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CORRECTION METHODS FOR HEAT LEAKS IN ADIABATIC CALORIMETRY

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ABSTRACT

Extrapolation methods for the correction of heat leaks in adiabatic calorimetry are discussed to obtain a good estimate of the ideal temperature increment ΔT for three cases of the origin of heat leaks; radiation, conduction, and both. A good estimate of ΔT in the radiation case is obtained by the extrapolation of the cooling curve to the half point of the heating duration t_0, with an error of the order of t_0^2. A good estimate of ΔT in the conduction case is obtained by the extrapolation of the cooling curve to the half point of the heating duration and by dividing the resultant temperature increment by $(1 - H_1/3H_0)$, where H_0 is the heat capacity of the sample and H_1 is that of the electrical leads and supporting materials. A simple physical meaning of the correction factor is given.

INTRODUCTION

Adiabatic (Nernst) calorimetry is known to give the highest accuracy in measuring the specific heat [1], and has the capacity for the absolute determination of the specific heat. A nearly adiabatic condition is realized at low temperatures, where superconducting leads with good electrical conductivity and good thermal insulation can be used. The ideal adiabatic condition can not, however, be satisfied at higher temperatures where heat leaks are enhanced because of increased thermal conductivity of electrical leads due to the breakdown of superconductivity and because of increased heat leaks due to radiation. The condition becomes unfavorable in high magnetic fields even at low temperatures, because superconductivity breaks down in the electrical leads.

Two extrapolation methods for the correction of heat leaks have been adopted to estimate the temperature increment ΔT in the ideal adiabatic condition. An empirical method is the extrapolation of the cooling curve to the half point of the heating duration t_0 (see Fig. 2 below) [1]. A theoretical method is the extrapolation of the cooling curve to the starting time of heating, on the basis of the simple model of heat leakage proportional to the

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temperature difference between the sample and the external heat bath [2,3]. Another method is the "planimeter" method [4], that is the method using the area of the heating and cooling curve on the basis of the same heat leak model. The method is, however, somewhat cumbersome, and the extrapolation method seems sufficient when the heat leak is small. The planimeter method seems to be applied in a heat pulse method [5] beyond the condition originally imposed in ref. 4 where quasistationary heating is assumed.

The empirical and theoretical extrapolation methods seem to contradict one another. The simple model for heat leakage in the latter method is not justified in general, because heat leaks through electrical leads (and supporting materials) can not be neglected compared with those through radiation at relatively low temperatures. The partial differential equation for heat conduction has to be solved in the discussion of the extrapolation.

Extrapolation methods for the correction of heat leaks are discussed below in a simple one-dimensional model for the three cases of the origin of the heat leaks; radiation, conduction, and both. A good estimate of ΔT in the radiation case is shown to be obtained by the extrapolation of the cooling curve to the half point of heating duration, with an error of the order of t_0^2. A good estimate of ΔT in the conduction case is obtained by the extrapolation of the cooling curve after the intrinsic relaxation time of electrical leads to the half point of heating duration and by dividing the resultant temperature increment by a factor $(1 - H_1/3H_0)$ where H_0 is the approximate heat capacity of the sample without correction, and H_1 is that of the electrical leads and supporting materials. It is recommended that in the intermediate situation (with heat leaks through both radiation and conduction), the heat capacity of the electrical leads (and supporting materials) should be, if possible, negligible compared with that of the sample, and that the cooling curve should be extrapolated to the half point of t_0.

HEAT LEAK BY RADIATION

The sample of temperature T_1 with surface area S_r is assumed to exchange heat with an external heat bath of temperature T_0 by radiation as shown in Fig. 1. Heat flow is given by the following equation

$$\sigma S_r(T_1^4 - T_0^4) \simeq 4\sigma S_r T_0^3(T_1 - T_0) \equiv AT \quad (1)$$

where σ is the Stefan–Boltzmann constant and T is the temperature difference between the sample and the heat bath. The possible deviation from unity of emissivity of the sample and the external heat bath is neglected for simplicity. The deviation of the emissivity from unity can easily be included in the coefficient A. Heat exchange through rarefied gas takes the same functional form as eqn. (1) [6]. The heat capacity of the sample is H_0, and a constant power w is applied to the sample for the time

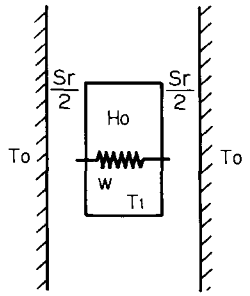

Fig. 1. Model of an adiabatic calorimeter with heat leak through radiation.

duration between 0 and t_0. The temperature difference T satisfies the following differential equation, on the assumption that the relaxation time within the sample is short enough compared with the relaxation time determined by eqn. (5) below to allow an approximation of homogeneity of the temperature in the sample

$$H_0 \frac{dT}{dt} = w[1 - U(t - t_0)] - AT \tag{2}$$

where $U(t)$ is the step function. The equation is expressed as

$$\frac{dT'}{dt'} = \frac{1}{t_0'}[1 - U(t' - t_0')] - T' \tag{3}$$

with the use of dimensionless variables defined as

$$t' = t/\tau_r \qquad t_0' = t_0/\tau_r \qquad T' = H_0 T/(w t_0' \tau_r) \tag{4}$$

where

$$\tau_r = H_0/A \tag{5}$$

is the relaxation time of the sample cooling due to the radiation.

The solution for $t' > t_0'$ satisfying the initial condition T' $(t' = 0) = 0$ is elementarily given by

$$T' = f(t') - f(t' - t_0')U(t' - t_0') \tag{6}$$

$$f(t') = \frac{1}{t_0'}[1 - e^{-t'}] \tag{7}$$

The Taylor expansion of eqn. (6) for $t' \geqslant t_0'$ gives

$$T' = 1 + \left(\frac{t_0'}{2} - t'\right) + \tfrac{1}{6}\left(3t'^2 - 3t_0't' + t_0'^2\right) + \dots \tag{8}$$

The extrapolation to $t_0'/2$, therefore, gives a good estimate of the temperature increment $\Delta T' = 1$ in the ideal adiabatic condition as shown in Fig. 2 with an error of the order of $t_0'^2$.

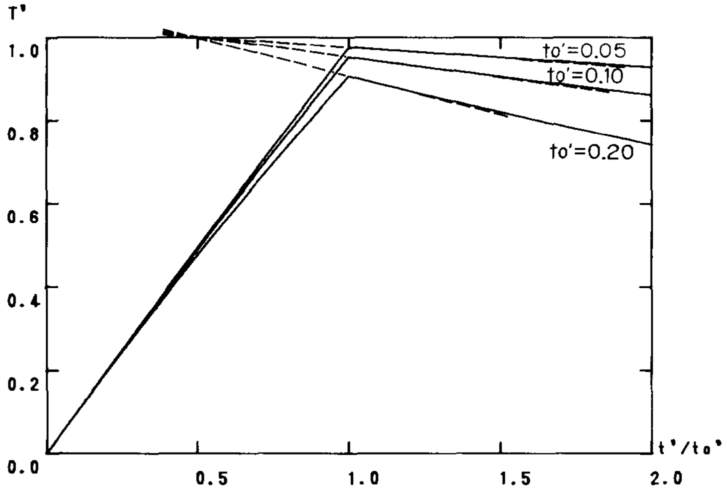

Fig. 2. Extrapolation method for the heat leak through radiation. Extrapolation to the half point of heating duration t'_0 gives a good estimate of ideal temperature increment.

The heat leak term approximated by eqn. (1) is essentially the same as that of refs. 2 and 3, but the present conclusion contradicts them. The origin of the contradiction can be traced back to a one-sided approximation made for the exponential function in the refs. 2 and 3 in which the first order Taylor expansion is made for $e^{t'_0}$ in spite of no approximation being made for $e^{-t'}$.

HEAT LEAK BY CONDUCTION

Heat leak by conduction through electrical leads (and supporting materials) becomes important at relatively low temperatures, where the radiation discussed above is not important because the rate of radiation proportional to T^4 is greatly diminished. Two extrapolation methods are discussed below on the basis of the nonstationary solution of the equation of heat conduction, and the latter one is concluded to be better from a practical point of view.

The model is shown in Fig. 3. H_0 is the heat capacity of the sample, w the heat power generated in the sample. κ is the conductivity, c the specific heat, l the length, and S_c the area of the cross section of the electrical lead, respectively. The partial differential equations of heat conduction expressed in dimensionless variables are

$$\frac{\partial T''(x', t'')}{\partial t''} = \frac{\partial^2 T''(x', t'')}{\partial x'^2} \tag{9}$$

$$\left[\frac{\partial T''(x', t'')}{\partial t''}\right]_{x'=1} = \frac{1}{t''_0}[1 - U(t'' - t''_0)] - k\left[\frac{\partial T''}{\partial x'}\right]_{x'=1} \tag{10}$$

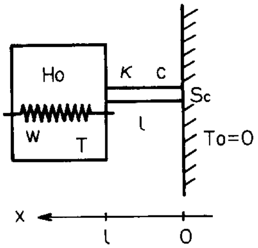

Fig. 3. Model of an adiabatic calorimeter with heat leak through conduction.

and the following initial and boundary conditions are imposed

$$T''(x', 0) = 0 \tag{11}$$

$$T''(0, t'') = 0 \tag{12}$$

The dimensionless variables and constants are defined as

$$t'' = t/\tau_i \tag{13}$$

$$t_0'' = t_0/\tau_i \tag{14}$$

$$x' = x/l \tag{15}$$

$$k = clS_c/H_0 \tag{16}$$

$$T'' = \frac{H_0}{wt_0''\tau_i} T \tag{17}$$

where

$$\tau_i = cl^2/\kappa \tag{18}$$

is the characteristic relaxation time of the electrical lead itself. The temperature of the external heat bath is taken as the origin of the temperature without loss of generality.

The operational calculus of Miksinski [7] (or isomorphic Laplace-transform calculus) gives the following solution for eqns. (9)–(12)

$$\{T''(x', t'')\} = \frac{1}{t_0''} \frac{1 - \exp(-st_0'')}{s} \frac{\sinh(\sqrt{s}\,x')}{s \cdot \sinh(\sqrt{s}) + k\sqrt{s}\,\cosh(\sqrt{s})} \tag{19}$$

The temperature of the sample ($x' = 1$) is, therefore, given by

$$\{T''(1, t'')\} = \frac{1}{t_0''}[\{f(t'')\} - \{f(t'' - t_0'')\}U(t'' - t_0'')] \tag{20}$$

where

$$\{f'(t'')\} = \frac{1}{s} \frac{\sinh(\sqrt{s})}{s \cdot \sinh(\sqrt{s}) + k\sqrt{s}\,\cosh(\sqrt{s})} \tag{21}$$

because the following general relation holds

$$\exp(-\lambda s)\{f(t)\} = \begin{cases} 0 & 0 \leqslant t \leqslant \lambda \\ \{f(t-\lambda)\} & 0 \leqslant \lambda \leqslant t \end{cases} \tag{22}$$

The inverse-transform of eqn. (21) is not found in published tables [8]. An approximate formula of eqn. (21) for small k may be obtained by the following expansion of eqn. (21) to the first order of k

$$\begin{aligned} \{f(t'')\} &\sim \frac{1}{s^2}\left[1 - k\frac{1}{\sqrt{s}}\coth(\sqrt{s})\right] \\ &= \{t''\} - k\left(\frac{\coth(\sqrt{s})}{s}\right)\frac{1}{s^{1.5}} \end{aligned} \tag{23}$$

The function in parentheses in the last line of eqn. (23) is inverse-transformed as

$$\begin{aligned} \frac{\coth(\sqrt{s})}{s} &= \frac{1}{s}\left[1 + 2\exp(-2\sqrt{s})\frac{1}{1-\exp(-2\sqrt{s})}\right] \\ &= \left\{1 + 2\sum_{n=1}^{\infty}\operatorname{cerf}\left(\frac{n}{\sqrt{t''}}\right)\right\} \end{aligned} \tag{24}$$

The second term in the last line is negligible for small t'' compared with the first term because, for example, $\operatorname{cerf}(1/\sqrt{0.5}) = 0.0455$. Equation (24) is, therefore, approximated for small t'' as

$$\frac{\coth(\sqrt{s})}{s} \sim \{1\} = \frac{1}{s} \tag{25}$$

Equation (23) with this approximation yields

$$\{f(t'')\} \sim \{t''\} - k\frac{1}{s}\frac{1}{s^{1.5}} = \{t''\} - k\left\{\frac{4}{3\sqrt{\pi}}t''^{1.5}\right\} \tag{26}$$

Equation (20) for $t'' > t_0''$ is, therefore, approximated for small t'' and t_0'' by

$$T''(1, t'') \sim \frac{1}{t_0''}\left\{t_0'' - k\frac{4}{3\sqrt{\pi}}\left[t''^{1.5} - (t'' - t_0'')^{1.5}\right]\right\} \tag{27}$$

The tangential line of the curve of eqn. (27) at $t'' = t_0''$ is given by

$$T''(1, t'') = 1 - k\frac{4}{3\sqrt{\pi}}\sqrt{t_0''} - 1.5k\frac{4}{3\sqrt{\pi}\sqrt{t_0''}}(t'' - t_0'') \tag{28}$$

Extrapolation of eqn. (28) to $t'' = t_0''/3$ gives $T'' = 1$, which is the temperature increment in the ideal adiabatic condition. Some examples of the present extrapolation method are illustrated in Fig. 4, where the heating and cooling curve is calculated from eqn. (20) with the numerical inverse-transform of eqn. (21) obtained by the FILT method [9], and the gradient of the curve at $t'' = t_0''$ is obtained numerically.

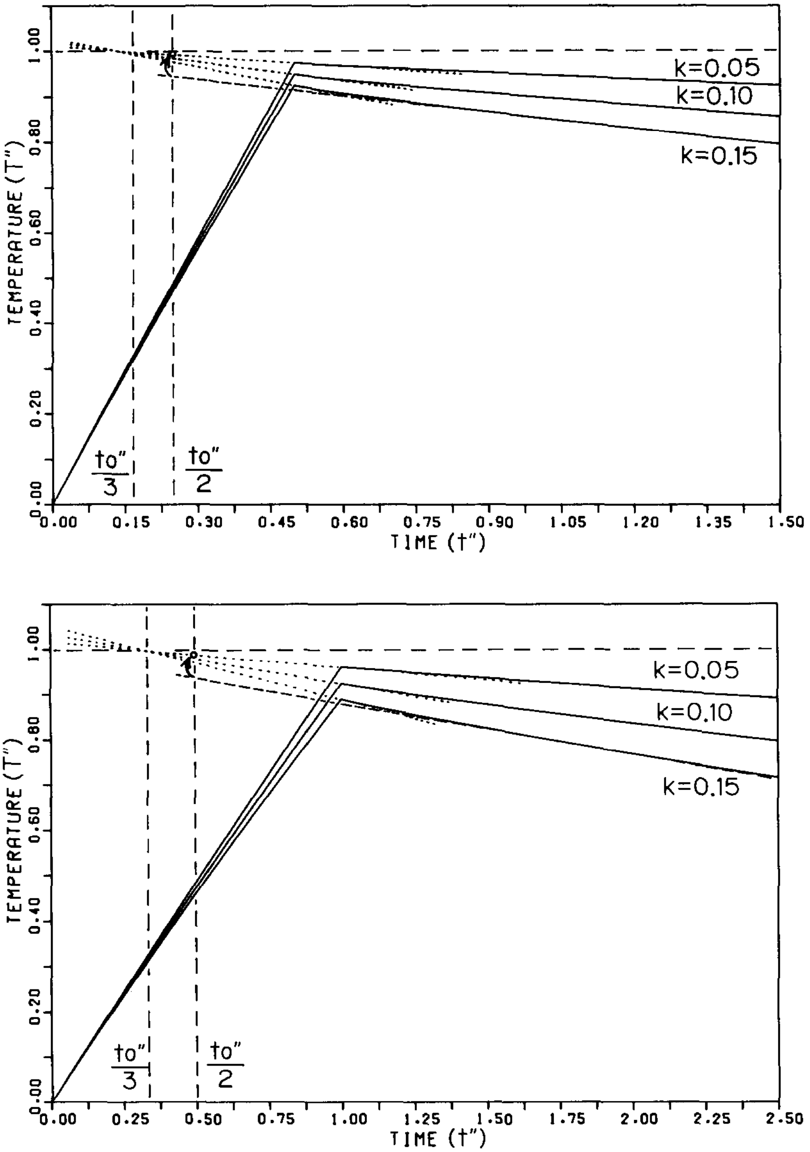

Fig. 4. Extrapolation method for the heat leak through conduction. Extrapolation to 1/3 of heating duration t_0'' of the initial slope of the cooling curve gives a good estimate of ideal temperature increment (dotted line). A more practical method is the extrapolation to $t_0''/2$ (broken line) and division by $(1-k/3)$ (arrow), where k is the heat capacity ratio of the supporting material to the sample. The latter method is only shown for a case of larger k to avoid confusion in the figure.

The extrapolation method discussed above is rather impractical to be applied to the analysis of experiments, because the gradient of the cooling curve changes rapidly immediately after t_0'' (as shown in Fig. 4) and such rapid change may not be measured accurately because of the possible delay in the response of the thermometer.

Another practical extrapolation method can be deduced from a different approximation applied to eqn. (21). Equation (21) is expanded to the second order of k as

$$\{f(t'')\} \sim \frac{1}{s^2}\left[1 - k\frac{1}{\sqrt{s}}\coth(\sqrt{s}) + k^2\left(\frac{1}{\sqrt{s}}\coth(\sqrt{s})\right)^2\right] \tag{29}$$

Equation (29) is inverse-transformed as

$$\begin{aligned}\{f(t'')\} \sim \{t''\} - k\{t''\}\cdot\left\{1 + 2\sum_{n=1}^{\infty}\exp(-\pi^2 n^2 t'')\right\} \\ + k^2\{t''\}\cdot\left[\left\{1 + 2\sum_{m=1}^{\infty}\exp(-\pi^2 m^2 t'')\right\}\right. \\ \left.\left\{1 + 2\sum_{n=1}^{\infty}\exp(-\pi^2 n^2 t'')\right\}\right]\end{aligned} \tag{30}$$

where dot-product means the convolution of Miksinski [7] defined as

$$\{a(t)\}\cdot\{b(t)\} = \left\{\int_0^t a(t-\tau)b(\tau)d\tau\right\} \tag{31}$$

The convolution and summation in eqn. (30) can be easily performed, which gives the following approximate expression for $t'' \gtrsim 0.5$

$$\begin{aligned}\{f(t'')\} \sim \Big\{\left(\tfrac{1}{45}k - \tfrac{2}{189}k^2\right) + \left(1 - \tfrac{1}{3}k + \tfrac{1}{15}k^2\right)t'' \\ + \left(-\tfrac{1}{2}k + \tfrac{1}{3}k^2\right)t''^2 + \tfrac{1}{6}k^2 t''^3\Big\}\end{aligned} \tag{32}$$

where $\exp(-\pi^2 n^2 t'')$ is neglected in the calculation of the coefficient of k^2 compared with unity for $n \geqslant 1$ and $t'' \gtrsim 0.5$ because $\exp(-\pi^2 \times 0.5)$, for example, is 0.0072 and negligible.

Some results of the approximate formula (32) are compared in Fig. 5 with the numerical results obtained by the FILT method [9] applied to the exact formula (21). The approximation is quite good for $0.5 \lesssim t'' \lesssim 4$ and $k \lesssim 0.1$ as shown in Fig. 5. The cooling curve after t_0'' is, therefore, given in the present approximation from eqns. (20) and (32) as

$$\begin{aligned}\{T''(1, t'')\} \sim \Big\{\left(1 - \tfrac{1}{3}k + \tfrac{1}{15}k^2\right) + \left(-\tfrac{1}{2}k + \tfrac{1}{3}k^2\right)(t_0'' - 2t'') \\ + \frac{1}{6t_0''}k^2\left[t''^3 - (t'' - t_0'')^3\right]\Big\}\end{aligned} \tag{33}$$

A linear extrapolation of eqn. (33) to $t'' = t_0''/2$ gives $T''(1, t'') = 1 - k/3$, leaving only error terms of the order of k^2. The cooling curve after about $t_0'' + 0.5$ should, therefore, be extrapolated to $t_0''/2$ and the resultant temper-

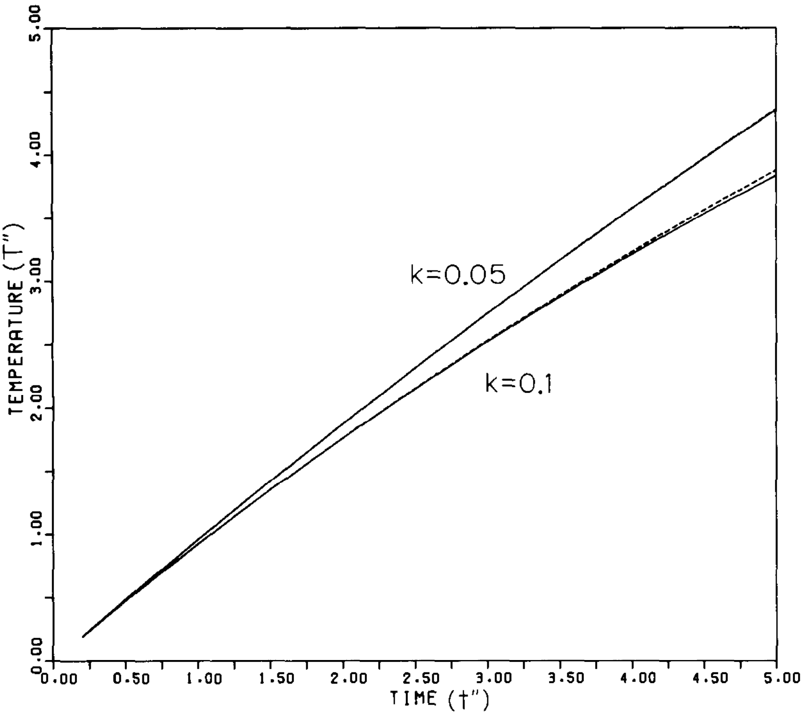

Fig. 5. Numerical result of the exact formula (21) for the heating process (solid curve) is well reproduced by the approximate formula (32) (broken curve).

ature increment should be divided by a factor $(1-k/3)$, which gives a satisfactory estimate of the increment in the ideal adiabatic condition as shown in Fig. 4 and Table 1.

The physical origin of the correction factor is rather simple. Spatial profile of the temperature in the electrical lead is well approximated by a

TABLE 1

Corrected temperature increment obtained by the practical extrapolation to $t_0''/2$ and division by $(1-k/3)$ in the case of heat leak through conduction, where k is the heat capacity ratio of the supporting material to the sample. The extrapolation line is fitted to the cooling curve between $t_0''+0.5$ and $t_0''+1.0$ by the least-squares method (see broken line in Fig. 4)

k	$\frac{T''(1, t'' \to t_0''/2)}{1-k/3}$	
	$t_0''=0.5$	$t_0''=1.0$
0.05	0.9991	0.9985
0.10	0.9965	0.9944
0.15	0.9928	0.9885

quadratic function of x' in the heating process in view of the result of the FILT calculation. It means that extra heat is necessary to raise the temperature of the electrical lead, and the heat is approximated by one third of the heat capacity of the electrical lead multiplied by the temperature increment in the sample. It suggests that the correction factor may be applicable to physical situations where heating of the electrical lead is expected with the temperature of the end point constant, even if the initial condition is not strictly the same as the one discussed here.

The correction parameter $k = clS_c/H_0$ is small in common experiments and can be easily estimated from the specific heat, length, and cross section, of lead wires (and supporting materials), and from the approximate heat capacity of the sample obtained from the experiment itself without correction. The correction is, of course, important in samples with small heat capacity.

HEAT LEAKS BY RADIATION AND CONDUCTION, AND PRACTICAL CONSIDERATIONS

The solution in the general case with radiation and conduction is shown to be expressed in a similar formula in the Appendix, and leads to eqns. (7) and (A5) [or (21)] in the two limiting cases of $\tau_r \to 0$ and $\tau_r \to \infty$, respectively. The accuracy of the limiting formulas can be checked by comparison of the numerical result of eqn. (6) or the numerical FILT calculation [9] applied to eqn. (A5) with the FILT results of the exact expressions (A1) or (A4), respectively. The accuracy in the temperature at $t' = 1$ calculated from the limiting formula is, for example, within 0.5% in a radiation dominant case where $\tau_r/\tau_c = 0.0012$, $\tau_i/\tau_r = 36$, $k = clS_c/H_0 = 0.043$, and $t_0' = 0.5$. τ_c is defined by (A3). The case corresponds roughly to 30 s heating of a 0.5 cm cube copper sample with unit emissivity in a heat bath of $T_0 = 570$ K with six $\varnothing 0.1 \times 10$ cm lead wires of stainless steel. The accuracy in the temperature at $t''' \ (\equiv t/\tau_c) = 1$ is within 0.015% in a conduction-dominant case where $\tau_r/\tau_c = 56$, $\tau_i/\tau_r = 0.00068$, $k = 0.038$, and $t_0''' = 0.05$. The case corresponds roughly to 30 s heating of the same sample at $T_0 = 4.2$ K. In the analysis of experiments, τ_i is estimated from the known thermal conductivity κ and the specific heat c of electrical leads. τ_c is estimated from κ, c and the heat capacity of the sample H_0. τ_r may be estimated from the excess cooling rate of the sample after several times of τ_i as compared with the rate expected from τ_c.

It is difficult to analyze the solution in the intermediate case by such approximate analytical methods as discussed above. The heat capacity of lead wires (and supporting materials) should, therefore, be made small enough in the intermediate case to be neglected when compared with that of the sample, and the cooling curve should be extrapolated to $t_0/2$.

Another practical consideration is worth mentioning concerning the initial condition of the radiation case. The extrapolation method is based on the initial condition T' $(t'=0)=0$, that is initial thermal equilibrium between the sample and the heat bath. The initial condition can be relaxed to allow intermittent heating in a heat bath of constant temperature, which is adopted in conventional experiments. We assume that first heating is initiated at t'_{init} (<0), is interrupted at $t'_{init}+t'_0$, and second heating is resumed again at $t'_{init}+t'_1$, and so on. The m-th heating is resumed at $t'=0$ $(=t'_{init}+mt'_1)$ and interrupted at t'_0. The temperature for $t'_0<t'<t'_1$ can be approximated by

$$T' \sim \{f(t')\} - \{f(t'-t'_0)\}U(t'-t'_0) + \frac{1}{t'_0}(a+bt'+ct'^2) \qquad (34)$$

where $\{f(t')\}$ is given by eqn. (7), because the temperature is shown to be expressed as

$$T' = \frac{1}{t'_0}\sum_{n=0}^{\infty}\Big(\{1-\exp[-(t'-nt'_1-t'_{init})]\}U(t'-nt'_1-t'_{init})$$
$$-\{1-\exp[-(t'-nt'_1-t'_0-t'_{init})]\}U(t'-nt'_1-t'_0-t'_{init})\Big) \qquad (35)$$

The extrapolation of the tangential line at $t'=0$ to $t'=t'_0/2$ gives $a/t'_0+b/2$, and the extrapolation of the tangential line at $t'=t'_0$ to $t'=t'_0/2$ gives $1+a/t'_0+b/2$ to the order of t'_0. The ideal temperature increment $\Delta T'=1$ is, therefore, obtained with an error of the order of $t_0'^2$ by taking the difference of the two extrapolation lines at $t'_0/2$. A corresponding consideration may not be applicable to the conduction case in practice, because the gradient of the cooling curve changes rapidly immediately after t''_0 as discussed above.

ACKNOWLEDGMENT

The numerical FILT calculation was made with the use of subroutine DLAPS3 of Fujitsu SSLII in Nagoya University Computation Center.

APPENDIX

The solution in the general case with radiation and conduction can be shown to be expressed in a similar formula to eqns. (7) and (20) with substitution of the following expression for $\{f(t')\}$

$$\{f'(t')\} = \frac{1}{s}\,\frac{\sinh\left(\sqrt{k_2 s}\right)}{(1+s)\sinh\left(\sqrt{k_2 s}\right) + k_1\sqrt{(k_2 s)}\cosh\left(\sqrt{k_2 s}\right)} \qquad (A1)$$

where time is normalized by τ_r as in eqn. (4), and parameters k_1 and k_2 are defined as

$$k_1 = \tau_r/\tau_c \qquad k_2 = \tau_i/\tau_r \tag{A2}$$

τ_c is the characteristic relaxation time of sample cooling by conduction

$$\tau_c = Hl/\kappa S_c \tag{A3}$$

The limit of eqn. (A1) with $\tau_r \to 0$ coincides with the operator expression of eqn. (6) in the radiation-dominant case in consideration of $k_1\sqrt{k_2} \to 0$.

Equation (A1) is conveniently rewritten for comparison with eqns. (20) and (21) of the conduction-dominant case as

$$\{f'(t''')\} = \frac{1}{s} k_1 \frac{\sinh\left(\sqrt{k_1 k_2 s}\right)}{(1 + k_1 s)\sinh\left(\sqrt{k_1 k_2 s}\right) + k_1\sqrt{(k_1 k_2 s)}\cosh\left(\sqrt{k_1 k_2 s}\right)} \tag{A4}$$

where time t''' is normalized by τ_c. The limit of eqn. (A4) with $\tau_r \to \infty$ coincides with the following modification of eqn. (21) in consideration of $k_1 k_2 = clS_c = k$

$$\{f'(t''')\} = \frac{1}{s} \frac{\sinh(\sqrt{ks})}{s \cdot \sinh(\sqrt{ks}) + \sqrt{ks}\cosh(\sqrt{ks})} \tag{A5}$$

where time t''' is normalized by τ_c as in eqn. (A4).

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