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Measurement of the Thermal Diffusivity of Liquids with a Laser-Induced Thermal-Grating Technique: An Experimental Investigation of Systemtic Errors

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The laser-induced thermal-grating technique has been developed to determine the thermal diffusivity of liquids and liquid mixtures. Here we report an experimental investigation of the systematic errors, which are caused by departures from one-dimensional heat conduction. In particular, the effects of cell wall and intersection angle of the heating beams have been thoroughly studied through measurements on toluene and methanol. A comparison has been made between experimental results and numerical predictions. The excellent agreement between experiments and theoretical predictions shows that the theoretical error analysis can be used for the estimation and accurate correction of systematic errors in measurements by this technique.

KEY WORDS: diffraction; laser-induced thermal grating; methanol; systematic error; thermal diffusivity; toluene.

1. INTRODUCTION

As an optical method, the laser-induced thermal-grating technique is suitable for the determination of the thermal diffusivity of liquids in a wide range of temperatures. This technique determines the thermal diffusivity of a sample by investigating the relaxation behavior of the diffracted intensity of the probing laser beam, and so it produces absolute values without calibration. In comparison with conventional measurement methods, this technique has the advantage that no sensors need to be inserted in the sample, especially when measurements on electrically conducting and

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corrosive liquids have to be made. Owing to the short measuring time (typically a few milliseconds), the small temperature rise in the sample during measurements (less than 0.1 K), and the very small sample volume (a few cubic millimeters), the effect of natural convection can be neglected. Moreover, application of this technique to anisotropic materials such as flowing polymer melts seems very promising.

In practice, the implementation of this measurement technique inevitably deviates from the ideal model. For example, systematic errors can be caused by deviations of experimental conditions from those of the ideal mathematical model such as departures from one-dimensional heat conduction. Nagasaka et al. [1] have analyzed the systematic errors inherent in thermal-diffusivity measurements via the thermal-grating technique using Green's function. A theoretical investigation of systematic errors has also been carried out by Wang et al. [2]. They have studied the dynamic behavior of the transient thermal grating during its excitation and relaxation by a numerical simulation. The systematic errors, which are caused by sample thickness, intersection angle, and absorption, etc., have been quantitatively analyzed with this simulation method. It was deemed that one can choose the experimental parameters such as the sample thickness d, the grating period Λ, and the absorption coefficient α to cancel the total error arising from these parameters to be almost-zero [1]. But, from experience, we have realized that it is difficult to find such combinations of experimental parameters due to difficulties in adjusting the optical arrangement. Another possibility is to try to estimate the systematic errors based on the error analysis and to correct the measured thermal diffusivity values for the errors after the measurements [2, 3]. Although we have thoroughly investigated the systematic errors with a simulation method, an experimental study of the systematic errors should be made to give the verification of the theoretical predictions. Here in particular, we try to study the effects of the cell wall and intersection angle of the heating beams in detail through measurements on toluene and methanol and to compare the experimental results with the theoretical predictions made by our numerical simulation.

2. MEASUREMENT TECHNIQUE

The theory and experimental apparatus are described in detail in Refs. 3–6. Here, a brief description of the measurement technique is presented.

To induce transient thermal grating, a short laser pulse ($0 < t \leqslant t_h$) is produced by chopping a laser beam ($\lambda_h = 488$ nm) mechanically. This heating laser beam is then split into two beams, which intersect at an angle θ in the thin sample and create a spatially periodic distribution of light

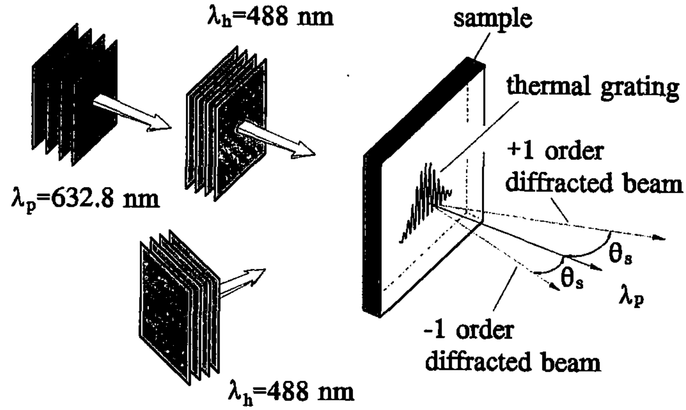

Fig. 1. Schematic of the laser-induced thermal grating technique.

intensity (Fig. 1). Along with the illumination of the liquid sample by the light pulse ($t > 0$), a grating-like temperature distribution (thermal grating) is gradually induced due to absorption of the laser intensity. A corresponding, periodic distribution of the refractive index n is then produced, which is called an optical grating. After the laser pulse ($t > t_h$), the periodic temperature distribution equalizes due to heat conduction in the sample. On the assumption of one-dimensional heat conduction in the modulation direction of temperature, the amplitude $\Delta T(t)$ of the thermal grating decays with a relaxation time

$$\tau = \frac{1}{a}\left(\frac{\Lambda}{2\pi}\right)^2 \tag{1}$$

where a is the thermal diffusivity of the liquid sample and Λ the grating period.

As shown in Fig. 1, a probing laser beam of wavelength λ_p ($\lambda_p = 632.8$ nm) penetrates the optical grating and is diffracted by it. The intensity of the first-order diffraction $I_1(t)$ is proportional to the square of $\Delta T(t)$ and expressed as

$$I_1(t) \propto \Delta T(t)^2 \propto \exp(-2t/\tau) \tag{2}$$

Therefore, the thermal diffusivity of the liquid sample can be determined by measuring the time dependence of the first-order diffracted intensity $I_1(t)$ and the grating period Λ

$$a = -\frac{1}{2}\left(\frac{\Lambda}{2\pi}\right)^2 \frac{d \ln I_1(t)}{dt} \tag{3}$$

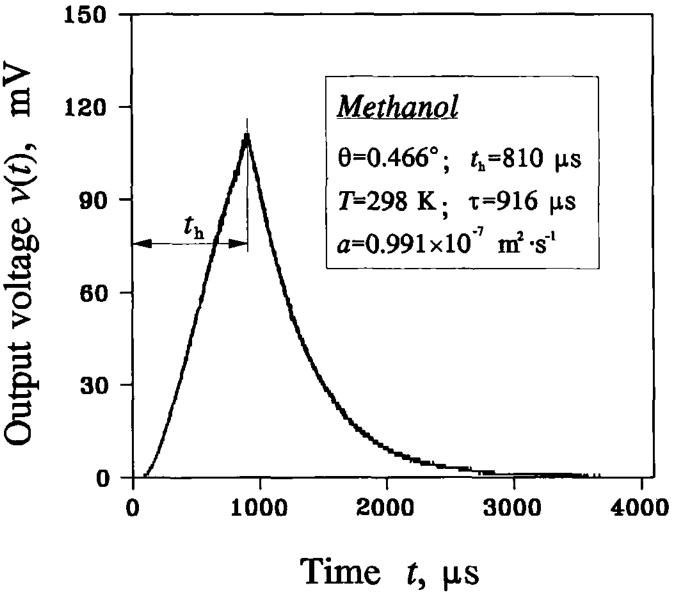

Fig. 2. Diffracted signal: a typical example of the output $V(t)$ from the photomultiplier.

In the measurements, the diffracted signal $I_1(t)$ is detected by means of a photomultiplier tube (PMT) in the homodyne scheme. The output $V(t)$ from the PMT is then amplified and recorded by a digital oscilloscope (Fig. 2). Subsequently, the $V(t)$ data are transferred to a personal computer, where they are fitted to

$$V(t) \propto I_1(t) \propto \Delta T(t)^2 \propto \exp(-2t/\tau) \tag{4}$$

by a least-squares calculation to obtain the relaxation time τ.

The grating period Λ is determined by measuring the diffraction angle θ_s instead of the intersection angle θ of the heating beams

$$\Lambda \approx \frac{\lambda_h}{\sin\theta} \approx \frac{\lambda_p}{\sin\theta_s}, \qquad \theta \ll 1 \tag{5}$$

In the present study, the diffraction angle θ_s is very small and varies from 0.20 to 1.5°. To determine θ_s accurately, we measure the intensity distributions of diffracted beams of +1 and −1 order with the photomultiplier tube (PMT) and an A/D converter. The distance ΔX between both peaks of the intensity distributions can then be determined. Hence, we can calculate the diffraction angle θ_s from ΔX and the distance L between the PMT and the crossing point of the heating beams in the sample [3, 6].

3. RESULTS AND DISCUSSION

There are seven experimental parameters which can cause deviations of the experimental conditions from those postulated by the ideal mathematical model and lead to systematic errors in the measurements [2]. These parameters are the sample thickness d, the intersection angle θ of the heating beams, the absorption coefficient α of the liquid sample, the Gaussian beam intensity distribution w and focusing f^* of the heating beams, the heating duration time t_h, and heating laser power P.

According to the theoretical error analysis [2], the error due to the heating duration time t_h is very small and negligible. This conclusion has also been experimentally confirmed by Nagasaka et al. [1] In our measurements, no influence of heating duration time t_h on the results for the thermal diffusivity a was observed, even though the measurements were done at $t_h = 1200\ \mu s$ ($d = 0.5$ mm and $\Lambda \approx 50\ \mu m$). Therefore, we conclude that any error of heating duration time t_h needs not be considered at the error correction. Similarly, the effect of heating laser power is also sufficiently small to be neglected.

The Gaussian beam intensity distribution w of the heating beams may cause heat flow to the unheated sample area in both x and y directions. A Gaussian temperature distribution, which results from the Gaussian beams, is superposed upon the sinusoidal grating structure and delays the relaxation of the thermal grating. As a result, the effect of Gaussian beam intensity distribution w of heating laser beams causes a reduction in the measured thermal-diffusivity value [1, 2, 7]. Due to the focusing of the probing and heating laser beams, which is necessary to intensify the diffracted signal and improve the signal-to-noise ratio significantly [3, 4], it is difficult to estimate the error arising from the Gaussian beam intensity distribution and to make correction to the measured thermal diffusivity results in actual practice. Hence, we try to choose the focusing parameter f^* and Λ in experiments to meet the condition $\Lambda/(f^* w) < 0.1$ [2], on which the effect of Gaussian beam intensity distribution w and focusing f^* is negligible.

To increase the absorption coefficient α, the liquid sample has to be doped with a dye. The amount of the dye to be added can usually be kept so small that the addition of the dye does not exert any influence on the thermophysical properties of the sample. According to the results of a numerical error analysis [2], the effect of the absorption coefficient α is negligible at $\Lambda/\alpha^{-1} < 0.05$. To meet this condition, we kept the grating period Λ approximately 50 μm and the absorption coefficient α smaller than 0.8 mm^{-1} in our measurements, which showed a sufficiently strong absorption for the measurements.

The results of the numerical simulation of the error due to the sample thickness d is shown as a function of Λ/d in Fig. 3 (see also Ref. 2), in which $a_{\text{Cal.}}$ stands for the calculated apparent thermal diffusivity, and $a_{\text{Ref.}}$ is the reference standard value. The calculations were carried out at $t_h = 1200$ μs, $\alpha = 0.8$ mm^{-1}, $\Lambda = 50$ μm, and $P_A = P_B = P/2 = 200$ mW, which approximately correspond to the generally selected values of experimental parameters in the measurements. The error $(a_{\text{Cal.}} - a_{\text{Ref.}})/a_{\text{Ref.}}$ due to the sample thickness increases with decreasing thermal diffusivity of the liquid sample. Nevertheless, the dependence of $(a_{\text{Cal.}} - a_{\text{Ref.}})/a_{\text{Ref.}}$ on the thermophysical properties is not obvious. With decreasing sample thickness d, the error $(a_{\text{Cal.}} - a_{\text{Ref.}})/a_{\text{Ref.}}$ increases because the heat loss through both sample windows becomes larger. At $\Lambda/d \approx 0.1$, which corresponds to the typical working condition, the error would be about 1.7%, and this error should be taken into account at the error corrections.

To verify the theoretical results, we have studied the effect of sample thickness d on the measured thermal diffusivity experimentally. The

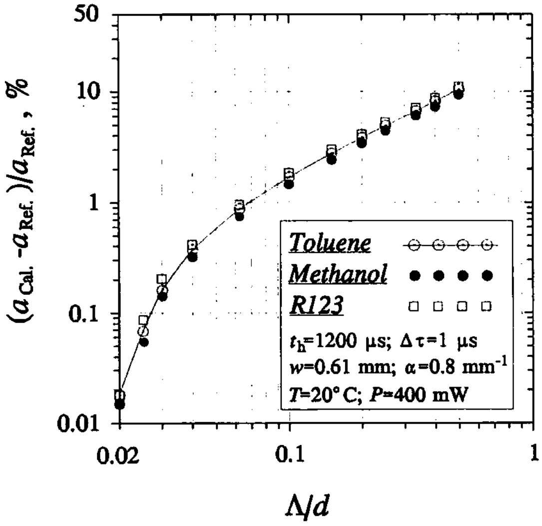

Fig. 3. Calculated results for the effect due to the sample thickness d as a function of Λ/d.

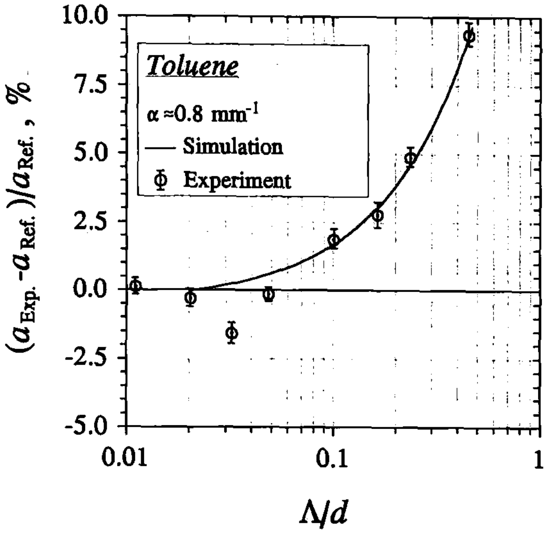

Fig. 4. Experimental results on toluene for the effect of sample thickness.

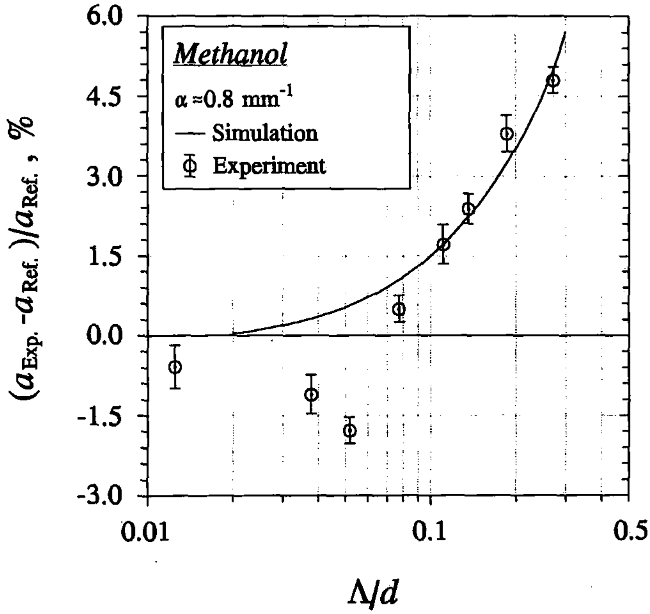

Fig. 5. Experimental results on methanol for the effect of sample thickness.

measurements were done at $t_h = 1200\ \mu s$, $\alpha \approx 0.8\ mm^{-1}$, $\Lambda \approx 50\ \mu m$, and $P \approx 400$ mW. The effects of the absorption coefficient α of the liquid sample, the Gaussian beam intensity distribution w and focusing f^* of the heating beams, the heating duration time t_h, and the heating laser power P can therefore be neglected. Figures 4 and 5 show experimental results of toluene and methanol, respectively, at room temperature as a function of Λ/d. The errors $(a_{Exp.} - a_{Ref.})/a_{Ref.}$ represent the deviations of the measured thermal-diffusivity values $a_{Exp.}$ from the reference standard values $a_{Ref.}$ of the thermal diffusivity of toluene and methanol. The values for $a_{Ref.}$ of toluene were calculated from the thermal conductivity λ [8], the specific heat capacity at constant pressure c_p [9], and the density ρ [9] using the relationship $a = \lambda/\rho c_p$. The values for $a_{Ref.}$ of methanol were taken from Ref. 10. In the measurements, the sample thickness d was changed from 0.1 to 2 mm, whereas the grating period Λ was kept nearly constant ($\Lambda \approx 50\ \mu m$ for toluene and $\Lambda \approx 55\ \mu m$ for methanol). From Figs. 4 and 5, we can see that the experimental results agree fairly well with the numerical results at $\Lambda/d \geqslant 0.1$. However, there are significant discrepancies between the measurement results and the values predicted by the numerical simulation, when Λ/d is smaller than 0.1. For an explanation we should know

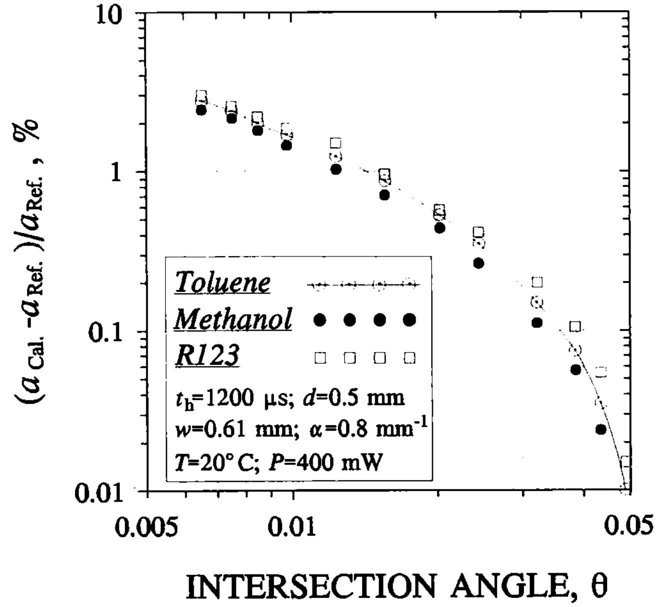

Fig. 6. Calculated results for the effect due to the intersection angle θ.

whether the thin grating assumption is always valid in the measurements. According to Eichler et al. [11], we obtain as the thin grating condition

$$Q=\frac{2\pi\, d\lambda_{\mathrm{p}}}{\Lambda^{2}n}\ll 1 \tag{6}$$

where n is the refractive index of the liquid sample. At $\Lambda/d=0.1$, the Q parameter has a value of 0.5 and increases with increasing larger sample thickness d. At $d=2$ mm, the Q parameter exceeds the value 2. The grating is then optically too "thick," and as a result, the relationship between the first-order diffracted intensity $I_1(t)$ and the amplitude $\Delta T(t)$ of the thermal grating, i.e., Eq. (2), is no longer valid, as stated in Ref. 1. This is the reason the measured values deviate significantly from the numerical results at a Q value greater than 0.5. Based on the measurement results, we conclude that the application of the thin grating assumption is no longer reasonable, when the Q parameter exceeds 0.5. It is therefore suggested that measurements should be done at $Q\leqslant 0.5$, and the effect due to the heat loss

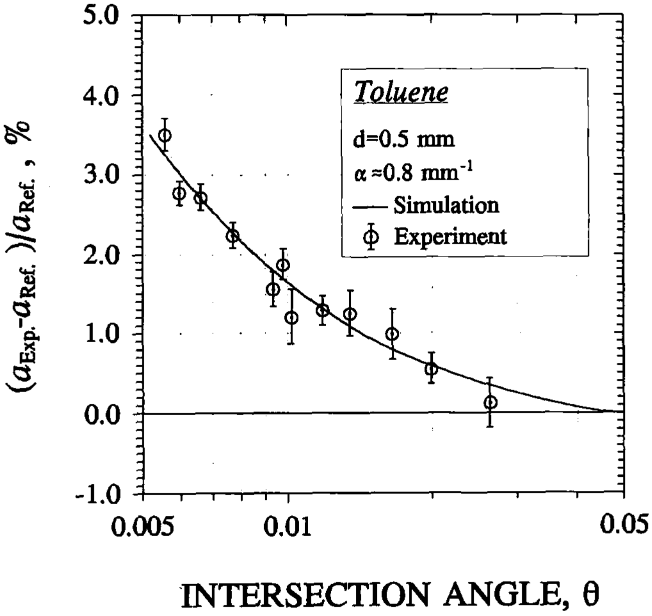

Fig. 7. Experimental results on toluene for the effect of intersection angle θ.

to the sample windows can then be taken into consideration at the error correction.

Usually, the sample thickness d is given (typically 0.5 mm), and the intersection angle θ can be adjusted to a certain value in experiments. It is, therefore, meaningful to study the effect due to the intersection angle of two heating beams at the given sample thickness $d = 0.5$ mm. Figure 6 shows the simulation results for the effect of the intersection angle θ at $d = 0.5$ mm, $t_h = 1200\,\mu$s, $\alpha = 0.8\ \text{mm}^{-1}$, $w = 0.61$, and $P_A = P_B = P/2 =$ 200 mW. Based on Fig. 6, we can perform the measurements at the given sample thickness d and a known intersection angle θ, and correct afterward the measured thermal-diffusivity values for the systematic error due to the sample thickness at the known intersection angle conveniently.

Figures 7 and 8 represent the experimental results for the effect of the intersection angle θ at $d = 0.5$ mm, $t_h = 1200\ \mu$s, $\alpha \approx 0.8\ \text{mm}^{-1}$, $w = 0.61$, and $P/2 = 200$ mW. The measurements were performed at room temperature, and toluene and methanol were used as liquid samples. As shown in Figs. 7 and 8, the measured values are in good agreement with

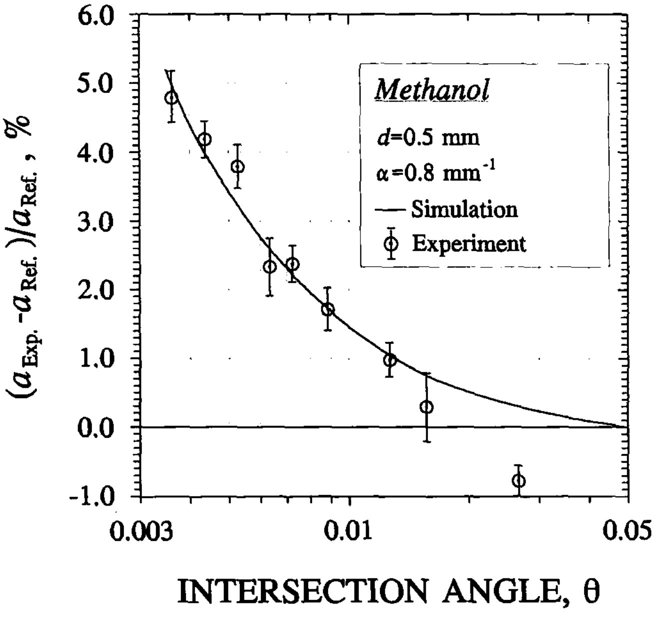

Fig. 8. Experimental results on methanol for the effect of intersection angle θ.

the numerical results, especially at $\theta \leqslant 0.01$. For larger intersection angles, the grating becomes "thick," which leads to large Q values and deviations of the measured results from the numerical ones. Measurements should therefore be made under the condition $\theta \leqslant 0.01$ at $d = 0.5$ mm, so that the thin grating condition $Q < 0.5$ is satisfied.

4. CONCLUSIONS

An experimental investigation of systematic errors has been performed, which are caused by departures from one-dimensional heat conduction. In particular, the effects of sample thickness and intersection angle of heating beams have been thoroughly studied through measurements on toluene and methanol. The experimental results were compared with the theoretical predictions made by a numerical simulation. The excellent agreement between experiments and numerical predictions shows that the theoretical error analysis can reliably used for the estimation and correction of systematic errors in the measurements by this technique.

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