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Temperature Oscillation Techniques for Simultaneous Measurement of Thermal Diffusivity and Conductivity[1]

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Simple temperature ocillation techniques are described for the fast measurement of thermal diffusivity and conductivity of liquids. The liquid specimen is a slab bounded above and below by a reference material. Two Peltier elements mounted on the outer surfaces of the reference layers generate temperature ocillations of these surfaces. Temperature waves propagate through the reference layers into the specimen. The thermal diffusivity of the specimen is deduced by measuring and evaluating the amplitude attenuation and/or the phase shift between the fundamental temperature oscillations at the surface of the liquid specimen and at a well-defined position inside the specimen. If the thermal diffusivity of the specimen is known, the thermal conductivity is determined by the measured amplitude attenuation and/or the phase shift between the fundamental temperature oscillations at the surface of the reference layer and at the surface of the specimen. Slab and semi-infinite body geometries are considered. Measurement cells are designed and experiments are carried out with water, ethanol, heptane, nonane, and glycerine. The results of the measurements of thermal diffusivity agree very well, and those of thermal conductivity reasonably well, with the data obtained from the literature.

KEY WORDS: ethanol; heptane; nonane; periodic techniques, temperature ocillations; thermal conductivity; thermal diffusivity; water.

1. INTRODUCTION

Thermal diffusivity is the important thermophysical property to describe transient heat conduction in a solid or steady liquid. Therefore a non-steady-state measurement technique is applicable. The presented temperature

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oscillation technique, based on a previously proposed method, combines the advantages of a steady-state measurement with the possibility to measure a property describing a non-steady state [1–5]. With modifications this method can be used for simultaneous thermal conductivity meaurement. Earlier applications were made only for solid materials [5]. To measure thermal diffusivity and conductivity of fluids, convection must be avoided.

The background of temperature oscillation techniques and an automated measurement system are presented in this paper. To confirm the practical applicability, experiments are carried out with different liquids and geometries. By computerized operations the measurement can be performed without attendance and thus be used, for instance, as a quality control device in a production process.

2. MEASUREMENT PRINCIPLE

The energy equation

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T \tag{1}$$

describes heat conduction in an isotropic solid or liquid with constant thermal conductivity. Here T is temperature, t time, and α the thermal diffusivity. The solution of Eq. (1) depends upon specimen geometry and boundary conditions.

At the nonadiabatic surfaces of the specimen, periodic temperature oscillations are generated with the period t_p and the constant angular frequency

$$\omega = \frac{2\pi}{t_p} \tag{2}$$

2.1. Temperature Oscillations in a Semi-Infinite Body

In this case the initial and boundary conditions are independent of the coordinates y and z. Thus the temperature will be a function of x and t only. The differential equation (1) is expressed in dimensionless coordinates containing the constant thermal diffusivity α,

$$\xi = x \left(\frac{\omega}{2\alpha}\right)^{1/2} \tag{3}$$

Introduction of the dimensionless time

$$\tau = \omega t \tag{4}$$

gives

$$\frac{\partial^2 T}{\partial \xi^2} = 2\,\frac{\partial T}{\partial \tau} \tag{5}$$

with the boundary conditions (subscript M indicates mean; subscript 0 indicates $x = 0$)

$$\xi = 0: \qquad T(\xi = 0, \tau) = T_{\mathrm{M}} + u_0 \cos \tau \tag{6}$$

$$\xi = \infty: \qquad \left.\frac{\partial T}{\partial \xi}\right|_{\xi = 0} = 0 \tag{7}$$

For large values of time all transient disturbances caused by starting the oscillations fade away and the known steady periodic solution of this problem is

$$T(\xi, \tau) = T_{\mathrm{M}} + u_0 e^{-\xi} \cos(\tau - \xi) = T_{\mathrm{M}} + u_x \cos(\tau - \xi) \tag{8}$$

The phase difference ΔG and the corresponding amplitude ratio between the surface $x = 0$ and a well-defined position x are

$$\Delta G = x\left(\frac{\omega}{2\alpha}\right)^{1/2} \tag{9}$$

and

$$\frac{u_0}{u_x} = \exp[x(\omega/2\alpha)^{1/2}] \tag{10}$$

Measurement of the phase difference or the amplitude ratio allows the thermal diffusivity α to be determined from Eq. (9) or (10), respectively.

2.2. Temperature Oscillations Within the Reference Layer Covering the Semi-Infinite Body

To evaluate the thermal conductivity, the heat flow entering the specimen is needed. For that purpose, the specimen is covered by a reference layer. The amplitude attenuation and the phase shift within the reference layer depend on the heat flow which is extracted by the specimen.

The temperature distribution in the reference (R) layer is determined by the energy equation which is analogous to Eq. (1):

$$\frac{\partial T_{\mathrm{R}}}{\partial t} = \alpha_{\mathrm{R}} \nabla^2 T_{\mathrm{R}} \tag{11}$$

Introducing the dimensionless coordinate of the reference layer (thickness D)

$$\zeta = x\left(\frac{\omega}{2\alpha_R}\right)^{1/2} \tag{12}$$

and the dimensionless time accoding to Eq. (4) gives, analogous to Eq. (5),

$$\frac{\partial^2 T_R}{\partial \zeta^2} = 2\,\frac{\partial T_R}{\partial \tau} \tag{13}$$

with the condition at the contact surface between the reference layer and the specimen temperatures

$$T_R(\zeta = 0, \tau) = T(\xi = 0, \tau) \tag{14}$$

and the heat flow from the reference layer must be equal to that entering the specimen. Heat flow per unit area is given by

$$-\lambda_R\left(\frac{\omega}{2\alpha_R}\right)^{1/2}\frac{\partial T_R}{\partial \zeta}\bigg|_{\zeta=0} = -\lambda\left(\frac{\omega}{2\alpha}\right)^{1/2}\frac{\partial T}{\partial \xi}\bigg|_{\xi=0} \tag{15}$$

where λ is thermal conductivity.

The steady periodic solution of this problem gives the amplitude ratio and the phase difference between the two surface ($x = 0$ and $x = -D$) of the reference layer (subscript D indicates $x = -D$):

$$\Delta G_R = \arctan\left[\tan(\zeta_R)\,\frac{C + \tanh(\zeta_R)}{1 + C\tanh(\zeta_R)}\right] \tag{16}$$

$$\begin{aligned}\left(\frac{u_D}{u_0}\right)_R &= \{[\cos(\zeta_R)(\cosh(\zeta_R) + C\sinh(\zeta_R))]^2 \\ &\quad + [\sin(\zeta_R)(\sinh(\zeta_R) + C\cosh(\zeta_R))]^2\}^{1/2}\end{aligned} \tag{17}$$

with

$$\zeta_R = D\left(\frac{\omega}{2\alpha_R}\right)^{1/2} \tag{18}$$

and

$$C = \frac{\lambda}{\lambda_R}\left(\frac{\alpha_R}{\alpha}\right)^{1/2} \tag{19}$$

From the measured phase difference or the amplitude ratio between the two surfaces of the reference layer and the thermal diffusivity of the reference material, the constant C can be determined from Eq. (16) or (17), respectively. With the known thermal diffusivity from Eq. (9) or (10), the thermal conductivity of the specimen can be determined from Eq. (19).

2.3. Steady Temperature Oscillations in a Slab

A slab $(0 \leqslant x \leqslant L)$ is considered with two diathermic surfaces. On each side, periodic surface-temperature oscillations are generated with the same constant angular frequency [Eq. (2)], but with different amplitudes and phases. The mathematical formulation of this problem is given as (subscript L indicates $x = L$)

$$\frac{\partial^2 T}{\partial \xi^2} = \frac{\partial T}{\partial \tau} \tag{20}$$

$$T(\xi_0 = 0, \tau) = T_M + u_0 \cos(\tau + G_0) \tag{21}$$

$$T\left(\xi_L = L\left(\frac{\omega}{\alpha}\right)^{1/2}, \tau\right) = T_M + u_L \cos(\tau + G_L) \tag{22}$$

with

$$\xi = x\left(\frac{\omega}{\alpha}\right)^{1/2} \tag{23}$$

The use of Laplace transform techniques yields the steady priodic solution of Eqs. (20)–(22). For convenience the complex solution is presented:

$$T^*(\xi, \tau) = T_M + \frac{u_L e^{iG_L} \sinh(\xi \sqrt{i}) - u_0 e^{iG_0} \sinh(\sqrt{i}(\xi - \xi_L))}{\sinh(\xi_L \sqrt{i})} e^{i\tau} \tag{24}$$

The complex amplitude ratio B^* between the points $x = L/2$ and $x = L$ becomes

$$B^* = \frac{2u_L e^{iG_L}}{u_L e^{iG_L} + u_0 e^{iG_0}} \cosh\left[\frac{L}{2}\left(\frac{i\omega}{\alpha}\right)^{1/2}\right] \tag{25}$$

The real phase difference ΔG and the real amplitude ratio are expressed as

$$\Delta G = \arctan\left(\frac{\mathrm{Im}[B^*]}{\mathrm{Re}[B^*]}\right) \tag{26}$$

and

$$\frac{u_L}{u_{L/2}} = \{(\mathrm{Re}[B^*])^2 + (\mathrm{Im}[B^*])^2\}^{1/2} \tag{27}$$

From a measurement of the phase or the amplitude at the two sides and in the center of the slab the thermal diffusivity α can be determined from Eqs. (23), (26) or (23), (27), respectively.

2.4. Temperature Oscillations Within the Reference Layer Covering the Slab

Consideration of the reference layer and of the generating temperature oscillations at the surface of the reference layer at the top and at the bottom surface of the specimen with the same constant angular frequency, but with different amplitudes and phases (see Fig. 1), gives the mathematical formulation of this problem as follows:

$$\frac{\partial^2 T_R}{\partial \zeta^2} = \frac{\partial T_R}{\partial \tau} \tag{28}$$

with

$$\zeta = x\left(\frac{\omega}{\alpha_R}\right)^{1/2} \tag{29}$$

and with the boundary conditions

$$T_R(\zeta = 0, \tau) = T(\xi = 0, \tau) \tag{30}$$

$$-\lambda_R\left(\frac{\omega}{2\alpha_R}\right)^{1/2} \frac{\partial T_R}{\partial \zeta}\bigg|_{\zeta=0} = -\lambda\left(\frac{\omega}{2\alpha}\right)^{1/2} \frac{\partial T}{\partial \xi}\bigg|_{\xi=0} \tag{31}$$

The steady periodic solution in the domain of $0 < x < L$ is analogous to Eq. (24). The solution in the domain of $-D < x < 0$ is given as

$$T_R^*(\zeta, \tau, \xi_L) = T_m + u_0 e^{i(\tau + G_0)} \cosh(\zeta\sqrt{i}) + C[u_R e^{i(\tau + G_R)} - u_0 e^{i(\tau + G_0)} \cosh(\xi_L\sqrt{i})] \frac{\sinh(\zeta\sqrt{i})}{\sinh(\xi_L\sqrt{i})} \tag{32}$$

where C is given by Eq. (19).

The complex amplitude ratio B_R^* between the points $x = -D$ and $x = 0$ becomes

$$B_R^* = \cosh(\zeta_D\sqrt{i}) - C\sinh(\zeta_D\sqrt{i})\left[\frac{(u_L/u_0)\, e^{i(G_L - G_0)} - \cosh(\xi_L\sqrt{i})}{\sinh(\xi_L\sqrt{i})}\right] \tag{33}$$

The real phase difference ΔG_R and the real amplitude ratio are expressed by

$$\Delta G_R = \arctan\left(\frac{\mathrm{Im}[B_R^*]}{\mathrm{Re}[B_R^*]}\right) \tag{34}$$

and

$$\frac{u_D}{u_0} = \{(\mathrm{Re}[B^*])^2 + (\mathrm{Im}[B^*])^2\}^{1/2} \tag{35}$$

The thermal diffusivity of the reference material and the specimen and the conductivity of the reference material are known. From measurement of the phase or the amplitude at both sides of the reference layer and of the other side of the specimen, the thermal conductivity of the specimen can be determined from Eqs. (33), (34) or (33), (35), respectively.

2.5. Evaluation of Measurements

Periodic temperature ocillations are generated at the surface of the reference layer by means of Peltier elements fed with a periodically oscillating electric voltage.

Every arbitrarilly shaped but strictly periodic temperature oscillation can be described mathematically by a Fourier series:

$$T(\tau) = \frac{a_0}{2} + \sum_{k=1}^{\infty} A_k \sin(k\tau + G_k) \tag{36}$$

with

$$a_k = \frac{1}{\pi}\int_0^{2\pi} T(\tau)\cos(k\tau)\,d\tau, \qquad k = 0, 1, 2, \dots \tag{37}$$

$$b_k = \frac{1}{\pi}\int_0^{2\pi} T(\tau)\sin(k\tau)\,d\tau, \qquad k = 1, 2, \dots \tag{38}$$

and

$$A_k = (a_k^2 + b_k^2)^{1/2}, \qquad \tan G_k = \frac{a_k}{b_k} \tag{39}$$

Thus the measured temperature oscillations are regarded as a superposition of several sinusoids of different frequencies, amplitudes, and phases. Each one represents a solution according to Eqs. (8), (24), and (32). In the experiments the fundamental oscillation ($k = 1$) is considered.

Amplitude and phase are evaluated from Eq. (39) by numerical integration according to Eqs. (37) and (38). Applied to the measured temperature at the surfaces of the reference layer and at the center or a well-defined position of the specimen, this yields the "measured" values of the phase difference and the amplitude ratio.

One-dimensional thermal conductivity is achieved by adiabatic surfaces of the frame. For the semi-infinite body, the amplitude of the temperature oscillation diminishes according to Eq. (10). At the distance of one thermal wavelength

$$\Lambda = 2\pi \left(\frac{2\alpha}{\omega}\right)^{1/2} \tag{40}$$

the amplitude is reduced by a factor of $\exp(-2\pi) = 0.0019$; thus the waves are very strongly attenuated. This implies that the solution for the semi-infinite specimen can be used for a specimen whose thickness is greater than one or two wavelengths.

3. EXPERIMENTS

Figure 1 shows a simplified scheme of the measurement cell with specimen bounded by two parallel reference layers. The liquid is filled into the flat cylindrical hollow space (diameter × thickness: 50 mm × 6 mm), formed by the two reference discs (diameter × thickness: 50 mm × 8 mm) made of stainless steel and a circular frame made of plexiglass. Filling and deaerating are done via the small tubes in the frame. The temperatures at the center of the specimen and at both surfaces of the reference layers are measured by means of Ni–CrNi thermocouples (diameter 0.1 mm), which are located on the centerline. The outside thermocouples are placed in

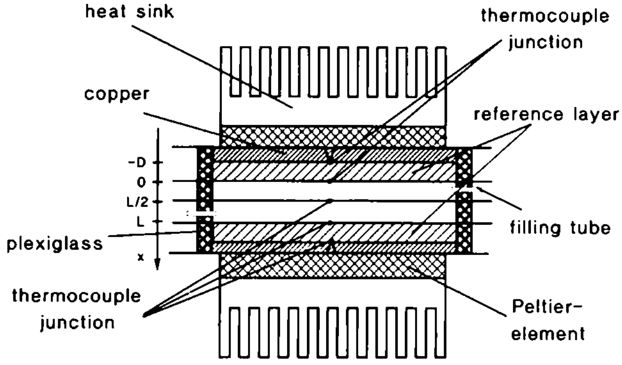

Fig. 1. Schematic diagram of the measurement apparatus.

a groove on the surfaces of the copper plates, and the thermocouple junctions are butt-welded on both sides of the reference layers. The temperatures of the two copper plates are changed periodically by means of two Peltier elements (square: 40 mm × 40 mm). To improve heat transfer, finned plates or cooled heat sinks are mounted on top of the Peltier elements.

It is possible to investigate solid specimen by replacing the frame by two identical solid plane spcimens with a thermocouple between them. If the upper copper plate and the upper reference layer are removed and the frame is extended, the specimen geometry is changed into a semi-infinite body.

The amplitude of the temperature oscillation inside the specimen is less than 1 K. To avoid convection, the measurement cell is turned in a vertical position. As one can see from Eqs. (3) and (23), the sensitivity coefficient of the thermal diffusivity on the frequency is constant. With decreasing temperature amplitudes, the error of the amplitude measurement increases. Thus the frequency is chosen so that the amplitude attenuation is about 0.5. To decide about geometry and amplitude or phase difference measurement, the sensitivity coefficients of the dimensionless space coordinate on the amplitude and phase difference of each geometry were studied. To determine the uncertainty of the thermal conductivity measurement, the uncertainty of the thermal properties of the reference material (up to 5%) must be added. The use of the slab and the phase difference measurement show the smallest measurement uncertainty. The advantage of the semi-infinite body is better filling and cleaning, especially for viscous liquids.

4. RESULTS

Experiments were carried out with five liquids and compared with values from the literature. To reduce the random error, the temperature was measured over two periods and the integration range of Eqs. (37) and (38), was shifted. Table I shows the measured thermal diffusivity and conductivity of all measurements with the highest deviation from values available from the literature [7]. The slab along with the phase difference equations were used to obtain the results given in Table I.

The measurement uncertainties depend on the various experimental parameters and on the measured medium. Of the liquids studied, only water has reliable values reported in the literature over a wide temperature range. The magnitudes of the measurement uncertainties of the semi-infinite body of thermal diffusivity are less than 3.5%, and of thermal conductivity less than 10%. The magnitudes of the measurement uncertainties of the

Table I. Experimental Results on Different Specimens

	Water	Ethanol	Heptane	Nonane	Glyerine
Diffusivity (10^{-8} $m^2 \cdot s^{-1}$)	14.12	9.323	8.538	8.673	9.705
Deviation from ref. 7 (%)	1.5	1.8	2.0	1.7	1.2
Conductivity ($W \cdot m^{-1} \cdot K^{-1}$)	0.6244	0.1780	0.1351	0.1324	0.2713
Deviation from ref. 7 (%)	4.3	2.9	7.9	0.3	5.1
Period (s)	100	100	150	150	200
Temperature (°C)	20	20	15	20	20

slab of thermal diffusivity are less than 2%, and of thermal conductivity less than 8%.

The most significant effects on the measurement uncertainties are thermocouple location, contact resistance of the thermocouples, and uncertainty of the thermophysical properties of the reference layer. The onset of free convection can be detected by the deformation of the sinusoidal temperature ocillations in the liquid. To reduce measurement uncertainty, the thermocouples are located in an isothermal plane.

5. CONCLUSIONS

The apparatus described perform very well. They can be used for absolute measurements slightly above or below ambient temperatures. The experiments have demonstrated the applicability of the method proposed for simultaneous determination of thermal diffusivity and conductivity. The slab apparatus achieves the best performance and can easily be modified for measurements at pressures different from ambient pressure. It the finned plates are thermostated, larger temperature differences from ambient temperature are possible.

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