THERMAL CONDUCTIVITY OF TRANSPARENT LIQUIDS

A Non-Steady-State Method of Measuring the Thermal Conductivity of Transparent Liquids

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A plane source method has been developed for non-steady-state measurements of the thermal conductivity of transparent liquids. The plane source is realized by using an electrically heated metal foil, suspended in the liquid. The temperature distribution is described by using the concept of instantaneous heat sources giving a simple expression of the optical path, which is recorded with wave-front-shearing interferometry. The thermal diffusivity is determined simply by measuring the positions of the fringes, locating the points where the gradient of the optical path is constant and knowing the times between two subsequent exposures. The accuracy of the method, which is an absolute one, is demonstrated. The average deviation from the recommended values of the thermal conductivity was found to be 0.37%.

I. Introduction

In recent years the non-steady-state measurements of thermal conductivity have proved to be a reliable technique used in precise work. The experimental arrangements are generally restricted to hot wire cells, where the change of temperature is observed in an electrically heated wire. Another possibility is to record the temperature in the liquid close to the source of heat. This is done by some experimenters with optical methods, which have the appealing advantage of not affecting the temperature distribution. There is, however, one particular disadvantage with the optical technique, when applied to hot wire cells, since in order to get the necessary optical path difference the temperature must be raised substantially above the initial temperature. For water the increase may be as high as one degree. The aim of the present work is to increase the sensitivity of optical methods without changing the accuracy. The most natural way of doing this is to extend the optical path in the liquid by using a continuous plane source of heat instead of a line source. This is done by utilizing a metal foil which is heated by a constant electrical current.

If it is assumed that we need the same output of power per unit length of the foil as with the hot wire, we would with the foil gain at least two things: firstly, the temperature increase at a particular point will be less and secondly the voltage drop between two points in the liquid can be kept at a low level because of the larger cross section of the foil, which is important when investigating conducting liquids.

2 F. M. VAN DER HELD and F. G. VAN DRUNEN, Physica 15, 865 [1949].
3 O. BRYNGDAHL, Arkiv Fysik 21, 289 [1962].
II. Theory of the Method

In order to describe the temperature distribution around the "hot foil" it is necessary to solve the differential equation of conduction of heat in three dimensions with the proper boundary conditions. This is most readily done by using the idea of the instantaneous point source which has proved most useful in the theory of conduction of heat. We take the solution

\[ v = \frac{Q}{8(\pi \kappa t)^{1/2}} \exp\left[ -\left( \frac{(x-x')^2 + (y-y')^2 + (z-z')^2}{4 \pi \kappa t} \right) \right] \]  

as fundamental. It may be interpreted as the temperature in an infinite solid due to an instantaneous liberation of a quantity of heat \( Q \) at a point \((x', y', z')\) and time \( t = 0 \). \( \kappa \) is the thermal diffusivity of the solid. By integrating the fundamental solution with regard to the appropriate variables one immediately obtains the desired temperature distribution. If we assume that the \( y \)- and \( z \)-axes in a system of orthogonal coordinates define the plane which contains the foil we get the solution of an instantaneous plane source by integrating equation (1) throughout the region \(-h \leq z \leq h\) and \(-d \leq y \leq d\), where \( 2h \) is the height and \( 2d \) is the width of the foil. With a constant heat liberation per unit area per unit time \( Q = q_c \rho \), we can integrate the obtained solution with regard to \( t' \) and get the solution of a continuous plane source.

\[ T(x, y, z, t) - T_0 = \frac{q}{8(\pi \kappa t)^{1/2}} \int_0^t dt' \exp\left[ -\left( \frac{y-y'}{\kappa t} \right)^2 \right] \int_{-d}^d dy' \exp\left[ -\left( \frac{y-y'}{\kappa (t-t')} \right)^2 \right] \cdot \int_h^{2h} dz' \exp\left[ -\left( \frac{z-z'}{\kappa (t-t')} \right)^2 \right] \]  

where \( T_0 \) is the initial temperature in the liquid. Rearranging equation (2) we get

\[ T(x, y, z, t) - T_0 = \frac{q}{8(\pi \kappa t)^{1/2}} \int_0^{2(\kappa t)^{1/2}} d\sigma \exp\left( -\frac{x^2}{\sigma^2} \right) \left[ \text{erfc}\left( \frac{y-d}{\sigma} \right) - \text{erfc}\left( \frac{y+d}{\sigma} \right) \right] \left[ \text{erfc}\left( \frac{z-h}{\sigma} \right) - \text{erfc}\left( \frac{z+h}{\sigma} \right) \right] \]  

where

\[ \text{erfc} u = \frac{2}{(\pi)^{1/2}} \int_u^\infty \exp\left( -\xi^2 \right) d\xi = 1 - \text{erf} u, \]  

which is a tabulated function.

When using an optical method to study the temperature distribution it is possible to choose a particular plane where the optical path is to be studied. To make it simple we look at the \( xy \)-plane in the middle of the foil. This makes it possible to get the factor \( 2 \text{erfc}\left( \frac{h}{4 \pi \kappa \Theta} \right) \) outside the integral in equation (3) with \( 0 < \Theta < t \). Since the time of an experiment is restricted to about 30 seconds because of the onset of convection we assume an experimental arrangement where \( h \) is big enough to make \( \text{erfc}\left( \frac{h}{4 \pi \kappa \Theta} \right) \) close to unity. If we put \( \Theta = 30 \text{ sec}, \kappa = 0.146 \text{ mm}^2/\text{sec}, \) and \( h = 13 \text{ mm}, \) the error is less than \( 10^{-5} \) and the temperature in the neighborhood of \( z = 0 \) becomes

\[ T(x, y, t) - T_0 = \frac{q}{4 \pi (\kappa t)^{1/2}} \int_0^{2(\kappa t)^{1/2}} d\sigma \exp\left( -\frac{x^2}{\sigma^2} \right) \left[ \text{erfc}\left( \frac{y-d}{\sigma} \right) - \text{erfc}\left( \frac{y+d}{\sigma} \right) \right]. \]  

This equation is strictly valid only if there is no heat transport along the foil because we assumed a distribution of line sources in the \( yz \)-plane. This should be a rather good approximation if the foil is very thin and of constant thickness. By computing \( \frac{T(0, 0, t) - T(0, d, t)}{d} \) we get an estimation of the temperature gradient in the foil and then it is easy to get the amount of heat which is transported along the "hot foil". It turns out that the ratio of this heat flow and the total liberation of heat per unit length is of the order of \( 10^{-3} \) when using a silver foil and \( 10^{-4} \) when using a platinum foil with a thickness of


8 × 10⁻³ mm. This means that equation (4) is a very good approximation of the temperature distribution in the liquid. It may be mentioned here that the problem of the isothermal foil has been solved by McLachlan.\(^\text{7}\)

III. Optical Analysis

In order to interpret the fringe pattern we need the optical path of each ray passing through the liquid in the neighborhood of the heated foil. The refractive index \(\mu\) is introduced with the expression \(\mu - \mu_0 = \left[T(x, y, t) - T_0\right] \cdot \partial \mu / \partial T\), where \(\mu_0\) is the refractive index of the liquid at the temperature \(T_0\). \(\partial \mu / \partial T\) can be considered a constant over the limited temperature range of an experiment. The optical path is now given by an integration of \(\mu - \mu_0\) with regard to \(y\), which gives

\[
R(x, t) = \frac{q}{4 \cdot \pi \cdot \sqrt{\left(\frac{\partial \mu}{\partial T}\right)}} \int_{-l}^{l} dy \int_0^2 \text{d}\sigma \exp\left(-\frac{x^2}{\sigma^2}\right) \left[\text{erfc}\left(\frac{y-d}{\sigma}\right) - \text{erfc}\left(\frac{y+d}{\sigma}\right)\right]
\]

where \(2l\) is the total extension of the cell enclosing the foil.\(^\text{7a}\) This equation may be transformed into the form

\[
R(x, t) = \frac{q \cdot d}{\pi \cdot \sqrt{\left(\frac{\partial \mu}{\partial T}\right)}} \cdot \left[1 + g(t)\right] \int_0^2 \text{d}\sigma \exp\left(-\frac{x^2}{\sigma^2}\right)
\]

where

\[
g(t) = \frac{\tau}{2 d \cdot (\pi)^{1/2}} \left[\exp\left(-\frac{(l+d)^2}{\tau}\right) - \exp\left(-\frac{(l-d)^2}{\tau}\right)\right] + (\pi)^{1/2} \left(\frac{l-d}{\tau}\right) \text{erfc}\left(\frac{l-d}{\tau}\right)
\]

\[
- (\pi)^{1/2} \left(\frac{l+d}{\tau}\right) \text{erfc}\left(\frac{l+d}{\tau}\right).
\]

\(\tau = (4 \cdot \Theta)^{1/2}\) and \(0 < \Theta < t\). As we have pointed out above the time is restricted to about 30 seconds due to the onset of convection and this means that with a properly designed cell \((l-d)/\tau\) is a relatively large number. Using an expansion of the error function for large values of the argument in equation (7) it is easy to show that

\[
|g(t)| < \frac{\tau}{d \cdot (\pi)^{1/2}} \left(\frac{l-d}{\tau}\right)^{-2} \cdot \exp\left(-\frac{(l-d)^2}{\tau}\right).
\]

If we put \(\tau = 0.146 \text{ mm}^2/\text{sec}, \Theta = t, t = 30 \text{ sec}, d = 2 \text{ mm},\) and \(l-d = 15 \text{ mm}\), which is the least favourable situation in a typical experiment we get \(|g(t)| < 10^{-6}\), which shows that it can be neglected in equation (6) giving

\[
R(x, t) = 2 \cdot d \cdot q \cdot \left(\frac{\partial \mu}{\partial T}\right) \cdot (t/\pi \cdot \tau) \cdot \left[\exp\left(-\frac{x^2}{4 \cdot \tau \cdot \pi}\right) - \left(\frac{x}{2 \cdot \tau}\right) \cdot \text{erfc}\left[\frac{x}{(4 \cdot \tau)^{1/2}}\right]\right]
\]

and

\[
dR/dx = - \frac{q \cdot d}{\pi} \cdot \left(\frac{\partial \mu}{\partial T}\right) \cdot \text{erfc}\left[\frac{x}{(4 \cdot \tau)^{1/2}}\right].
\]

The gradient of the optical path difference becomes independent of the width of the heat source because \(2q \cdot d\) is the heat liberation per unit length of the foil and this is important from an experimental point of view, since the determination of the thermal conductivity will not be affected by a possible error in this quantity. Another thing which should be noted is that the equations (8) and (9) are applicable to the extreme cases of an infinite plane source and a line source, that is when \(d\) approaches infinity or zero, because the same expressions of the optical path difference apply in these two cases.\(^3, 5\)

\(^7\) N.W. McLachlan, Phil. Mag. 36, 600 [1945]; 37, 216 [1946].

\(^{7a}\) This is an exact expression of the optical path even if the heat transport along the foil would be considerable because it is irrelevant where the “line sources” are situated in the \(yz\)-plane when computing the optical path.
IV. Apparatus and Technique

In order to realize as closely as possible a plane source we used foils made of silver, with the approximate dimensions: length 190 mm, width 12 and 60 mm (see Table 1) and a thickness of 0.0084 mm. The problem of supporting such a thin metal foil in the liquid, was solved by passing it over two electrically insulated cylinders attached to copper frames as shown in Fig. 1. The electrical joints above and below the cylinders were made by soldering the foil to heavier pieces of silver which were connected via a resistor to a storage battery. The cell used in the experiments consisted of two concentric copper cylinders with a diameter of 130 and 160 mm, put together to form an annulus with connections to a circulating water bath. The cell windows were of interferometer quality and could be adjusted to a parallelism within ± 2 minutes of arc. The temperature was measured with a Beckmann thermometer, and the drift was always kept below ± 0.005 degrees over a period of several hours before a measurement was taken.

The optical system used in these experiments was a wave-front-shearing interferometer described by Bryngdahl in his paper on thermal conductivity 3. However, as a light source we used a 1.0 milliwatt helium-neon gas laser with a wavelength of 6328 Å. The laser light made the interference fringes (Fig. 2 *) much narrower, which markedly improved the reproducibility of the measurements. The recording system consisted of a Nikon F camera, which automatically fed the film and could be electrically triggered with a microswitch and a synchronous motor. During the evaluations of the interferograms which were done with an ordinary comparator, we used the fact that for one particular fringe the derivative \( (dR/dx) \) is always constant. This means that the corresponding \( x^2 \)-values must be a linear function of the time \( t \) according to Eq. (9). We made plots of \( x^2 \) versus \( t \) for the three uttermost fringes (the distance between two symmetrical fringes equals \( 2x \)) and then determined the slope of these lines. Then we used the fact that for three consecutive fringes

\[
(dR/dx)_{x_1} - (dR/dx)_{x_2} = (dR/dx)_{x_2} - (dR/dx)_{x_3}
\]

or from equation (9)

\[
\text{erfc}[x_1/(4 \times t)^{1/2}] + \text{erfc}[x_3/(4 \times t)^{1/2}] - 2 \text{erfc}[x_2/(4 \times t)^{1/2}] = 0.
\]

\( x \) can now be determined by iteration from this relation. This technique is very satisfactory because it is not necessary to know either the output of heat or the value of \((\overline{\mu}/\overline{\Sigma})\), and it is actually possible to determine the thermal diffusivity by measuring the fringe separations and the times between subsequent exposures, which should be compared with the dimension of \( \chi \) (m²/sec).

V. The Influence of the Thickness of the Heater

When deriving the formula (4) we had to assume that the thickness of the heater was zero. It is possible to estimate this approximation by considering the extra heat flow which must be supplied in order to raise the temperature of the foil itself. The additional heat production which is necessary becomes

\[
Q_I = 2 \nu c_f \rho \frac{dT}{dt} x = \nu,
\]

where \( 2 \nu \) is the thickness of the foil, and the index \( f \) refers to the foil. Using equation (4) we get

\[
\left(\frac{dT}{dt}\right)_{x = \nu} = \frac{Q}{4c_p(\pi \times t)^{1/2}} \exp \left(-\frac{\nu^2}{4 \times t}\right) \cdot \left[\text{erfc} \frac{y-d}{(4 \times t)^{1/2}} - \text{erfc} \frac{y+d}{(4 \times t)^{1/2}}\right].
\]

This expression is a function of \( \nu \) and is about twice as high in the middle of the foil as at the edge. Taking the least favourable value of \( \nu \) and is about twice as high in the middle of the foil as at the edge. Taking the least favourable value of \( (dT/dt)_{x = \nu} \) we get

\[
Q_f/Q = \frac{c_f \rho \nu}{c_p \rho (\pi \times t)^{1/2}} \exp \left(-\frac{\nu^2}{4 \times t}\right) \cdot \left\{1 - \text{erfc}\left(\frac{d}{(4 \times t)^{1/2}}\right)\right\}
\]

which is a decreasing function of time. After 1 second \( Q_f/Q \) is already below \( 3.7 \times 10^{-3} \), where we have used the following numerical values: \( \nu = 4.3 \times 10^{-3} \) mm, \( \chi = 0.146 \) mm²/sec, \( \rho = 1 \) g/cm³, \( c_p = 10.5 \) g/cm³, \( c_f = 4.18 \) Joules/g.deg., and \( c_I = 0.23 \) Joules/g.deg. It means that the power needed to raise the temperature of the foil is negligible after one

* Fig. 2 on p. 1008 a.
Fig. 2. Interference fringes from run no. 6.

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second and the only impact this may have is a shifting of the time scale \( \tau \), which is irrelevant with the evaluation technique used in these measurements, where just the times between two successive exposures are necessary.

**VI. Effect of Radiation from the Foil**

The heat loss per unit area of the foil due to radiation into a “blade” enclosure is

\[
Q_r = \bar{\sigma} (T_1^4 - T_2^4)
\]

where \( \bar{\sigma} \) is the Stefan-Boltzmann constant. Since \( T_1 \approx T_2 \) we get approximately

\[
Q_t = 0.08 \bar{\sigma} T^3 \Delta T,
\]

where \( \Delta T \) may be estimated from equation (4) by putting \( x = 0 \). \( \Delta T \) is then of course a function of \( y \) and to get a mean value \( \Delta T \) we take an average over \( -l \leq y \leq l \), which gives

\[
\Delta T = q (t/\pi \kappa)^{1/8} \left[ 1 - \text{erfc} [l/(\pi \Theta t)^{1/2}] + (\pi \Theta t/\pi \kappa)^{1/8} \exp(-l^2/\Theta t) - 1 \right]
\]

or with typical values of \( l, t \) and \( \kappa \)

\[
\Delta T = \frac{Q}{Q_{cp}} (t/\pi \kappa)^{1/8}
\]

and finally

\[
Q_t/Q = 0.08 \frac{\bar{\sigma} T^3}{Q_{cp}} (t/\pi \kappa)^{1/8}.
\]

At the end of the longest runs, when \( t = 20 \) seconds \( Q_t/Q \) is less than \( 2 \times 10^{-4} \), which shows that the effect of radiation can be neglected.

**VII. The Increase of Temperature in the Liquid**

At the beginning of an experiment the temperature is uniform throughout the cell, but during the measurement there is an increase of temperature, which is biggest in the immediate vicinity of the foil. One can get the temperature in the foil from equation (10), where we have put \( x = 0 \). This temperature is obviously too high if the thermal diffusivity would be refered to it, since use is only made of the three uttermost fringes in the determination of \( \kappa \). In order to get an exact value it is necessary to take an average along the path of the light corresponding to each fringe and then compute the temperature by using an iteration technique. A less elaborate way is to take an average in the following manner

\[
\Delta T_x = t^{-1/2} \int_0^t \left[ x_3 (x_3 - x_1)^{-1} x_2 \right] \frac{dx}{x_1}
\]

where \( t_x \) is the total time of an experiment. Taking into account the actual extension of the temperature distribution as shown above we get

\[
\Delta T_x = \frac{q (t_x/\pi \kappa)^{1/8} \left[ \frac{1}{2} (1 + 2 \xi_1^2) \text{erfc} \xi_1 - \frac{1}{2} (1 + 2 \xi_3^2) \text{erfc} \xi_3 - (\pi)^{-1/2} \xi_1 \exp(-\xi_1^2) + (\pi)^{-1/2} \xi_3 \exp(-\xi_3^2) \right]}{6 \bar{\sigma} T^3/\pi \kappa}
\]

where \( x = 2 \xi (\pi \kappa t)^{1/4} \). \( T_0 + \Delta T_x \) is then the mean temperature of the experiment.

The temperature increase becomes on the average 0.12 degrees, where we have used a cellwidth \( l = d + 15 \) mm. A temperature correction which is less than 0.2 degrees can eventually be neglected in routine work because this means a shift in the thermal conductivity of about 0.05 percent for water an this is well below the experimental error.

**VIII. Onset of Convection**

When a temperature gradient is applied to a liquid at rest, the resulting density gradient produces an instability which eventually leads to a convective motion in the fluid. This problem has been studied a great deal but is still far from being fully understood. In the steady-state techniques the absence of convective motion is generally deduced from critical values of the Rayleigh number

\[
R = g \rho \alpha \delta \Delta T \delta^3/\eta \kappa
\]

where \( g \) is the gravity, \( \rho \) the density, \( \alpha \) the thermal expansion, \( \eta \) the viscosity, \( \kappa \) the thermal diffusivity, \( \Delta T \) the temperature difference, and \( \delta \) is a characteristic dimension. One of the problems in the steady-state techniques is to minimize this dimension of the apparatus and thus keep \( R \) below its critical value. In the case of vertical cylinders it has been shown that convection is not important until \( R \) reaches a value of about 1000, when \( \delta \) is chosen as the thickness of the fluid. Applying this to hot wire cells VAN DER HELD and VAN DRUNEN suggested that to the instantaneous temperature distribution in the non-steady-state, it is possible to find a pseudo-steady-state situation and convection is likely to
occur when this pseudo-steady-state reaches a critical value of \( R = 1000 \). The reason why it is necessary to introduce the concept of pseudo-steady-state is that one needs some estimation of the radius of the expanding high temperature fluid which surrounds the heating wire. The \( \delta \)-value obtained by using this assumption turns out to be \( 1.499(\pi t)^{1/4} - r_0 \), where \( r_0 \) is the radius of the wire and can usually be neglected.

When trying to approach the convection situation with the hot foil one must realize the very complex situation because here we are dealing with a two dimensional temperature distribution. It is thus necessary to keep in mind the qualitative nature of the following treatment. To get some estimation of the thickness of the expanding high temperature liquid we take a mean value of the extension of the temperature field by considering \( x \) as a function of \( T \).

The average becomes

\[
\Delta x(y, t) = \frac{1}{T_t - T_0} \int_{T_0}^{T_t} T(x, t) \, dT
\]

It turns out that \( \Delta x(y, t) \) is rather independent of \( y \) except when \( y = 0 \) or take an average of \( \Delta x(y, t) \) from \( y = -d \) to \( y = d \) and assume that \( d/(\pi t)^{1/4} \) is so large that \( \text{erfc}[d/(\pi t)^{1/4}] \) is negligible compared to 1. If \( \delta \) is then chosen as 2 \( \Delta x \), the instantaneous temperature field is approximated by a linear distribution with an extension equal to \( \delta \) and which has the same average extension as the actual one. Introducing this value of \( \delta \), which becomes \( (\pi t)^{1/4} \), into the Rayleigh criterium (11) and putting \( \Delta T = (Q/g \, c_p) \cdot (t/\pi \, x)^{1/4} = T_t - T_0 \) we get an explicit expression for the time of onset of convection

\[
t_c = \left[ R \eta \, c_p \over \pi \, g \, x \, Q \right]^{1/4}.
\]

This equation seems to give the right dependence of \( t_c \) on \( Q \) and choosing \( R = 930 \) the experimental and calculated values are in fair agreement (Table 1).

One thing that should be noted is that the characteristic dimension is of the same form \( C(\pi t)^{1/4} \), where \( C \) is some constant, in the cases discussed above. This fact indicates a possibility to choose \( (\pi t)^{1/4} \) as the characteristic dimension in the different non-steady-state techniques and disregard all attempts to physically justify the choice of \( \delta \). In that case one would just have different critical values of the Rayleigh number depending upon the geometrical shape of the heat source.

**IX. Results and Discussion**

In order to test the method and the apparatus designed, a series of measurements of the thermal conductivity of water were made which are shown in Table 1. The purity of the distilled water was checked by a determination of the refractive index giving \( \mu_0 = 1.33396 \) at 25°C and 5461 Å. To get an idea about the reproducibility of the technique we chose to make a number of experiments at approximately the same temperature and as can be seen from the table the average deviation from the recommended values is 0.37%. According to the theoretical treatment, the optical path difference does not depend on the width of the foil, and in order to test this experimentally, we made measurements with two different \( d \)-values 6 mm and 30 mm. When \( d \) approaches zero the theory predicts a temperature distribution corresponding to that of the hot wire cell and this case is previously investigated. As can be seen from the table, there is no change in the measured thermal conductivity when using foils of different geometries, but it should be mentioned that the determination of the fringe separations is not so critical when using a wide foil because the time scale is extended, and it is easier to get good accuracy of the slope in the plots of \( x^2 \) versus \( t \). The power output per unit length was also varied to see the possible influence on the experimental data.

The onset of convection limits the time of the measurement which reduces the problem of long time thermostatic control. Comparing this technique where the temperature gradient is recorded with the methods where an absolute value of the temperature is obtained, it is evident that the temperature control is even less important because a small drift of the temperature may be tolerated as long as the gradient in the liquid is negligible.

In this new method it is very easy to obtain the thermal diffusivity without resort to constants of the apparatus (the magnification being omitted) or properties of the liquid, however, in order to get the thermal conductivity \( \lambda \) it is necessary to know the specific heat and the density. It appears when using equation (9) that it is possible to compute \( \lambda \) if the
Table 1. Experimental data. The width and the thickness of the foil were 60.3 mm and 8.48 \times 10^{-3} \text{ mm} in the experiments 1 through 7, and 12.05 mm and 8.26 \times 10^{-3} \text{ mm} in the experiments 8 through 11. Some of the runs were interrupted after 20 sec, when there was no sign of convection.

The aim of this work was to improve the sensitivity of the optical methods by extension of the optical path difference in non-steady-state measurements. If one would try to extend the foil further then to a width of say 100 mm it would probably be difficult to suspend it in the liquid, and some kind of multiple reflection cell seems more realistic from a practical point of view. This possibility is now being investigated.

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