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1999 Jpn. J. Appl. Phys. 38 2988

(http://iopscience.iop.org/1347-4065/38/5R/2988)

Jpn. J. Appl. Phys. Vol. 38 (1999) pp. 2988–2992
Part 1, No. 5A, May 1999

Two-Dimensional Effects on Measurement of Thermal Diffusivity by AC Calorimetric Method: I. Conditions for Precise Measurement

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(Received January 6, 1999; accepted for publication February 26, 1999)

In the application of an ac calorimetric method to precise thermal diffusivity measurement of a thin material, one-dimensional temperature wave propagation is required. In a relatively thick material, one-dimensional temperature wave propagation is deformed and in the strict sense, two-dimensional temperature wave propagation should be taken into account. In the present paper, we propose the condition of the maximum thickness of a material required to measure thermal diffusivity within an accuracy of ±2% in the framework of one-dimensional temperature wave propagation, based upon the results obtained analytically for the two-dimensional temperature wave propagation.

KEYWORDS: ac calorimetry, thermal diffusivity, thick material, experimental condition, thermophysical properties

1. Introduction

An ac calorimetric method has been extensively applied to thermal diffusivity measurement of a variety of thin materials from polymer to diamond.[1–3] In this method, chopped light is partly applied to the front or the rear surface of a sample and the irradiated part is moved along the surface (see Fig. 1). When the thickness of a sample, d, is thin enough, one-dimensional temperature wave propagation takes place along the surface. To measure ac temperature, a thermocouple with a diameter smaller than the thermal diffusion length is attached to the front or the rear surface. The decay or the phase shift of ac temperature is measured as a function of the relative displacement at a position in $|x| \geq w$. From the measurement of the ac temperature we obtain the thermal diffusivity. This method has the following merits.

1. The attachment of a thermocouple does not affect the precision of the measurement at all.[4]
2. The effect of heat loss, which is dominant in the measurement of materials with low thermal diffusivity such as a polymer film, can be corrected analytically.[2,5]
3. The edge effects that occur at the boundary of materials with high thermal diffusivity, such as a diamond plate, can be corrected analytically.[3,6]

To carry out the measurement, there is a limitation on the thickness of a platelike sample, since in the above method, one-dimensional temperature wave propagation along the surface of a sample is assumed. For the thickness of a sample, d, the following condition is roughly required to attain one-dimensional temperature wave propagation:[1]

$$kd \ll 1, \tag{1}$$

where both the temperature wave number and the temperature decay constant are given by the same quantity, k, as[7,8]

$$k = \sqrt{\frac{\omega}{2D}}. \tag{2}$$

In eq. (2), ω is the angular frequency and D is the thermal diffusivity of a sample.

When the thickness of a sample, d, is too large, a temperature gradient across a sample appears and then, we are no longer able to regard the plate-like sample as a one-dimensional system. Yamane *et al.*[9] considered such a case in which two-dimensional temperature wave propagation takes place in a platelike sample. They analyzed the behavior of ac temperature waves in the cross section given in Fig. 1 based upon the Fourier transform of the impulse response. Then, they pointed out that, if a thermal diffusivity measurement is performed in the region of $x/(d/\pi) > 100$, where x is the axis along the surface of a platelike sample with its origin at the edge of the irradiated part, the expression for one-dimensional ac temperature wave propagation can be applied to the analysis of thermal diffusivity along the x-direction. However, the above region should depend not only on ω but also on D. We need a general condition for measuring thermal diffusivity along the x-direction. As seen in eq. (2), by using ω and D we can express characteristic length, $1/k$, which is called the thermal diffusion length, in ac temperature wave propagation. Therefore, it is reasonable to consider the condition for the magnitude of kd to measure thermal diffusivity along the x-direction in a platelike sample with thickness d. In the present paper, by solving the differential equation of heat conduction in a two-dimensional system, such as a platelike sample, under the boundary conditions given in Fig. 1, we will propose the required magnitude of kd for measuring thermal diffusivity within an accuracy of ±2%.

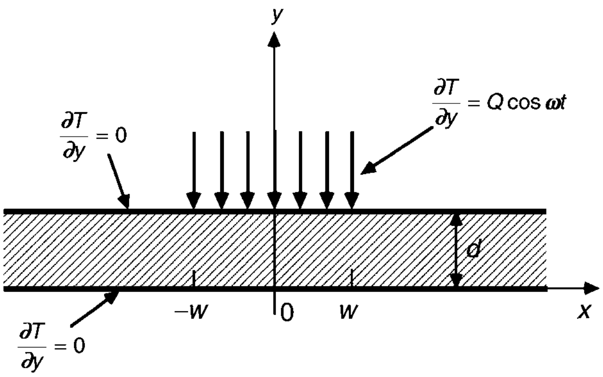


Fig. 1. A system for a platelike material. The thickness is d and the modulated heat is applied in the region from $x = -w$ to w. The boundary conditions are given in the illustration. For the units of Q, see text.

2. Theoretical Analyses

The heat conduction equation in a two-dimensional system is given by

$$\frac{\partial T(x,y,t)}{\partial t} = D\left(\frac{\partial^2 T(x,y,t)}{\partial x^2} + \frac{\partial^2 T(x,y,t)}{\partial y^2}\right). \quad (3)$$

First, we will analyze the temperature distribution in the system shown in Fig. 1. The boundary conditions are

$$\frac{\partial T(x,y,t)}{\partial y} = 0, \quad \text{at } y = 0, \quad (4)$$

$$\frac{\partial T(x,y,t)}{\partial y} = 0, \quad \text{at } y = d \text{ and in } |x| > w, \quad (5)$$

$$\frac{\partial T(x,y,t)}{\partial y} = Q\cos\omega t, \quad \text{at } y = d \text{ and in } |x| \le w, \quad (6)$$

where Q is the applied heat flux divided by the thermal conductivity of a material. As can be seen from eqs. (4) and (5), in the present analysis we assume that there is no heat leakage to the surroundings. To solve eq. (3), let us define $T(x, y, t)$ as

$$T(x,y,t) = \mathrm{Re}\{\theta(x,y)\exp(\mathrm{i}\omega t)\}. \quad (7)$$

Then, we can rewrite eq. (3) as

$$2\mathrm{i}k^2\theta(x,y) = \frac{\partial^2\theta(x,y)}{\partial x^2} + \frac{\partial^2\theta(x,y)}{\partial y^2}. \quad (8)$$

Let us define a Fourier transform and its inverse Fourier transform, respectively, by

$$\hat{\theta}(s,y) = \int_{-\infty}^{\infty}\theta(x,y)\exp(-\mathrm{i}sx)\mathrm{d}x, \quad (9)$$

$$\theta(x,y) = \frac{1}{2\pi}\int_{-\infty}^{\infty}\hat{\theta}(s,y)\exp(\mathrm{i}sx)\mathrm{d}s. \quad (10)$$

Substituting eq. (10) into eq. (8), we can obtain

$$\frac{\partial^2\hat{\theta}(s,y)}{\partial y^2} - \xi^2\hat{\theta}(s,y) = 0, \quad (11)$$

where $\xi^2 = s^2 + 2\,\mathrm{i}k^2$ and $\mathrm{Re}\xi$ is positive. We can solve the differential equation of eq. (11) with the boundary conditions of eqs. (4)–(6) as

$$\hat{\theta}(s,y) = \frac{2Q\sin sw\cosh\xi y}{\xi s\sinh\xi d}. \quad (12)$$

As a result, from the inverse Fourier transform of eq. (12) using eq. (10) we obtain $\theta(x, y)$ as

$$\theta(x,y) = \frac{Q}{\pi}\int_{-\infty}^{\infty}\exp(\mathrm{i}sx)\frac{\sin sw\cosh\xi y}{\xi s\sinh\xi d}\mathrm{d}s. \quad (13)$$

Finally, since we wish to obtain $T(x, y, t)$, let us define the following relation:

$$\theta(x,y) = R(x,y)\exp\{\mathrm{i}\phi(x,y)\}. \quad (14)$$

Then, $T(x, y, t)$ is given by

$$T(x,y,t) = R(x,y)\cos\{\omega t + \phi(x,y)\}. \quad (15)$$

It should be stressed that as given in eq. (13), by integration we can obtain $\theta(x, y)$ without any approximation and finally get $T(x, y, t)$. This is a useful result for considering explicitly the experimental conditions required for precise measurement.

3. Results of Analyses

In the numerical calculations, the important quantities are the thickness d, the half-width of the irradiated part w, the thermal diffusivity D and the angular frequency ω. For both Q and T, we need only these relative values, and in the analyses we chose them arbitrarily. Furthermore, both D and ω are related through eq. (2). Then, we used k instead of D and ω. As typical examples, we show the distributions of ac temperature waves in the two-dimensional cross section in Fig. 1 for $d = 0.005\,\mathrm{m}$, $w = 0.01\,\mathrm{m}$, $k = 1\,\mathrm{m}^{-1}$ and $d = 0.005\,\mathrm{m}$, $w = 0.01\,\mathrm{m}$, $k = 10\,\mathrm{m}^{-1}$ in Figs. 2 and 3, respectively. For these cases, the isothermal contours are drawn only to guide the eyes so that the contours in both figures lie nearly with the same separation on the right side. As seen in Figs. 2 and 3, the contours on the right side are almost normal to the upper and lower surfaces. Therefore ac temperature waves seem to propagate in a manner close to one-dimensional behavior. In the following we will consider these results in detail.

Based upon the result of $T(x, y, t)$ for $d = 0.01\,\mathrm{m}$ and $k = 1\,\mathrm{m}^{-1}$, we draw $\log R$ as a function of x at $y = 0$ and $y = d$ in Fig. 4 and ϕ as a function of x at $y = 0$ and $y = d$ in Fig. 5. From them, the derivative of $\log R$ with respect to x,

$$k_{\mathrm{a,app}} = -\frac{\partial\log R}{\partial x}, \quad (16)$$

divided by k is plotted in Figs. 6(a) and (b) for $k = 1\,\mathrm{m}^{-1}$ and $100\,\mathrm{m}^{-1}$, respectively, and the derivative of ϕ,

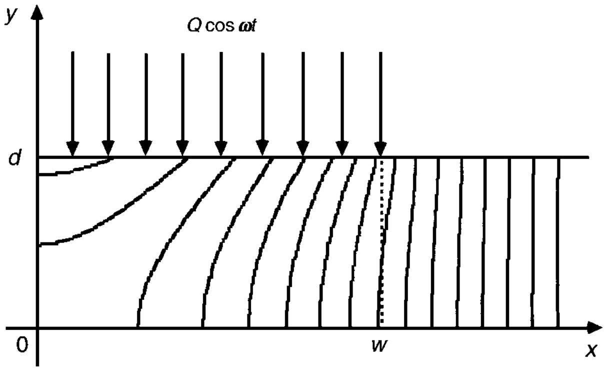


Fig. 2. Isothermal contours in a two-dimensional cross section for the model when $d = 0.005\,\mathrm{m}$, $w = 0.01\,\mathrm{m}$ and $k = 1\,\mathrm{m}^{-1}$.

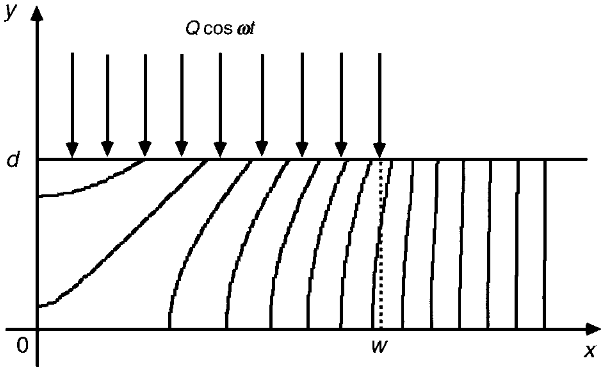


Fig. 3. Isothermal contours in a two-dimensional cross section for the model when $d = 0.005\,\mathrm{m}$, $w = 0.01\,\mathrm{m}$ and $k = 10\,\mathrm{m}^{-1}$.

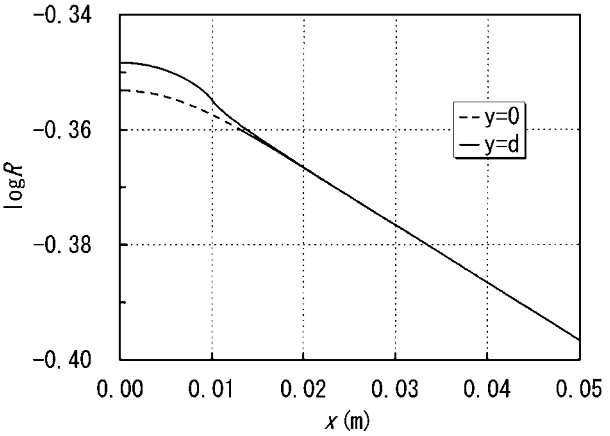


Fig. 4. Relation for log$R$ versus x at $y = 0$ and $y = d$, when $d = 0.01$ m, $w = 0.01$ m and $k = 1\,\mathrm{m}^{-1}$. The slope becomes linear slightly above $x = 0.01$ m ($= w$) and it is equal to $k = 1\,\mathrm{m}^{-1}$.

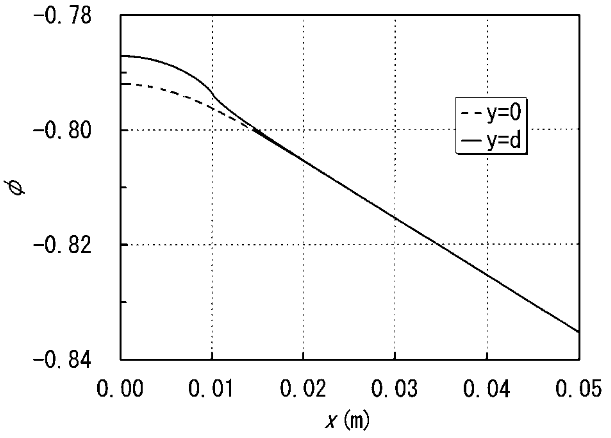


Fig. 5. Relation for ϕ versus x at $y = 0$ and $y = d$ when $d = 0.01$ m, $w = 0.01$ m and $k = 1\,\mathrm{m}^{-1}$. The slope becomes linear slightly above $x = 0.01$ m ($= w$) and it is equal to $k = 1\,\mathrm{m}^{-1}$.

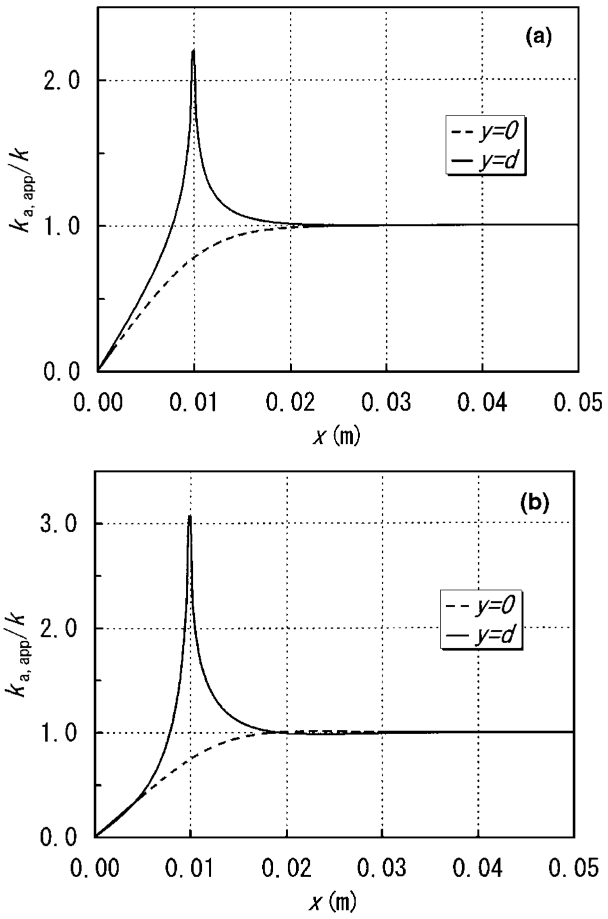


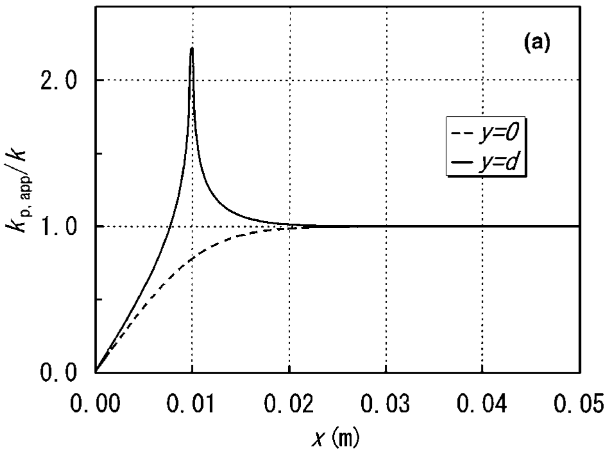
Fig. 6. Ratio of the apparent decay constant $k_{\mathrm{a,app}}$ and k as a function of x at $y = 0$ and $y = d$ when $d = 0.01$ m and $w = 0.01$ m. (a) for $k = 1\,\mathrm{m}^{-1}$ and (b) for $k = 100\,\mathrm{m}^{-1}$.

$$k_{\mathrm{a,app}} = -\frac{\partial \phi}{\partial x}, \tag{17}$$

divided by k in Figs. 7(a) and (b) for $k = 1\,\mathrm{m}^{-1}$ and $100\,\mathrm{m}^{-1}$, respectively. In Figs. 4 and 5, both the slopes, $k_{\mathrm{a,app}}$ and $k_{\mathrm{p,app}}$, of log$R$ versus x and ϕ versus x, respectively, are linear, i.e., as seen in Figs. 6(a) and (b) and 7(a) and (b) both $k_{\mathrm{a,app}}/k$ and $k_{\mathrm{p,app}}/k$, respectively, become close to unity in the region where $x > w$ (and $x < -w$) where $w = 0.01$ m. This indicates that one-dimensional temperature wave propagation holds in the region of $x > w$ (and $x < -w$). As seen from the behavior of $k_{\mathrm{a,app}}/k$ and $k_{\mathrm{p,app}}/k$ in Figs. 6(a) and (b) and Figs. 7(a) and (b), respectively, the deviation from k is significant at $x = w$, i.e., the apparent thermal diffusivity deviates from the true thermal diffusivity markedly at $x = w$. Therefore, if the condition guarantees one-dimensional temperature wave propagation at $x = w$, the one-dimensional temperature wave propagation holds in the entire region of $x \geq w$ (and $x \leq -w$). This means that, if we verify the conditions for measuring thermal diffusivity within an accuracy of ±1% at $x = w$ (and $x = -w$), we can obtain thermal diffusivity with much higher accuracy in the region of $x > w$ (and $x < -w$). Next, we will briefly discuss the particular behaviors of the curve, $k_{\mathrm{a,app}}/k$ versus x or $k_{\mathrm{p,app}}/k$ versus x, near the edge of the heated region of $x = w$ in Figs. 6(a) and (b) and 7(a) and (b) in the following.

First at $x = 0$, in all of the curves $k_{\mathrm{a,app}}/k$ and $k_{\mathrm{p,app}}/k$ become almost zero. This means that at the center of the heated region, $x = 0$, ac temperature waves propagate almost parallel to the y-direction. At the edge of the heated region, $x = w$ (and $x = -w$) peaks appear in $k_{\mathrm{a,app}}/k$ and $k_{\mathrm{p,app}}/k$ when ac temperature is detected on the front surface while on the other hand, gentle slopes appear on the rear surface. It is much easier to understand the above behavior based upon the curves in Figs. 4 and 5. If only one-dimensional temperature wave propagation takes place, the slopes of log$R$ versus x and ϕ versus x are constant in the region of $x \geq w$ and bend downward as x decreases in $0 < x < w$.[1] Then, the curve of $k_{\mathrm{a,app}}/k$ versus x or $k_{\mathrm{p,app}}/k$ versus x in one-dimensional temperature propagation lie between the solid curve and the dotted curve both in Figs. 4 and 5. Owing to the interference of ac temperature waves, the solid curve deviates upward and the dotted curve deviates markedly downward. As a result, the characteristic behaviors of the curve, $k_{\mathrm{a,app}}/k$ versus x or $k_{\mathrm{p,app}}/k$ versus x, near the edge of the heated region take place.

Then, let us consider the experimental condition at the end points of the irradiated region, i.e., $(x, y) = (w, d)$ and $(w, 0)$. At the front surface of $(x, y) = (w, d)$, generally we observe that the apparent thermal diffusivity is larger than the true thermal diffusivity, as can be seen in the behavior of $k_{\mathrm{a,app}}/k$



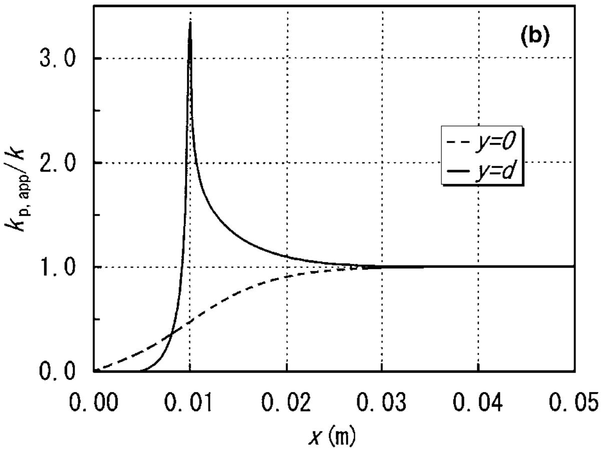


Fig. 7. Ratio of the apparent wave number $k_{\mathrm{p,app}}$ and k as a function of x at $y = 0$ and $y = d$ when $d = 0.01$ m and, $w = 0.01$ m. (a) for $k = 1\,\mathrm{m}^{-1}$ and (b) for $k = 100\,\mathrm{m}^{-1}$.

and $k_{\mathrm{p,app}}/k$ versus x in the region of $x \geq w$ of Figs. 6(a) and 6(b), and the apparent thermal diffusivity is largest at $x = w$. Then, we will search for the condition under which the apparent thermal diffusivity is 2% larger than the true thermal diffusivity at $x = w$ numerically, i.e., this is the condition for the thermal diffusivity measurement within an accuracy of ±2% at $(x, y) = (w, d)$. In Fig. 8, at $k_{\mathrm{a,app}}/k = 1.01$ or $k_{\mathrm{p,app}}/k = 1.01$, kd is drawn as a function of kw by a single curve, since both $k_{\mathrm{a,app}}/k$ and $k_{\mathrm{p,app}}/k$ are expressed as functions of kd and kw which are derived from eq. (13). At large kw, kd is 0.31 for $k_{\mathrm{a,app}}/k$. If d becomes smaller, the condition for one-dimensional temperature wave propagation holds more strongly. Therefore, the condition that we can attain the thermal diffusivity measurement within an accuracy of 2% is given by

$$kd \leq 0.31 \tag{18}$$

for the measurement at the front surface. At $k_{\mathrm{p,app}}/k = 1.01$, the same analysis can be made, then the condition under which we can attain the thermal diffusivity measurement within an accuracy of 2% is given by

$$kd \leq 0.02. \tag{19}$$

On the other hand, at the rear surface of $(x, y) = (w, 0)$ the apparent thermal diffusivity is smaller than the true thermal diffusivity. Then, we will search for the severest condition under which the apparent thermal diffusivity is 2% smaller than the true thermal diffusivity. In Fig. 9, at $k_{\mathrm{a,app}}/k = 0.99$ or $k_{\mathrm{p,app}}/k = 0.99$, kd is plotted as a function of kw. At large kw, kd is 0.55 for $k_{\mathrm{a,app}}/k$. If d becomes smaller, the condition for one-dimensional temperature wave propagation holds more sufficiently. Therefore, the condition that we can attain the thermal diffusivity measurement within an accuracy of 2% is given by

$$kd \leq 0.55 \tag{20}$$

for the measurement at the rear surface. At $k_{\mathrm{p,app}}/k = 0.99$, the same analysis can be made, then the condition in which we can attain the thermal difffusivity measurement within an accuracy of 2%, is given by

$$kd \leq 0.03. \tag{21}$$

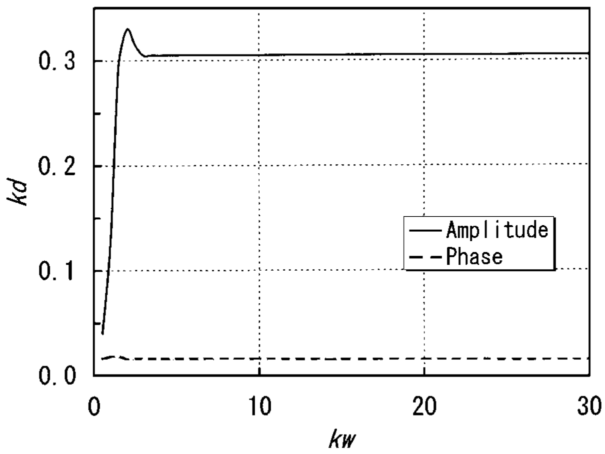


Fig. 8. Relation of kd versus kw when $k_{\mathrm{a,app}}/k = 1.01$ (solid line) and $k_{\mathrm{p,app}}/k = 1.01$ (dashed line) at the front surface. Generally the measurement is performed at a sufficiently large w and therefore, kd becomes flat.

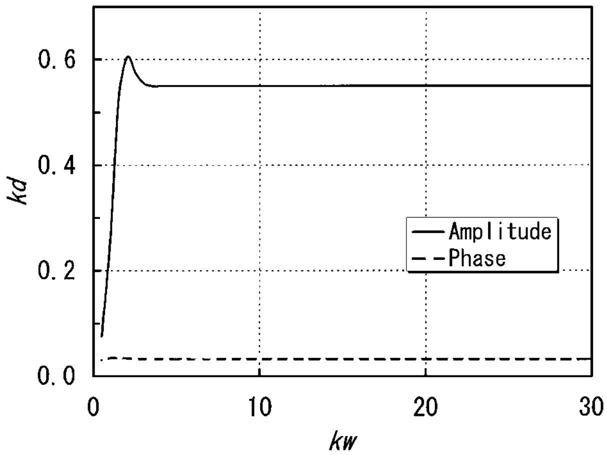


Fig. 9. Relation of kd versus kw when $k_{\mathrm{a,app}}/k = 0.99$ (solid curve) and $k_{\mathrm{p,app}}/k = 0.99$ (dashed line) at the rear surface. Generally the measurement is performed at a sufficiently large w and therefore, kd becomes flat.

4. Discussion

As shown above, the condition for the thickness of a sample required for thermal diffusivity measurement using an ac calorimetric method depends on whether the ac temperature detection is carried out at the rear or the front surface of a sample and also whether we use the amplitude or the phase of the ac temperature. When thermal diffusivity measurement is performed under the conditions that the ac temperature is detected at the rear surface of a sample and the amplitude of the ac temperature is used for the analysis, as seen in eq. (20), at 1 Hz the maximum thickness of the samples is calculated as

$$\begin{aligned} d_{\max} &= 3\,\text{mm} && \text{for aluminum,} \\ d_{\max} &= 1\,\text{mm} && \text{for Al}_2\text{O}_3\text{ ceramics,} \\ d_{\max} &= 0.1\,\text{mm} && \text{for polyethylene.} \end{aligned}$$

If we use 0.01 Hz for the measurement, the maximum thickness becomes ten times as thick as the above values. Therefore, the ac calorimetric method to measure thermal diffusivity can be applied not only to thin samples but also to relatively thick samples, where the sample thickness is comparable to those required to use a laser flash method.

The present results can be easily extended to a case in which there is anisotropy in the thermal diffusivity in the x- and y-directions of a material. In such a case, according to the degree of anisotropy the scale of the x- and y-directions should be modified.

Finally, it should be noted that the conditions for thermal diffusivity measurement in an ac calorimetric method cannot be applied to the condition for heat capacity measurement in an ac calorimetric method. This will be discussed in another paper.

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