the plane (α, X). The values of α and X for which δ reaches its absolute minimum are taken as the correct experimental values of the above parameters.

3. EXPERIMENTAL PROCEDURE AND RESULTS

First of all, we performed the calibration of the capacitive cell by heating the copper disk C_0 and simultaneously recording the capacitive signal $V(t)$ and the temperature change $\theta^{(0)}(t)$ of C_0. As in Ref. 2, the signal $V(t)$ and the change of gap between the plates, d_{gap}, are assumed to be linked by a quadratic relation,

$$d_{\mathrm{gap}} = c_1 V + c_2 V^2 \tag{13}$$

where c_1 and c_2 are parameters to be determined.

In the calibration experiment d_{gap} is known and given by $\beta_{\mathrm{Cu}} h_0 \theta^{(0)}(t)$ (see Section 1). Consequently, from a best-fit analysis it is possible to obtain the parameters (c_1, c_2) by which one can relate the thermal expansion to the electric signal.

Once the system has reached its initial equilibrium conditions again, one proceeds to the second experiment by heating the disk C_1. An example of heating curves for a specimen of fused silica with a length of 1.00 cm is shown in Fig. 2. The height of the copper disks C_1 and C_2 was 1.00 cm. For a given choice of (α, X) we evaluate the sum square deviation

$$\delta = \sum_{i=1}^{N} [d_i - c_1 V_i - c_2 V^2]^2 \tag{14}$$

where d_i is given by Eq. (11) and V_i is the capacitive signal determined in this second experiment at time t_i. Exploring the behavior of δ as a function of α and X, one obtains a matrix of values like that shown in Table I, referring to a specimen of fused silica at 25°C. From Table I, it is

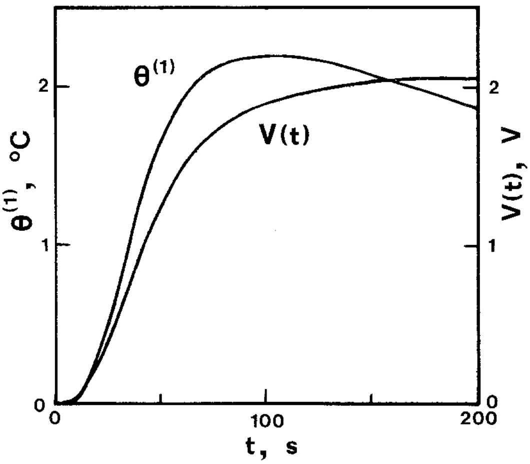

Fig. 2. Heating curves obtained for a sample of fused silica. $\theta^{(1)}$ is the temperature change of the copper disk C_1, and $V(t)$ the capacitive signal.

immediately possible to deduce the minimum value of δ, corresponding to the pair of values $(8 \times 10^{-3}, 290)$. By performing several measurements on the same specimen of fused silica, a repeatibility of about 5% in α, and about 3% in X, has been found. The mean values for α and X are $8.1 \times 10^{-3}\ \mathrm{cm}^2 \cdot \mathrm{s}^{-1}$ and 289, respectively. These values are in good agreement with those found in the literature [6] and also with those obtained by means of an experimental apparatus in which the dilatometric sensor was substituted by a thermocouple inserted in C_2 [3]. We also performed a set of measurements on a specimen of Zerodur and deduced a mean value of $10.2 \times 10^{-3}\ \mathrm{cm}^2 \cdot \mathrm{s}^{-1}$ for the diffusivity and 235 for X, in good agreement with the results obtained in Ref. 3.

Table I. Values of the Sum Square Deviation δ, Given by Eq. (14), as a Function of the Thermal Diffusivity α and of the Reduced Conductivity Ratio $X = k'b/kd$ for a Specimen of Fused Silica at Room Temperature[a]

	α				
X	6	7	8	9	10
270	0.141	0.062	0.314	0.725	1.207
290	0.635	0.149	0.029	0.106	0.287
310	1.435	0.655	0.263	0.084	0.159

[a] The values of the diffusivities are given in $10^{-3}\ \mathrm{cm}^2 \cdot \mathrm{s}^{-1}$.

One point to investigate is the uncertainty affecting our measurements of α and X arising from experimental uncertainties connected with the reading of data. Let us consider an ideal specimen with $\alpha = 10^{-2}\ \mathrm{cm}^2 \cdot \mathrm{s}^{-1}$, $X = 300$, and length $= 1$ cm (copper disk C_2 with a length of 1 cm) subjected to heating as shown in Fig. 2.

Let us also assume an ideal situation with $c_1 = 1$ and $c_2 = 0$. Under these conditions, we calculate the theoretical dilation and the corresponding signal function $V(t)$. By truncating each numerical value of this function to the first decimal place, we simulate the reading uncertainty of the experimental data coming from the recorder. The function obtained in this way can be considered, in our computer experiment, as an "experimental" function to which our numerical procedure of data analysis can be applied in order to obtain the values of α and X giving δ its absolute minimum. By comparing these values with those originally assumed for the two parameters, we find relative deviations of the order 1%. Therefore this computer experiment shows that part of the absence of repeatibility of our results can be attributed to uncertainties in the data, due to the instabilities of the whole electronic equipment, of the temperature of the specimen, and so on. Another part depends on the uncertainty affecting the determination of c_1 and c_2 in the calibration experiment (due, again, to the mentioned instabilities). In fact, if in the above computer experiment one gives c_1 and c_2 a resonable change of 1%, relative deviations of 4 and 2% in α and X are found.

In conclusion, the intrinsic accuracy of the present method is high, because the unwanted heat exchanges between specimen and environment are almost eliminated. The uncertainty of the measured values is essentially due to the presence of the thermal and electrical instabilities of the apparatus. Any reduction of these instabilities would be much more significant here than in other competitive methods, where a systematic error is always introduced by the temperature field detectors.

REFERENCES

1. M. Omini, A. Sparavigna, and A. Strigazzi, *Meas. Sci. Technol.* **1**:166 (1990).
2. M. Omini, A. Sparavigna, and A. Strigazzi, *Meas. Sci. Technol.* **1**:1228 (1990).
3. A. Sparavigna, M. Omini, A. Pasquarelli, and A. Strigazzi, *Int. J. Thermophys.*, in press (1992).
4. J. V. Beck, *J. Heat Trasfer* **110**:1046 (1988).
5. A. Sparavigna, G. Giachello, M. Omini, and A. Strigazzi, *Int. J. Thermophys.* **11**:1111 (1990).
6. Y. Shapira and D. Lichtman, in *Method of Experimental Physics, Vol. 14*, G. L. Weissler and R. W. Carlson, eds. (Academic Press, New York).