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APPLICATION OF A MOMENTS METHOD AND OF LAPLACE TRANSFORMS TO HEAT TRANSFER EXPERIMENTS*

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We present a mathematical method based on Laplace transform techniques for the analysis of heat capacity and thermal conductivity measurements, for the case of thin film samples on substrates of finite lengths. The method is a further development of the heat pulse technique. This mathematical analysis is capable of separating the heat capacity and thermal conductivity of the sample from those of the substrate, thus eliminating the need for an additional measurement on the substrate alone. This fact substantially reduces the errors and complexity of the experiment and also makes the heat pulse technique the only one capable of obtaining thermal parameters on thin films in a single experiment. The analysis of the experimental data is performed by calculating several moments of the temperature rise in two thermometers as a function of time. Special considerations are taken to adapt the method for on-line computer experiments.

Thin films have very interesting physical properties. However, because of their thickness, only very seldom are the films self-supporting. The use of a substrate, therefore, is the usual solution. This fact does not influence the electrical measurements made on the film, but is a very big drawback in thermal transfer experiments. Even for relatively thick films, the heat capacity and thermal conductivity of the substrate are not negligible compared to those of the film. All the present methods for heat capacity measurements for thin films, including the a.c. method [1], thermal relaxation methods [2], or pulse methods [3–4], lack the possibility of separation of the substrate thermal parameters from those of the film in one experiment. The only way to get absolute results concerning the film alone is to perform an additional experiment on the substrate alone [5] and to substract the parameters of the substrate from those of the combined assembly of film and substrate.

In this work we present a method based on the heat pulse technique [3–8] by which it would be possible to measure and separate the film and the substrate thermal parameters in one experiment. In the regular heat pulse technique, a $\delta(t)$-shaped heat pulse is introduced at one end of a one-dimensional sample. The other end of the sample is attached to a constant-temperature bath. At one point along the sample the time-dependence of the temperature is measured. From the shape of the pulse at that point and the amount of heat introduced to the sample by the δ pulse, the heat capacity and thermal conductivity can be deduced with the help of the mathematical method described in references [7–8].

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In this work the heat transfer equation is solved for a one-dimensional problem with specified boundary conditions. The solution is based on Laplace transforms, and the desired parameters are found by calculating the moments of the temperature rise at two points along the sample. The method is based on the general ideas shown in reference [6], and is therefore one particular case. All the Laplace transforms and their derivatives used in this work do exist mathematically, as proven in reference [6].

The sample covers only one part (usually half) of the substrate, thus dividing the substrate into two non-equivalent regions. One thermometer is placed in each region. By calculating the moments of the temperature rise in each thermometer, we get relations involving the heat capacities and thermal conductivities of the sample and the substrate. Using these relations, the parameters for the sample and substrate alone can be deduced. In Section 1 of this work we present the mathematical problem, its solution, and the calculation of the moments. The experiments performed using this method are especially suitable for on-line computer analysis. In Section 2 we present special considerations necessary for an on-line experiment, especially those needed to overcome problems involving noise and finite measuring time. In order to check the method, we have used the analogy between one-dimensional heat transfer and electrical transmission line equations. We have built an analog electrical circuit, on which we measured successfully the parameters of the circuit by the use of an on-line computer experiment and the mathematical method outlined here. In Section 3 we describe that analog circuit and the experimental results.

Theory

Description of the mathematical problem

A thin elongated sample with the attached heater and two thermometers is shown in Fig. 1. The heater and the thermometers are lines of length W parallel to the width of the sample. Assuming a negligible thickness-to-length ratio for the sample and substrate (usually of the order of 5.10^{-3} or lower), the resulting heat flow geometry is one-dimensional. The direction of this dimension will be called the x-axis. The one-dimensional heat conduction equation for the sample is

$$K' \frac{\partial^2 T(x, t)}{\partial x^2} = C'\rho \frac{\partial T(x, t)}{\partial t} \tag{1}$$

where K' is the bulk thermal conductivity, C' is the bulk specific heat, ρ is the density and $T(x, t)$ is the temperature rise at point x and time t. It is convenient to use one-dimensional quantities $C = C'\rho Wh$ and $K = K'Wh$ for the specific heat C and thermal conductivity K, respectively. We introduce $R = 1/K$, the thermal resistivity. Actually, C and R are the heat capacity and thermal resistivity of unit length of the sample. By using these notations we get the heat transfer equation as

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

In our case, we attach the heater at $x = 0$. The other end of the sample is thermally "grounded" to a heat sink, at $x = 2l$, i.e. it has a constant temperature. The heat capacity and thermal resistivity of a unit length are C_1 and R_1 for $x \leq l'$, and C_2 and R_2 for $x \geq l'$, respectively, where l' divides the sample into two regions. In the actual experimental set-up, one of the regions represents the substrate alone, and the other region represents the sample and substrate. The quantities C_1, C_2, R_1 and R_2 are continuously distributed along the sample in the appropriate regions.

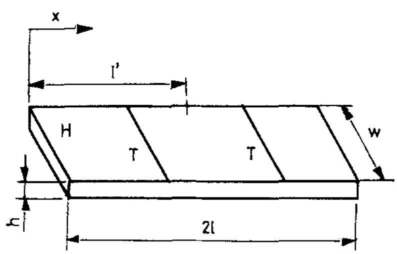

Fig. 1. The one-dimensional geometry (H = heater, T = thermometer)

At time $t = 0$ a short-duration heat pulse of magnitude Q calories is supplied to the heater. The pulse will be treated mathematically as a δ function in time.

The boundary conditions for the problem are

$T(2l, t) = 0$ at all t;
$T(x, 0) = 0$ for $x \neq 0$;
$\dot{Q}(O, t) = Q\delta(t)$, where $\dot{Q}(0, t)$ is the heat flow at $x = 0$ at time t.

The two thermometers are placed at x_1 and x_2, where $x_1 \leq l'$ and $x_2 > l'$.

The mathematical problem of one-dimensional heat flow is identical to the problem of an electrical transmission line with negligible inductance and leakage [9, 10] described in Fig. 2. In this circuit the capacitances and resistances are

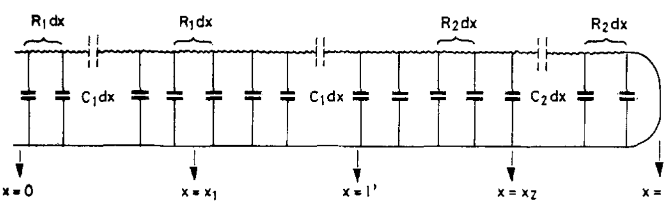

Fig. 2. Electrical analog model for the heat flow problem: two transmission lines connected in series

C_1 and C_2, and R_1 and R_2, respectively. The changes from C_1 to C_2 and from R_1 to R_2 occur at $x = l'$. In the electrical analog circuit, the voltage V and the current I replace the temperature T and the heat current $\dot{Q}$, respectively.

Laplace transform solution for $T(x, t)$

The Laplace transforms [9, 10] for Eq. (2) with the complex variable s are

$$\begin{aligned} T(x, s) &= T(0, s) \cos h(x \cdot h(s)) - \dot{Q}(0, s) \frac{R}{h(s)} \sin h(x \cdot h(s)) \\ \dot{Q}(x, s) &= \dot{Q}(0, s) \cos h(x \cdot h(s)) - T(0, s) \frac{C \cdot s}{h(s)} \sin h(x \cdot h(s)) \end{aligned} \tag{3}$$

where $h^2(s) = C \cdot R \cdot s$

In our case there are two regions. For $x \leq l'$ (region I) we get $h_1^2(s) = C_1 \cdot R_1 \cdot s$, and for $x \geq l'$ (region II) we get $h_2^2(s) = C_2 \cdot R_2 \cdot s$. In region II the point $x = 0$ (needed for Eq. (3)) is given by the point $x = l'$, and x will be replaced by $x - l'$.

We thus obtain four equations, two for $x \geq l'$ and two for $x \leq l'$. By using the boundary conditions specified in the preceding Section we get

$$\begin{aligned} &T(2l, s, = 0 \qquad \text{for all } s \\ &\dot{Q}(0, s) = Q \end{aligned}$$

Using the continuity of $T(x, s)$ and $\dot{Q}(x, s)$ at l', together with the boundary conditions, we eliminate $T(l', s)$ and $\dot{Q}(l_, s)$ and get

for $x \geq l'$

$$T(x, s) = \frac{Q}{s^{\frac{1}{2}}} \frac{\left(\frac{R_2}{C_2}\right)^{\frac{1}{2}} \sin h((2l - x)(C_2 \cdot R_2 \cdot s)^{\frac{1}{2}})}{\cos h(l'(C_1R_1s)^{\frac{1}{2}}) \cdot \cos h((2l - l')(C_2R_2s)^{\frac{1}{2}}) + \left(\frac{C_1R_2}{C_2R_1}\right)^{\frac{1}{2}} \sin h(l'(C_1R_1s)^{\frac{1}{2}}) \cdot \sin h((2l - l')(C_2R_2s)^{\frac{1}{2}})} \tag{4}$$

and for $x \leq l'$

$$T(x, s) = \frac{Q}{s^{\frac{1}{2}}} \frac{\left(\frac{R_1}{C_1}\right)^{\frac{1}{2}} \cos h((2l - l')(C_2R_2s)^{\frac{1}{2}}) \sin h((l' - x)(C_1R_1s)^{\frac{1}{2}}) + \left(\frac{R_2}{C_2}\right)^{\frac{1}{2}} \sin h((2l - l')(C_2R_2s)^{\frac{1}{2}}) \cdot \cos h((l' - x)(C_1R_1s)^{\frac{1}{2}})}{\cos h(l'(C_1R_1s)^{\frac{1}{2}}) \cdot \cos h((2l-l')(C_2R_2s)^{\frac{1}{2}}) + \left(\frac{C_1R_2}{C_2R_1}\right)^{\frac{1}{2}} \cdot \sin h(l'(C_1R_1s)^{\frac{1}{2}}) \cdot \sin h((2l - l')(C_2R_2s)^{\frac{1}{2}})} \tag{5}$$

The moments method

The moments $f_n(x)$ are defined for any integer n as $f_n(x) = \int\limits_0^\infty T(x, t)t^n dt$. Applying the general rules of the Laplace transform at $s = 0$, we get for positive n [10]

$$f_n(x) = (-1)^n \left(\frac{\partial^n T(x,s)}{\partial s^n}\right)_{s=0}$$

and for negative n

$$f_n(x) = \int\limits_0^\infty ds\Big(\int\limits_s^\infty d\sigma\Big)^{|n|-1} T(x, \sigma)$$

As shown in Ref. [6], all these derivatives and integrals exist for the case of thermal conduction.

We will calculate the moments for $n = -1$, 0, 1 and 2. These four moments will give simple relations, enabling one to calculate C_1, C_2, R_1 and R_2. Since $T(x, t)$ is an experimental value, we can compute the moments $f_n(x)$ directly and thus obtain the desired physical parameters.

Substituting the dimensionless quantities

$$\gamma = \frac{l'}{l} \qquad \beta = \frac{2l - l'}{l}$$

we get for $x \geq l'$ (region II) with $\alpha = \dfrac{2l - x}{l}$

$$f_0(x) = Q \cdot \alpha \cdot l \cdot R_2$$

$$f_1(x) = \frac{Q \cdot \alpha \cdot l^3 \cdot R_2}{6} [3\gamma^2 C_1 R_1 + (3\beta^2 - \alpha^2)C_2 R_2 + 6\beta\gamma C_1 R_2] \tag{6}$$

$$f_2(x) = \frac{Q \cdot \alpha . l^5 \cdot R_2}{60} [25\gamma^4 C_1^2 R_1^2 + (25\beta^4 - 10\alpha^2\beta^2 + \alpha^4)C_2^2 R_2^2 + $$
$$+ 120\beta^2\gamma^2 C_1^2 R_2^2 + (100\beta^3 \cdot \gamma - 20\alpha^2\beta\gamma)C_1 C_2 R_2^2 + 100\beta\gamma^3 C_1^2 R_1 R_2 +$$
$$+ (30\beta^2\gamma^2 - 10\alpha^2\gamma^2)C_1 C_2 R_1 R_2]$$

and for $x \leq l'$ (region I) with $\alpha = \dfrac{l' - x}{l}$

$$f_0(x) = Q \cdot l(\alpha R_1 + \beta R_2) \tag{7}$$

$$f_1(x) = \frac{Q \cdot l^3}{6} [(3\alpha\gamma^2 - \alpha^3)C_1 R_1^2 + 2\beta^3 C_2 R_2^2 + 6\gamma\beta^2 C_1 R_2^2 + (3\beta\gamma^2 - 3\alpha^2\beta + 6\alpha\beta\gamma)C_1 R_1 R_2]$$

$$f_2(x) = \frac{Q \cdot l^5}{60} [(\alpha^5 + 25\alpha\gamma^4 - 10\alpha^3\gamma^2)C_1^2 R_1^3 + (5\alpha^4\beta + 100\alpha\beta\gamma^3 + 25\beta\gamma^4 - 30\alpha^2\beta\gamma^2 -$$
$$- 20\alpha^3\beta\gamma) \cdot C_1^2 R_1^2 R_2 + 120\beta^2\gamma^2 C_1^2 R_2^3 + (120\alpha\beta^2\gamma^2 + 100\beta^2\gamma^3 - 60\alpha^2\beta^2\gamma)C_1^2 R_1 R_2^2 +$$
$$+ 16\beta^5 C_2^2 R_2^3 + (20\beta^3\gamma^2 - 20\alpha^2\beta^3 + 40\alpha\beta^3\gamma)C_1 C_2 R_1 R_2^2 + 80\beta^4\gamma C_1 C_2 R_2^3]$$

The computation of $f_{-1}(x)$ can be done numerically, using the relation $f_{-1}(x) = \int_0^\infty T(x, s)\,ds$, with the quantities R_1, R_2, C_1, C_2, l', x and l as parameters of the integral. Evaluation of $f_{-1}(x)$ was done with a computer program. A full discussion and the relevant results are presented in Appendix *A*.

Working point

The purpose of the mathematical solution presented in the previous sections is to enable us to find out (with the help of a heat pulse and two thermometers, at points x_1 and x_2) the heat capacity and thermal resistivity of sample and substrate, namely R_1, R_2, C_1 and C_2. To accomplish this we need at least four independent equations. Therefore, we measure experimentally $T(x, t)$ at $x = x_1$ and $x = x_2$ and calculate up to four of the moments presented in the previous Section and Appendix *A*. We thus have eight independent equations, more than needed to find R_1, R_2, C_1 and C_2. Due to the complicated form of $f_2(x)$ and the limitation on the working point with $f_{-1}(x)$, we choose to use only $f_0(x)$ and $f_1(x)$. It is obvious that we can calculate the other moments, too, or at least use their values to check the validity of the results obtained using only $f_0(x)$ and $f_1(x)$.

By inspection of the expressions in the previous Section, we see that one of the thermometers must be at $x_1 < l'$ (note that x_1 cannot be equal to l'), and the other at $x_2 \geq l'$.

Most of the experimental errors are contained in the measured value of $T(x, t)$ and are propagated to $f_i(x)$. In order to decrease the influence of these errors, we must choose x_1 and x_2 so that the coefficients of C_1, C_2, R_1 and R_2 in Eqs (6) and (7) will be of about the same order of magnitude. As a result we choose our working point to be:

$$l' = l \qquad x_1 = 0.5 \cdot l \qquad x_2 = 1.5 \cdot l.$$

Of course, there are many other working points that can serve as well, depending on the special considerations for each experiment. For our choice we get:

$$\begin{aligned}
f_0(x_1) &= Q \cdot l \cdot (0.5R_1 + R_2) && x < l \\
f_0(x_2) &= Q \cdot l \cdot 0.5 \cdot R_2 && x \geq l \\
f_1(x_1) &= \frac{Q \cdot l^3}{6}\left[\frac{11}{8} C_1 R_1^2 + 2C_2R_2^2 + 6C_1R_2^2 + \frac{21}{4} C_1R_1R_2\right] && x < l \\
f_1(x_2) &= \frac{Q \cdot l^3 R_2}{12}\left[3C_1R_1 + \frac{11}{4} C_2R_2 + 6C_1R_2\right] && x \geq l.
\end{aligned} \tag{8}$$

Mathematical considerations for experimental use of the method

The experimental technique involves the measurement of $T(x_1, t)$ and $T(x_2, t)$ simultaneously. The best way to do this is with the help of an on-line computer with two analog-to-digital inputs. In order to calculate the moments, we must integrate $T(x, t)$ numerically from $t = 0$ to $t = \infty$. This large integration range creates two problems. The first is encountered while computing moments of order $n < 0$. The main contribution to these moments arises for small t's. However, at these points the measured $T(x, t)$ has very small values, and the experimental error due to electronic noise in this time region can be enormous. The best way to solve this problem is to smooth out the digital values for $T(x, t)$ at small t's, using an analytical form of $T(x, t)$ for $t \to 0$. This problem is treated in the following Section.

The second problem arises from the fact that the measurement and the computer memory are naturally limited to finite t, while the integration is done to $t = \infty$. We can try to measure $T(x, t)$ up to very large value of t so that we can neglect the remainder. Naturally, in this way the calculation of high-order moments demands higher and higher t's, and the experimental difficulties are very big. Therefore, we prefer to find an analytical way to extrapolate $T(x, t)$ for $t \to \infty$. This problem is treated in the next to following section.

Expansion for $t \to 0$

We can see that $T(x, s)$ is a function of $s^{\frac{1}{2}}$ and involves $\cos(hs^{\frac{1}{2}})$ and $\sin(hs^{\frac{1}{2}})$ divided by $s^{\frac{1}{2}}$. Therefore, we choose to expand $T(x, s)$ in terms of expressions of the form $\frac{e^{-\alpha s^{\frac{1}{2}}}}{s^{\frac{1}{2}}}$. We then use the mathematical relation that the inverse transform Laplace of $\frac{e^{-\alpha s^{\frac{1}{2}}}}{s^{\frac{1}{2}}}$ is $\frac{e^{-\frac{\alpha^2}{4t}}}{(\pi t^{\frac{1}{2}})}$ to get expansions for $T(x, t)$ for $t \to 0$.

We use the same definitions of α, β and γ given previously, and define another dimensionless parameter $A = \left(\frac{C_1 R_2}{C_2 R_1}\right)^{\frac{1}{2}}$ and get for $x \leq l'$

$$T(x, t) = Q\left(\frac{R_1}{C_1}\right)^{\frac{1}{2}} \frac{1}{(\pi t)^{\frac{1}{2}}} \left[e^{-\frac{l^2(\gamma-\alpha)^2 C_1 R_1}{4t}} - \frac{1-A}{1+A} e^{-\frac{l^2(\alpha+\gamma)^2 C_1 R_1}{4t}} + \right.$$

$$+ \frac{1-A}{1+A} e^{-\frac{l^2(3\gamma-\alpha)^2 C_1 R_1}{4\cdot t}} - \frac{4A}{(1+A)^2} e^{-\frac{l^2}{4t}(2\beta(C_2R_2)^{\frac{1}{2}} + (\gamma+\alpha)(C_1R_1)^{\frac{1}{2}})^2} - \frac{4A}{(1+A)^2} \cdot$$

$$\left. \cdot\, e^{-\frac{l^2}{4t}((3\gamma-\alpha)(C_1R_1)^{\frac{1}{2}} + 2\beta(C_2R_2)^{\frac{1}{2}})^2} + \text{much smaller terms} \right]$$

while for $x \geq l'$

$$T(x, t) = \frac{Q}{1+A}\left(\frac{R_2}{C_2}\right)^{\frac{1}{2}} \frac{2}{(\pi \cdot t)^{\frac{1}{2}}} \left[e^{-\frac{l^2}{4t}(\gamma(C_1R_1)^{\frac{1}{2}} + (\beta-\alpha)(C_2R_2)^{\frac{1}{2}})^2} - \frac{1-A}{1+A} \cdot \right.$$

$$\cdot\, e^{-\frac{2}{4t}(3\gamma(C_1R_1)^{\frac{1}{2}} + (\beta-\alpha)(C_2R_2)^{\frac{1}{2}})^2} - \frac{1-A}{1+A} e^{-\frac{l^2}{4t}(\gamma(C_1R_1)^{\frac{1}{2}} + (3\beta-\alpha)(C_2R_2)^{\frac{1}{2}})^2} + \qquad (9)$$

$$\left. + e^{-\frac{l^2}{4t}(\gamma(C_1R_1)^{\frac{1}{2}} + (\beta+\alpha)(C_2R_2)^{\frac{1}{2}})^2} + \text{much smaller terms} \right]$$

Using the parameters chosen for the working point previously, we get $\alpha = 0.5$, $\beta = 1$ and $\gamma = 1$, and the explicit expressions for the expansions are:

$$T(0.5l, t) = Q\cdot\left(\frac{R_1}{C_1}\right)^{\frac{1}{2}}\frac{1}{(\pi\cdot t)^{\frac{1}{2}}}\left[e^{-\frac{l^2}{4t}0.25C_1R_1} - \frac{1-A}{1+A}e^{-\frac{l^2}{4t}2.25C_1R_1} - \frac{1-A}{1+A}\cdot\right.$$
$$\cdot e^{-\frac{l^2}{4t}6.25C_1R_1} - \frac{4A}{(1+A)^2}e^{-\frac{l^2}{4t}(2(C_2R_2)^{\frac{1}{2}}+1.5(C_1R_1)^{\frac{1}{2}})^2} -$$
$$\left. - \frac{4A}{(1+A)^2}e^{-\frac{l^2}{4t}(2.5(C_1R_1)^{\frac{1}{2}}+2(C_2R_2)^{\frac{1}{2}})^2} + \dots\right]$$
$$T(1.5l, t) = \frac{Q}{1+A}\left(\frac{R_2}{C_2}\right)^{\frac{1}{2}}\frac{2}{(\pi t)^{\frac{1}{2}}}\left[e^{-\frac{l^2}{4t}((C_1R_1)^{\frac{1}{2}}+0.5(C_2R_2)^{\frac{1}{2}})^2} - \right. \tag{10}$$
$$- \frac{1-A}{1+A}e^{-\frac{l^2}{4t}(3(C_1R_1)^{\frac{1}{2}}+0.5(c_2R_2)^{\frac{1}{2}})^2} - \frac{1-A}{1+A}e^{-\frac{l^2}{4t}(C_1R_1)^{\frac{1}{2}}+2.5(C_2R^{c})^{\frac{1}{2}})^2} -$$
$$\left. - e^{-\frac{l^2}{4t}((C_1R_1)^{\frac{1}{2}}+1.5(C_2R_2)^{\frac{1}{2}}} + \dots\right]$$

This expansion is reduced to the first term for very small t. However, we have to specify what is meant by "small t" during an on-line computer experiment.

The only way to evaluate t during the experiment is to compare it with $t_{max}(x)$, which is the time at which the temperature $T(x, t)$ has its maximum value $T_{max}(x)$. t_{max} is a directly measured value. We will now give estimates for $t_{max}(x)$.

Using one-term expansion in Eq. (10) as an approximate form of $T(x, t)$, we get

$$T(0.5l, t) = Q\left(\frac{R_1}{C_1}\right)^{\frac{1}{2}}\frac{1}{(\pi t)^{\frac{1}{2}}}e^{-\frac{l^2}{4t}0.25C_1R_1}$$
$$T(1.5l, t) = \frac{Q}{1+A}\left(\frac{R_2}{C_2}\right)^{\frac{1}{2}}e^{-\frac{l^2}{4t}((C_1R_1)^{\frac{1}{2}}+0.5(C_2R_2)^{\frac{1}{2}})^2} \tag{11}$$

From Eq. (11) we can calculate $t_{max}(x)$ simply by differentiation, and get:

$$t_{max}(0.5l) = \frac{l^2}{8}C_1R_1$$
$$t_{max}(1.5l) = \frac{l^2}{2}((C_1R_1)^{\frac{1}{2}} + 0.5(C_2R_2)^{\frac{1}{2}})^2 \tag{12}$$

It is impossible to find t_{max} analytically for any longer (and more meaningful) expansion of the form used in Eq. (10).

It can be shown by numerical calculation that for $x = 0.5\cdot l$ we have

$$0.9\frac{l^3}{8}\cdot C_1\cdot R_1 < t_{max} \leq \frac{l^2}{8}\cdot C_1\cdot R_1.$$

The situation for $x = 1.5l$ is more complicated, and by calculation of the same type we get a much wider range for t_{max}, namely

$$0.64 \frac{l^2}{2} ((C_1R_1)^{\frac{1}{2}} + 0.5(C_2R_2)^{\frac{1}{2}})^2 \leq t_{max}(1.5l) \leq \frac{l^2}{2} ((C_1R_1)^{\frac{1}{2}} + 0.5(C_2R_2)^{\frac{1}{2}})^2$$

Coming back to the expansion for $t \to 0$ (Eq. 10), we can see that the error obtained in $T(0.5l, t)$ by neglecting the second term will be less than 2%, even up to $t = t_{max}$. Therefore, we can use a one-term expansion in order to smooth out the data near $t = 0$ by a simple least squares fit up to a certain fraction of t_{max}.

The situation for $x = 1.5 \cdot l$ is more complex. It can be seen that for the case $C_2R_2 \ll C_1R_1$ it is difficult to use a one-term expansion, even for very small t. However, for the more realistic case where $C_1R_1 \simeq C_2R_2$, we get that up to $t = 0.2t_{max}$ the contribution of the second term will be less than 1%. Using Eq. (12), $t_{max}(1.5l) > 4 \cdot t_{max}(0.5 \cdot l)$. Thus, the absolute value of t at $x = 1.5l$ is large enough for accurate numerical calculations. If one wants to calculate negative moments without a prior knowledge of the ratio C_2R_2/C_1R_1, and to reduce errors by smoothing out the data near $t = 0$, it is advisable to use the one-term expansion given in Eq. (11), with the range of t's proposed in this Section. After completing the calculation and getting C_1, C_2, R_1 and R_2, the validity of the selected range must be checked for the case of $C_2R_2 \ll C_1R_1$, namely to recalculate all the thermal parameters once again, but with a reduced t range for the fit for $t \to 0$.

In passing, we can note that the validity of any fit to experimental data can always be checked by statistical methods, as part of the calculation of the parameters of the proposed fit.

We note that for $x = 0.5l$, if $t_{max} = \frac{1}{8} l^2C_1R_1$ (a rather good approximation for any t_{max}), we have

$$T(t_{max}) = 0.968 \cdot \frac{Q}{C}$$

Expansions for $t \to \infty$

We were unable to find an analytic expression for an expansion for $T(x, t)$ for $t \to \infty$. However, we will show that there is a range of time values (in terms of t_{max}) that give a one-term expansion for $T(x, t)$. Suppose the function $T(x, s)$ has α_v ($v = 1, 2, 3 \ldots$) poles of first order; then $T(x, t)$ can be expanded

$$T(x, t) = \sum_{v} \text{Rez}\,(\alpha_v)e^{\alpha_v \cdot t} \tag{13}$$

Therefore, we look for the poles of Eqs (4) and (5). We divide Eqs (4) and (5) into two parts; the first is the nominator divided by $s^{\frac{1}{2}}$, and the second is the denominator. It can be seen that for the two equations the nominator divided by $s^{\frac{1}{2}}$ has no poles. Therefore, all the poles in the equations are the zero's of the denominators, namely the zero's of

$$f(s^{\frac{1}{2}}) = \cos h(\gamma \cdot l(C_1R_1s)^{\frac{1}{2}}) \cdot \cos h(\beta \cdot l(C_2R_2s)^{\frac{1}{2}}) + \\ + A \sin h(\gamma \cdot l(C_1R_1s)^{\frac{1}{2}}) \sin h(\beta \cdot l(C_2R_2s)^{\frac{1}{2}})$$

where β, α and A were defined in the previous Sections.

By substituting

$$\begin{aligned} r &= \gamma \cdot l(C_1R_1)^{\frac{1}{2}} + \beta \cdot l(C_2R_2)^{\frac{1}{2}} \\ p &= \gamma \cdot l(C_1R_1)^{\frac{1}{2}} - \beta l(C_2R_2)^{\frac{1}{2}} \end{aligned} \tag{14}$$

we get

$$f(s^{\frac{1}{2}}) = \frac{1}{2}(1 + A) \cos h(r \cdot s^{\frac{1}{2}}) + \frac{1}{2}(1 - A) \cosh (ps^{\frac{1}{2}})$$

Since s is real, then $s^{\frac{1}{2}}$ is either real or purely imaginary. By inspection of Eq. (14) and taking into account that $A > 0$, $r > 0$, and $|r| > |p|$, we can see that $s^{\frac{1}{2}}$ cannot be both real and a solution for $f(s^{\frac{1}{2}}) = 0$. Thus, we can define $s^{\frac{1}{2}} = iu$ and solve the equation

$$f(u) = (1 + A) \cos (r \cdot u) + (1 - A) \cdot \cos (p \cdot u) \tag{15}$$

It can also be seen that all the roots of $f(u)$ in Eq. (15) are simple roots, and using Eq. (13) one gets

$$T(x, t) = \sum_{\nu} \mathrm{Rez}\,(-u_{\nu}^2)e^{-u_{\nu}^2 \cdot t} \tag{16}$$

where u_{ν} are the roots of $f(u)$. The residue in Eq. (16) is calculated from Eqs (4) and (5).

From the expansion (16) we see that for large enough t's we can use a one-term expansion. As previously suggested for small t, one can use a one-term approximation as a fit for large t, using a term of the form De^{-Et}. The validity of such a fit can be checked by statistical methods during the experiment.

In order to get an estimate of the permitted range for this one-term approximation, we have used a computer program to calculate the first two solutions of Eq. (15) for a wide range of physical parameters C_1, C_2, R_1 and R_2. For each set of solutions one calculates the ratio between the first two terms in the expansion of Eq. (16). After completing the calculation for a wide range of C_1, C_2, R_1 and R_2, we limited ourselves to the range of physical interest. The main requirement is that C_1R_1 and C_2R_2 will differ by not more than one order of magnitude. This requirement can easily be fulfilled by the right choice of the substrate. Moreover, this requirement is vital for the regular mathematical analysis of the heat pulse method [8], namely a one-dimensional heat flow.

Restricting the ratio C_1R_1/C_2R_2 as mentioned gives a one-term expansion (Eq. (16)) in region II at $x = 1.5l$ for $t > 2 \cdot t_{max}(1.5 \cdot l)$, and in region I at $x = 0.5 \cdot l$ for $t > 10 \cdot t_{max}(0.5 \cdot l)$. Since $t_{max}(1.5 \cdot l) > 4 \cdot t_{max}(0.5 \cdot l)$, the absolute values of t involved in the two locations are about the same.

Finally, we wish to note that it is advisable to check the conditions for one-term expansion after computing C_1, C_2, R_1 and R_2, in the same way as mentioned in the former Section.

Electrical analog experiment

An experimental check of the mathematical theory can be done by making an experiment on an electrical set-up like that of Fig. 2 (except that in our case the transmission lines were replaced by discrete elements). The reasons for choosing this procedure to verify the theory, instead of a heat transfer experiment, are accuracy and simplicity in the experimental set-up and the actual measurements.

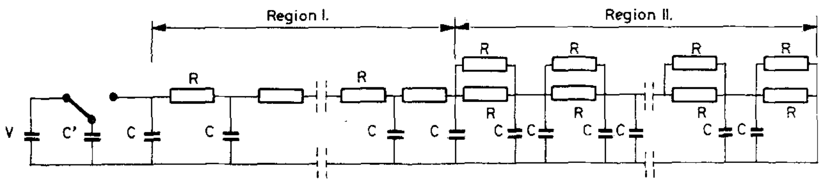

Fig. 3. Electrical analog experimental set-up

The discrete transmission line was composed of resistors and capacitors, and is shown in Fig. 3. The δ function heat pulse Q was obtained by charging a capacitor C' to a known voltage and subsequently discharging it through the transmission line. The voltage V was measured at the desired points, namely $x = 0.5l$ and $x = 1.5l$, by two analog-to-digital inputs of a P.D.P.-8 on-line computer at varying rates between 1000 and 3000 points per second. At every input 100 points were measured as background ($t < 0$) and 500 points were recorded from $t = 0$ to t greater than t_{max}. The recorded points were presented by the computer on a storage scope to enable a first check of the pulse. Least squares fits for the small and large t regions were done for every pulse. The fits were done by a one-term approximation at the range recommended in Sections 2.1 and 2.2. The fits were checked by statistical methods and by presenting the fitted line on the scope and comparing to the measured data. The moments f_0 and f_1 were calculated from the experimental $T(x, t)$ points and the fits. Using Eq. (8), the parameters C_1, C_2, R_1 and R_2 were derived. A complete heat pulse measurement and data analysis lasts about 3 minutes. We performed three sets of experiments. First, we used a homogeneous discrete "transmission line" of 28 elements, each made of one capacitor and one resistor. We measured the resistance and capacity by the standard heat pulse method [7]. Secondly, we repeated the experiment using the same discrete "transmission line", but the measurements were made at points equivalent to $x = 0.5l$ and $x = 1.5l$. Thirdly, we carried out the experiment using a heterogeneous discrete "transmission line" of 14 regular elements in series with 14 elements, each made of two resistors and two capacitors in parallel, as presented in Fig. 3.

Table 1

Results of the electrical analog experiments

Experiment No.	"Thermometer" position	f_0	f_1	$R, k\Omega$	$C, \mu F$
1	$x = l$	0.5809	0.2047	9.430	0.104
2	$x = 0.5\,l$	0.8690	0.2681	9.405	0.103
2	$x = 1.5\,l$	0.2895	0.1076	9.400	0.103
3	$x = 0.5\,l$	0.5799	0.1194	9.414	0.102
3	$x = 1.5\,l$	0.1454	0.09033	9.386	0.104

In this configuration we get $C_2 = 2C_1$ and $R_2 = 0.5R_1$, and again we measured the time-dependence at two points, equivalent to $x = 0.5l$ and $x = 1.5l$.

The results from all the measurements are presented in Table 1. In this Table the various experiments are presented as first, second and third experiment, as described above. The values denoted as R and C in Table 1 are the resistance and capacitance, respectively, of one resistor and one capacitor, as computed from the experimental results after taking into account the capacitor C'. These results are slightly different from the indicated values of $R = 10\ k\Omega \pm 5\%$ and $C = 0.1\ \mu F \pm$ $\pm 5\%$, but within the indicated errors, and smaller than the relative value of one element ($\approx 7\%$) in this discrete system. Since the difference is a feature common to the three measurements, we attribute this difference to the discrete nature of the "transmission line" used. This fact is in contrast to the case of a thermal transport experiment, which has a continuous nature. As the mathematical analysis is based on a continuous medium, no additional error will be added to thermal property experiments.

Discussion

Using a Laplace transform technique, we have solved the heat transfer equation for the case of a one-dimensional sample. A δ-shaped heat pulse is applied at one end, and the sample contains two regions with different heat capacities and thermal conductivities.

Using this solution and the calculation of experimentally attainable moments, we found a way to evaluate the heat capacities and thermal conductivities of both the substrate and the sample in a single experiment. The only assumptions in the model are those which were used in the regular heat pulse method [5]. The main advantage of this analysis is for absolute measurements of thermal parameters of thin film in cases where the parameters of the substrate and sample are of about the same order of magnitude and neither can be neglected.

The method is particularly good for on-line computer experiments similar to those used by us in the electrical analog experiment. The mathematics presented here enables the user to avoid experimental errors due to noise by using fits to the beginning and tail of the measured heat pulses, and to check his results by the computation of other moments. We have recommended a working point where $x_1 = 0.5l$, $x_2 = 1.5l$ and $l' = l$, but of course many other points can be used.

The accuracy of the measurements can be greatly improved by repeating the heat pulse many times before analysing the data, thus decreasing the influence of electronic noise.

A real-life thermal transfer experiment has to be performed, using two small thermometers. The sample has to be prepared by depositing the desired thin film on only half (if $l' = l$) of the substrate length. The thermometers will then be attached to the centers of the two regions and the temperature rises will be measured and fed to the on-line computer simultaneously.

Appendix A: Computation of $f_{-1}(x)$

The computation of $f_{-1}(x)$ cannot be done analytically. Therefore, we will give numerical estimates. From section "Theory. The moments method" we have:

$$f_{-1}(x) = \int_0^\infty T(x, s)ds$$

The physical quantities R_1, R_2, C_1, C_2, l, l′ and x are presented here as parameters. In order to perform the computer numerical evaluation of $f_{-1}(x)$, we have transformed s to the interval [0, 1]. We get for $x \leq l'$

$$f_{-1}(x) = \frac{2Q}{\gamma}\left(\frac{R_1}{C_1}\right)^{\frac{1}{2}} \int_0^1 \frac{dx}{(1-x)^2} \frac{\cos h\left(\frac{\beta}{\gamma}\left(\frac{C_2R_2}{C_1R_1}\right)^{\frac{1}{2}} \frac{x}{1-x}\right) \sin h\left(\frac{\alpha}{\gamma}\frac{x}{1-x}\right)}{\cos h\frac{x}{1-x} \cos h\left(\frac{\beta}{\gamma}\left(\frac{C_2R_2}{C_1R_1}\right)^{\frac{1}{2}} \frac{x}{1-x}\right)} +$$

$$+ \frac{A \sin h\left(\frac{\beta}{\gamma}\left(\frac{C_2R_2}{C_1R_1}\right)^{\frac{1}{2}} \frac{x}{1-x}\right) \cos h\left(\frac{\alpha}{\gamma}\frac{x}{1-x}\right)}{A \sin h\frac{x}{1-x} \sin h\left(\frac{\beta}{\gamma}\left(\frac{C_2R_2}{C_1R_1}\right)^{\frac{1}{2}} \frac{x}{1-x}\right)}$$

and for $x \geq l'$

$$f_{-1}(x) = \frac{2Q}{\alpha l C_2}\int_0^1 \frac{\mathrm{d}x}{(1-x)^2} \frac{\sin h\frac{x}{1-x}}{\cos h\left(\frac{\beta}{\alpha}\frac{x}{1-x}\right)\cos h\left(\frac{\gamma}{\alpha}\left(\frac{C_1R_1}{C_2R_2}\right)^{\frac{1}{2}}\frac{x}{1-x}\right) + A \sin h\left(\frac{\beta}{\alpha}\frac{x}{1-x}\right) \cdot \sin h\left(\frac{\gamma}{\alpha}\left(\frac{C_1R_1}{C_2R_2}\right)^{\frac{1}{2}}\frac{x}{1-x}\right)}$$

where α, β, γ and A were defined earlier.

At our working point $x = 0.5l$, we have $\frac{\alpha}{\gamma} = 0.5$ and $\frac{\beta}{\gamma} = 1$. We also limit ourselves to the range of physical interest, i.e. $C_1R_1 = C_2R_2$. Representative calculations were done for the interval $0.8 \le \frac{C_2R_2}{C_1R_1} \le 1.2$. First we assume $A \ge 1$ (which is equivalent to $C_1 \ge C_2$) and we obtain

$$f_{-1}(0.5l) = 1.1850 + 0.2671 \left(\frac{C_1R_2}{C_2R_1}\right)^{\frac{1}{2}} + 0.1943 \left(\frac{C_2R_2}{C_1R_1}\right)^{\frac{1}{2}} \tag{A1}$$

For $1 \le \frac{C_1R_1}{C_2R_2} \le 1.45$, the maximum error for all the mentioned range is less than 0.1%, while the mean error is 0.03%. For a wider interval we obtain similar expansions, but with a larger error margin.

For $A \le 1$ (or $C_2 \ge C_1$) we get

$$f_{-1}(0.5l) = 1.1810 - 0.2927 \left(\frac{C_1R_2}{C_2R_1}\right)^{\frac{1}{2}} - 0.1722 \left(\frac{C_2R_2}{C_1R_1}\right)^{\frac{1}{2}} \tag{A2}$$

For $0.65 \le \left(\frac{C_1R_2}{C_2R_1}\right) \le 1$, the involved errors are about the same as in Eq. (A1). For $x = 1.5l$ we get $\frac{\beta}{\alpha} = 2$ and $\frac{\gamma}{\alpha} = 2$. If we choose the same range of values for A as in Eqs (A1) and (A2), we get for $C_1 > C_2$

$$f_{-1}(1.5l) = 0.2856 - 0.03626 \left(\frac{C_1R_2}{C_2R_1}\right) - 0.06334 \left(\frac{C_1R_1}{C_2R_2}\right)^{\frac{1}{2}} \tag{A3}$$

and for $C_2 > C_1$

$$f_{-1}(1.5l) = 0.30548 + 0.04355 \left(\frac{C_1R_2}{C_2R_1}\right)^{\frac{1}{2}} + 0.06965 \left(\frac{C_1R_1}{C_2R_2}\right)^{\frac{1}{2}} \tag{A4}$$

The maximum error in (A3) and (A4) is 0.3%, and the mean error is 0.1%.

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RÉSUMÉ — On présente une méthode mathématique reposant sur les techniques de transformation de Laplace, pour l'analyse des mesures de capacité calorifique et de conductivité thermique, dans le cas des échantillons en pellicules minces sur des substrats de longueurs finies. La méthode représente un développement ultérieur de la technique des pulsations de chaleur. Cette analyse mathématique est capable de séparer la capacité calorifique et la conductivité thermique de l'échantillon de celles du substrat, en éliminant ainsi la nécessité d'une mesure suppplémentaire du substrat seul. Ce fait réduit notablement les erreurs et la complexité de l'expérience et rend la technique des pulsations thermiques la seule capable d'obtenir des paramètres thermiques de pellicules minces à partir d'une seule expérience. L'analyse des données expérimentales s'effectue en calculant à divers moments l'augmentation de la température de deux thermomètres, en fonction du temps. Des mesures spéciales ont été prises pour adapter la méthode à des expériences sur ordinateur en ligne.

ZUSAMMENFASSUNG — Eine auf Laplace-scher Transformationstechnik beruhende mathematische Methode zur Analyse von Wärmekapazitäts- und Wärmeleitungsmessungen für Dünnschichtproben auf Substraten begrenzter Längen wird vorgestellt. Die Methode ist eine Weiterentwicklung der Wärmepulstechnik. Diese mathematische Analyse vermag die Wärmekapazität und Wärmeleitfähigkeit der Probe von denen des Substrats zu trennen, wodurch sich die zusätzliche Messung des Substrats allein erübrigt. Hierdurch werden Fehler und Komplexität des Versuchs bedeutend reduziert und die Wärmepuls-Methode erweist sich als erstes Verfahren bei dem die thermischen Parameter dünner Filme in einem einzigen Versuch ermittelt werden können. Die Analyse der Versuchsdaten wird durch die Berechnung verschiedener Momente des Temperaturanstiegs in zwei Thermometern als Funktion der Zeit durchgeführt. Besondere Maßnahmen wurden bei der Anpassung der Methode für on-line Computerversuche berücksichtigt.

Резюме — Представлен математический метод, основанный на преобразования Лапласа, для анализа измерений теплоемкости и термопроводимости тонкопленочных образцов на подложке конечный длины. Этот метод является дальнейшим развитием техники теплового импульса. Математический анализ позволяет разделить теплоемкость и термопроводность образца и подложки, что, следовательно, устраняет необходимость дополнительного измерения самой подложки. Метод значительно понижает ошибки и сложность эксперимента и делает метод теплового импульса единственным методом получения термических параметров тонких пленок с помощью одного измерения. Анализ экспериментальных данных проводили путем вычисления нескольких моментов подъема температуры в двух термометрах как функцию времени. Представлены соображения о применимости к этому ЭВМ, работающей в режиме «на линии».