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Thermal Diffusivity of Semitransparent Materials Determined by the Laser-Flash Method Applying a New Analytical Model[1]

H. Mehling,[2,3] G. Hautzinger,[2] O. Nilsson,[2] J. Fricke,[2] R. Hofmann,[4] and O. Hahn[4,5]

We have developed an analytical model to determine the thermal diffusivity of nonscattering materials from samples with low optical thickness and opaque boundaries with arbitrary emissivities. The paper outlines the new analytical model and describes measurements on two samples: a microscope slide glass and a high-grade fused quartz plate. Results show that the new model applied to measurements on gold- or graphite-coated samples leads to the same results as if a conventional model is used on gold-coated samples.

KEY WORDS: analytical model; glass; laser-flash method; radiative heat transfer; thermal conductivity; thermal diffusivity.

1. INTRODUCTION

The laser-flash method is a widely used method to determine the thermal diffusivity of materials at high temperatures. It is based on the assumption that the laser pulse is absorbed at the front surface of the sample and that the heat is transported via purely diffusive processes to the sample rear surface, where the temperature rise is monitored by an infrared detector.

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[2] Bavarian Center for Applied Energy Research (ZAE Bayern), Am Hubland, D-97074, Würzburg, Germany.

[3] To whom correspondence should be addressed.

[4] Fraunhofer Institut für Silicatforschung, Neunerplatz 2, D-97082 Würzburg, Germany.

[5] Present address: Max-Planck Institut für Polymerforschung, Ackermannweg 10, D-55122 Mainz, Germany.

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Analytical solutions of the diffusivity equation exist for adiabatic [1] or nonadiabatic boundary conditions [2, 3].

For many years research has been performed in order to extend the applicability of the laser-flash method to semitransparent samples [4–8]. Extensive numerical simulations of the coupled radiative/conductive heat transfer in semitransparent, nonscattering media with opaque, either highly reflecting or absorbing boundaries of different thicknesses have been done by Andre and Degiovanni [7]. Their results show that if the sample has highly reflecting boundaries and an optical thickness less than 0.1, conventional analytical models [1–3] will lead to the correct thermal diffusivity. This is easily understood, as due to the low optical thickness, thermal emission within the sample is negligible and radiative heat transfer between the sample surfaces is suppressed by their high reflectivity, i.e., low emissivity.

Opaque and highly reflecting boundaries are usually realized by coating the sample surfaces with a gold layer using physical vapor deposition (PVD). This is time-consuming and requires special equipment. Additionally, a graphite layer is necessary on both sides to absorb the laser pulse at the front side and to increase emission in the infrared at the rear side of the sample.

It would be advantageous if the boundary conditions were opaque, but otherwise arbitrary. In that case, a simple graphite coating would be sufficient. If the sample is still optically thin, thermal emission within the sample is negligible. But in addition to the diffusive heat transfer within the sample, there is now a direct radiative heat transfer between the front and the rear surfaces of the sample. Due to the low optical thickness of the sample, these two heat fluxes simply add linearly. This is therefore called the diathermic case: the two heat fluxes are not coupled by the temperature distribution within the sample but only by the temperatures at the boundaries. An approximate model for this case was first suggested by Blumm et al. [9]. We now outline an exact analytical solution for this case which is given in detail in Ref. 10.

2. ANALYTICAL SOLUTION

One-dimensional heat conduction is mathematically described by the Fourier equation

$$\frac{\partial T(x, t)}{\partial t} = \alpha \frac{\partial^2 T(x, t)}{\partial x^2} \tag{1}$$

with

$$T(x, t) := T_{\text{sample}}(x, t) - T_{\text{sur}} \tag{2}$$

Here $T_{\text{sample}}(x, t)$ is the sample temperature at point x and time t, T_{sur} the temperature of the surrounding and α the thermal diffusivity of the sample. If the pulse length of the laser and its penetration depth in the sample are short enough to be neglected, the initial temperature distribution within the sample can be described by a delta function. For adiabatic boundary conditions we have

$$\left.\frac{\partial T(x, t)}{\partial x}\right|_{x=0} = 0 \qquad \text{and} \qquad \left.\frac{\partial T(x, t)}{\partial x}\right|_{x=l} = 0 \tag{3}$$

Equations (1) and (3) lead to the solution of Parker et al. [1] for the rear-side temperature:

$$T(l, t) = T_\infty \left\{1 + 2 \sum_{n=1}^{\infty} (-1)^n \exp\left(-\frac{n^2\pi^2\alpha}{l^2} t\right)\right\} \tag{4}$$

Here T_∞ is the sample temperature after thermal equilibrium is reached within the sample in the adiabatic case and l is the sample thickness. The boundary conditions for heat losses at the front and rear surfaces of the sample, which are assumed to be linear in $T(T(0, t) \ll T_{\text{sur}}$ and $T(l, t) \ll T_{\text{sur}})$, can be written as

$$\left.\frac{\partial T(x, t)}{\partial x}\right|_{x=0} = hT(0, t) \qquad \text{and} \qquad \left.\frac{\partial T(x, t)}{\partial x}\right|_{x=l} = -hT(l, t) \tag{5}$$

instead of Eq. (3). For radiative heat losses

$$h = \frac{4\sigma\varepsilon T_{\text{sur}}^3}{\kappa} \tag{6}$$

where σ is the Stefan–Boltzmann constant, κ the thermal conductivity, and ε the emissivity of the sample. The corresponding solution is [3]

$$T(l, t) = 2T_\infty l \sum_{n=1}^{\infty} \beta_n \exp(-\alpha\beta_n^2 t) \frac{\beta_n \cos(\beta_n l) + h \sin(\beta_n l)}{(\beta_n^2 + h^2)\, l + 2h} \tag{7}$$

where β_n are the positive roots of

$$\tan(\beta_n l) = \frac{2\beta_n h}{\beta_n^2 - h^2}; \qquad \beta_1 = 0 \tag{8}$$

This is henceforth called the conventional model.

If we deal with a diathermic sample which has an opaque coating (e.g., graphite), the boundary conditions of Eq. (3) become

$$\left.\frac{\partial T(x,t)}{\partial x}\right|_{x=0} = hT(0,t) + h\eta(T(0,t) - T(l,t))$$
$$\left.\frac{\partial T(x,t)}{\partial x}\right|_{x=l} = -hT(l,t) - h\eta(T(0,t) - T(l,t)) \tag{9}$$

The term $h\eta(T(0,t) - T(l,t))$ is added to include the direct radiative heat transfer between the front and the rear surfaces of the sample. This direct radiative heat transfer is proportional to the emittances of the front and rear surfaces [included in h; see Eq. (6)], the location and width of the totally transparent spectral regions (fitted by the parameter η), and the temperature differences between the surfaces $T(0,t) - T(l,t) \leqslant T_{sur}$ assumed). As shown by Ref. 10, a detailed knowledge of the dependence of η on the optical properties is not necessary for the determination of the thermal diffusivity in the diathermic case. It can be shown [10] that Eqs. (7) and (8) remain valid for the diathermic case if the following substitution is used:

$$\begin{aligned} h &\to h && \text{for even } n \\ h &\to h(1+2\eta) && \text{for odd } n \end{aligned} \tag{10}$$

This can be written in a single expression:

$$h \to h[1 + \eta(1 + (-1)^{n+1})] \tag{11}$$

Comparison with numerical results and first measurements are given in Refs. 11 and 12. The result can also be expressed by the Biot number $N_{Bi} := hl$ and $y_n := \beta_n l$:

$$T(l,t) = 2T_\infty \sum_{n=1}^{\infty} y_n \exp\left(-\frac{\alpha y_n^2}{l^2} t\right) \frac{y_n \cos(y_n) + B_{Bi} \sin(y_n)}{y_n^2 + N_{Bi}^2 + 2N_{Bi}} \tag{12}$$

where y_n are the positive roots of

$$\tan(y_n) = \frac{2y_n N_{Bi}}{y_n^2 - N_{Bi}^2}; \qquad y_1 = 0 \tag{13}$$

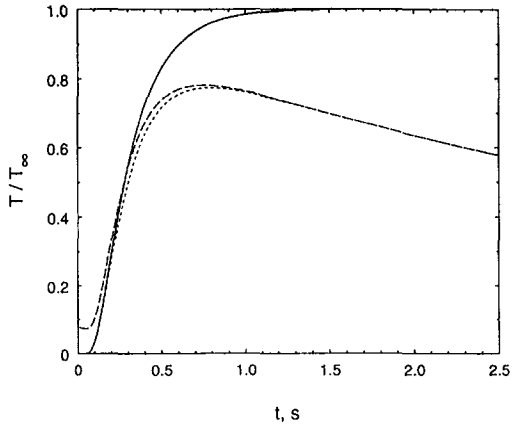

Fig. 1. Comparison of the laser-flash curves for a nontransparent sample $\eta = 0$ with Biot numbers $N_{Bi} = 0$ (solid line) and $N_{Bi} = 0.2$ (dotted line) as well as for a diathermic sample with $\eta = 0.4$ and $N_{Bi} = 0.2$ (dashed line). The thermal diffusivity was $\alpha = 0.5\ \text{mm}^2 \cdot \text{s}^{-1}$.

and the substitution

$$N_{Bi} \to N_{Bi}[1 + \eta(1 + (-1)^{n+1})] \tag{14}$$

for the diathermic case. Figure 1 shows a comparison of the laser-flash curves for nontransparent samples $\eta = 0$ with Biot numbers $N_{Bi} = 0$ and $N_{Bi} = 0.2$ as well as for a diathermic sample with $\eta = 0.4$ and $N_{Bi} = 0.2$. The thermal diffusivity was $\alpha = 0.5\ \text{mm}^2 \cdot \text{s}^{-1}$. The presence of the direct radiative heat transport between the front and the rear surfaces of the sample in the diathermic case leads to an immediate increase in the rear surface temperature. After the maximum temperature is reached, the difference in temperature between the diathermic and the nondiathermic case vanishes because the front and rear surfaces of the sample have the same temperature and therefore the direct radiative flux vanishes.

3. MEASUREMENTS

Two samples with absorption but no scattering of radiation have been investigated: a microscope slide glass and a high-grade fused silica platelet. The microscope slide glass has a content of 72.2% SiO_2, 14.3% Na_2O, 1.2% K_2O, 6.4% CaO, 4.3% MgO, and 1.2% Al_2O_3 and a thickness of $l = 1.01$ mm. The thickness of the high-grade fused silica sample was $l = 1.07$ mm.

The optical thickness of a sample of homogeneous thickness l consisting of a nonscattering material was determined from the measured normal transmittance T and reflectance R [13] using

$$R = \rho\left(1 + \frac{(1-\rho)^2\,\tau^2}{1-\rho^2\tau^2}\right) \tag{15}$$

and

$$T = \frac{(1-\rho)^2\,\tau}{1-\rho^2\tau^2} \tag{16}$$

These relations include multiple reflections but no interference.

$$\tau = \exp(-l/l_{\text{abs}}) \tag{17}$$

is the transmissivity within the material and ρ the reflectivity of the boundary according to Fresnel's formula. The ratio of the thickness of the sample l to the mean free path for absorption l_{abs} is equal to the optical thickness. Figure 2 shows the optical thickness for both samples. Both samples are optically thick for $\lambda \geqslant 5$ μm and optically thin for $\lambda \leqslant 2.6$ μm. The intermediate range might cause deviations from our ideal model of a diathermic sample.

The laser-flash measurements were performed with a Nd : Yag laser of 1064-nm wavelength. An MCT detector sensitive in the range from 3 to

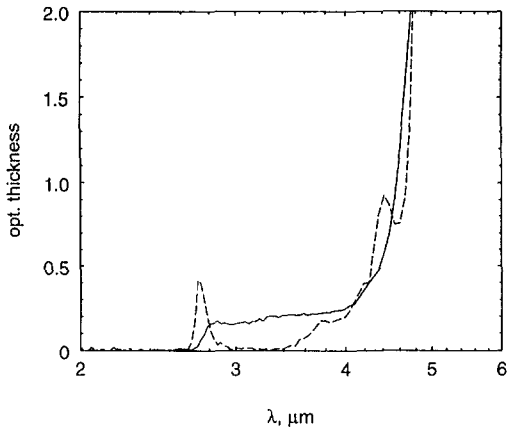

Fig. 2. Optical thickness of microscope slide glass (solid line) with a thickness of 1.01 mm and high-grade fused quartz (dashed line) with a thickness of 1.07 mm.

11 μm was used for monitoring the temperature rise at the rear side of the sample. At each temperature, the thermal diffusivity α was determined as the average over four individual laser-flash measurements.

4. RESULTS

4.1. Microscope Slide Glass

The derived thermal diffusivity of the microscope slide glass is shown in Fig. 3. Measurements on gold-coated samples lead to the same results no matter if the conventional model or the new one with direct radiative heat transfer included is used. This is due to the fact that direct radiative heat transfer is strongly suppressed by the low emissivity of the gold layer.

If no gold layer is applied, direct radiative heat transfer between the graphite layers occurs and increases with temperature. This leads to a systematic overestimation of the thermal diffusivity with increasing temperature (up to 10% at 773 K) if the conventional model is used. The evaluation with the new model provides the correct diffusivities, without the application of a gold layer.

4.2. High-Grade Fused Quartz

The second sample was a high-grade fused quartz platelet. Figure 4 shows basically the same features as for the microscope slide glass, except

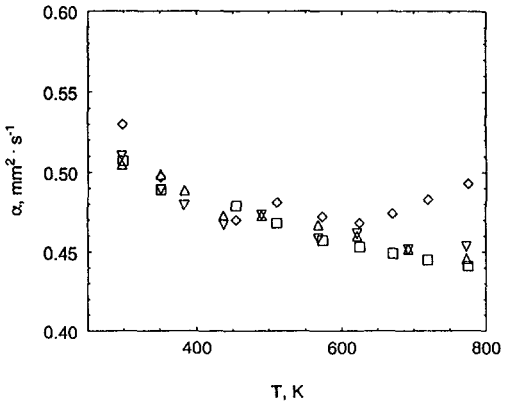

Fig. 3. Derived thermal diffusivity α of microscope slide glass: gold-coated evaluated with conventional (∇) and new ($\triangle$) model; only graphite coated evaluated with conventional ($\diamond$) and new ($\square$) model.

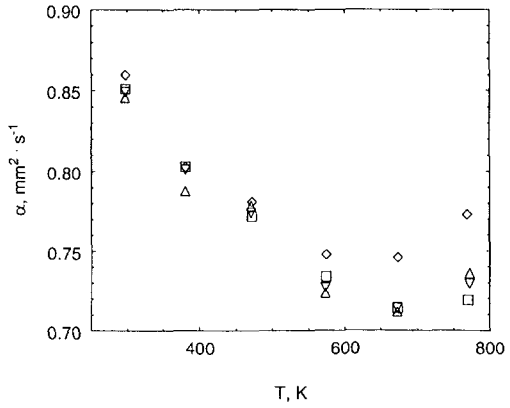

Fig. 4. Derived thermal diffusivity α of high-grade fused quartz: gold-coated evaluated with conventional (∇) and new ($\triangle$) model; only graphite coated evaluated with conventional ($\diamond$) and new ($\square$) model.

for a slight increase in the calculated thermal diffusivity from 670 to 770 K, even for the gold-coated sample. This might be due to the fact that the sample is not strictly diathermic (compare Fig. 2). However, it should be noted that the experimental error is ± 0.015 mm$^2 \cdot$ s^{-1} at 773 K.

5. CONCLUSIONS

A new analytical model to determine the thermal diffusivity of diathermic samples was presented. Measurements on two glass samples with coatings of low and high emissivity were evaluated with a conventional and the new analytical model. The results show that measurements on graphite-coated samples evaluated with the new analytical model lead to the correct thermal diffusivities. The coating of the sample with gold and graphite can thus be replaced by a simple graphite coating.

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