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Subsecond Measuring Technique for In-plane Thermal Diffusivity at Local Area by the Forced Rayleigh Scattering Method[1]

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In the course of a chemical reaction or structural changes in a material, the thermal diffusivity of the material changes in response to its varying conditions. Real-time information in the form of successive data on the thermal diffusivity during the process is indispensable for an adaptive control system for material production or processing. In the present study, a measurement technique for in-plane thermal diffusivity in micro-scale and at a high repetition rate has been studied. In view of the application of this method for fast processes, the effects of property changes during a single measurement time on the measured value have been quantitatively evaluated. We have developed a measurement system of the in-plane thermal diffusivity by means of the forced Rayleigh scattering method, an optical technique with a measurement time less than 1 ms and at a micro-scale local observation area. In the present system, each thermal diffusivity measurement was conducted every 10 ms; i.e., the repetition rate was up to 100 data per second. The results of preliminary measurements of a polymethylmethacrylate plate with several repetition rates verify the reliability of this technique in tracing high-speed phenomena.

KEY WORDS: dynamic monitoring system; forced Rayleigh scattering method; real-time measurement; thermal diffusivity.

1. INTRODUCTION

In many fields such as semiconductor manufacturing, polymer engineering, and food processing, thermal control plays an important role and even has a

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critical influence on the quality of the final product. For example, during an internal structural change or a chemical reaction process in a material, the thermal diffusivity is expected to vary with the progress of the process [1]. Therefore, dynamic feedback utilizing real-time data of the thermal diffusivity can be a means which makes possible a more flexible process control than when using only temperature or pressure measurement. Although various methods to measure the thermal diffusivity of solids and liquids have been studied [2, 3], many techniques are not suitable for application to real-time monitoring of a material under a changing process.

The forced Rayleigh scattering method (FRSM) is a technique for measuring the thermal diffusivity that has the capability to carry out high-speed and real-time sensing. The following characteristics of the FRSM are valuable for measuring the thermal diffusivity in real-time: (a) a non-destructive method, which can be applied for *in situ* monitoring of a changing process; (b) time-resolved measurement with a high repetition rate; and (c) micro-scale sensing. The aim of this study is to develop the FRSM as a "real-time thermal-diffusivity monitoring system" that can be applied to tracing a rapid process in micro-scale. We have previously reported a basic experimental system utilizing the FRSM and the preliminary results of continual measurements of a biopolymer in a transition state [4]. In the present paper, a theoretical consideration of the error analysis for property variations during a single measurement process is discussed. Moreover, an experimental setup that can be used for continual measurements with a temporal resolution of 10 ms has been developed, and validation of the system based on measurements of a polymethylmethacrylate plate with various repetition rates is also described.

2. PRINCIPLE OF MEASUREMENT

The FRSM can measure the thermal diffusivity of a sample in a non-contact manner, and the principle is based on two phenomena, a transient thermal grating and diffraction [5, 6]. Fig. 1 shows a schematic image of the principle. A sample is heated by an interference pattern created by two short-pulsed laser beams of equal wavelength and intensity. A temperature rise corresponding to the interference pattern of heating beams is generated. After the heating, the excited temperature distribution decays exponentially by heat conduction. The time profile of the temperature distribution in the sample can be described as

$$T(x,t) = T_0 + T_1 \exp\left[-a\left(\frac{2\pi}{\Lambda}\right)^2 t\right]\cos\left(\frac{2\pi x}{\Lambda}\right), \tag{1}$$

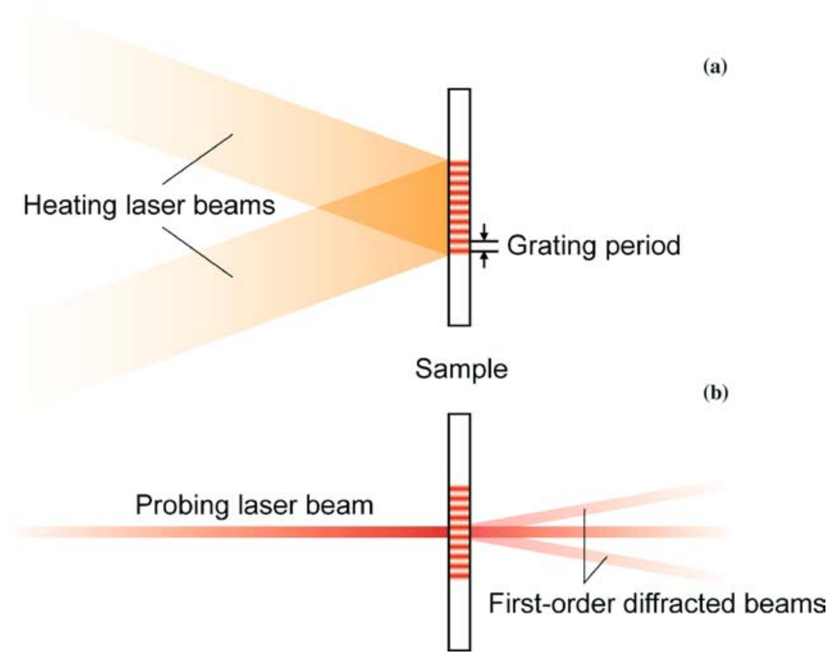

Fig. 1. Schematic image of the principle of the forced Rayleigh scattering method (FRSM). This is based on (a) a transient thermal grating and (b) diffraction.

where T_0 is the mean temperature rise and T_1 is a coefficient representing the temperature distribution due to the thermal fringes of laser heating, which are independent of time t and position x. Λ is the grating period of the interference pattern, and a is the thermal diffusivity of a sample. Under the existence of the temperature distribution, the refractive index gradient caused by the temperature variation acts as a transient optical grating. A probing laser beam with a non-absorbing wavelength passes through the heated region and is diffracted by the grating. The intensity of the first-order diffracted beam is proportional to the square of the temperature change, based on diffraction theory [7]. Therefore, the intensity of the first-order diffracted beam I_1 after the laser heating can be expressed as a function of time t as

$$I_1 \propto \exp\left(-\frac{2t}{\tau}\right), \tag{2}$$

where τ is the relaxation time of heat conduction. Consequently, the thermal diffusivity of the sample can be determined by analysis of the temporal decay of the diffracted beam, and the equation takes the form,

$$a = \frac{1}{\tau}\left(\frac{\Lambda}{2\pi}\right)^2. \tag{3}$$

3. EVALUATION OF THE UNCERTAINTY FACTOR DURING A SINGLE MEASUREMENT

In the basic theory of the FRSM, the thermophysical properties of a sample are assumed to be uniform on the time scale of a single measurement, like almost all measuring techniques for the thermal diffusivity. However, with a sample undergoing rapid and drastic change, the property variation during a single measurement cannot be negligible. Considering the application of this method to real-time monitoring of a changing material, it is necessary to evaluate the uncertainty factor due to property changes during the measurement period. In the present study, we have constructed a theoretical model to estimate the influence quantity, which is the magnitude of uncertainty due to the property change in a single measurement process of the FRSM. Here, we assume that the thermal-diffusivity changes of a sample can be described as a function of time, as in the following equation:

$$a = a_0\left[1 + \zeta\,(t)\right], \tag{4}$$

where a_0 is the initial thermal diffusivity of the sample at the time when the measurement is started and t is the elapsed time after heating. Using Eq. (4), the heat conduction problem under the same initial and boundary conditions for the basic theory of the FRSM is considered. The time-dependent temperature distribution at position x and at time t can then be expressed as

$$T\,(x,t) = T_0 + T_2 \exp\left\{-a_0\left(\frac{2\pi}{\Lambda}\right)^2\left[t + \int \zeta\,(t)\,dt\right]\right\}\cos\left(\frac{2\pi x}{\Lambda}\right), \tag{5}$$

where T_2 is independent of position and time. As can be seen from the comparison between Eqs. (1) and (5), the effects of property change appear only in the term containing the temporal variation. Fig. 2 provides an image of the temperature evolution described in Eq. (5). For the case when the thermal diffusivity increases (case I), the temperature decay is quickened, and the thermal diffusivity decrease impedes the attenuation (case II). The matter of concern is the magnitude of the effect on the actually measured value of the thermal diffusivity. Thus, we consider the case where the obtained signal including the effect of property variation

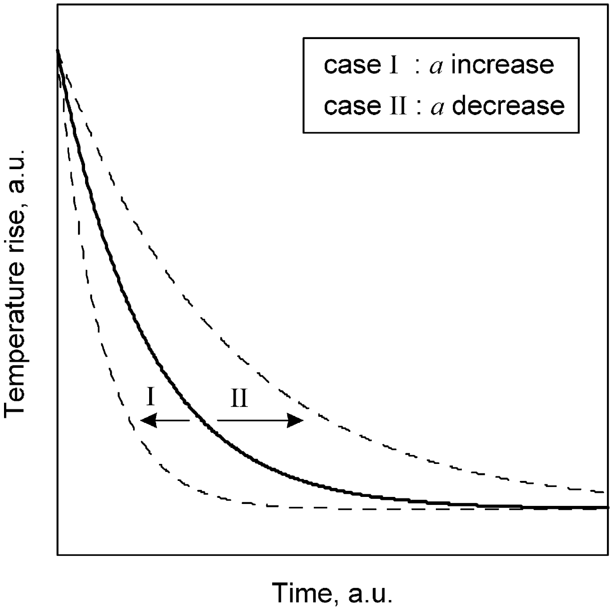

Fig. 2. Attenuation process of the temperature rise by laser heating when the property of the sample is varying. Cases I and II indicate the time profile of sample temperature with increasing and decreasing thermal diffusivity, respectively.

is analyzed based on the scheme with the constant property assumption. The influence quantity can be estimated from the comparison between the relaxation time of the temperature with and without the property variation. Here, the thermal diffusivity of the sample is assumed to vary linearly with time;

$$\zeta(t) = \psi t, \tag{6}$$

where ψ is the rate of change of the thermal diffusivity. Although this assumption is simple, it is a proper approach for a measuring technique with a short measurement period. The relaxation time of heat conduction τ_{meas} from the temperature distribution described in Eq. (5) can be written as

$$\tau_{\text{meas}} = \frac{-1 + \sqrt{1 + \frac{2\psi}{a_0}\left(\frac{\Lambda}{2\pi}\right)^2}}{\psi}. \tag{7}$$

By substituting Eq. (7) into Eq. (3), the measured thermal diffusivity a_{meas} can be calculated. Then, a deviation of a_{meas} to a_0 gives us the quantity of the uncertainty factor. The interrelation between this deviation and the amount of property variation during the same time scale clearly indicates the influence. Fig. 3 shows the results of this analysis. In the case of an increasing property, the uncertainty is positive, and vice versa. In these calculations, a_{meas} is the mean value of the thermal diffusivity at the starting and completion points in a single measurement. This analytical result is due to the linear assumption of the temporal evolution of the property described in Eq. (6). The actual time required for the present FRSM setup to carry out a measurement is on the order of several hundred microseconds, for which many rapid processes can meet this linear premise. Based on the principle of the FRSM, any other properties whose variations can be converted to a change in the thermal diffusivity have an influence on temperature decay. This is why only the thermal diffusivity variation during the relaxation process of the temperature rise caused by laser heating is treated here. From this analytical model, we can quantitatively evaluate the effects of the property change on the measured value, and these results illustrate the possibility of applying the FRSM to real-time monitoring of high-speed changing processes.

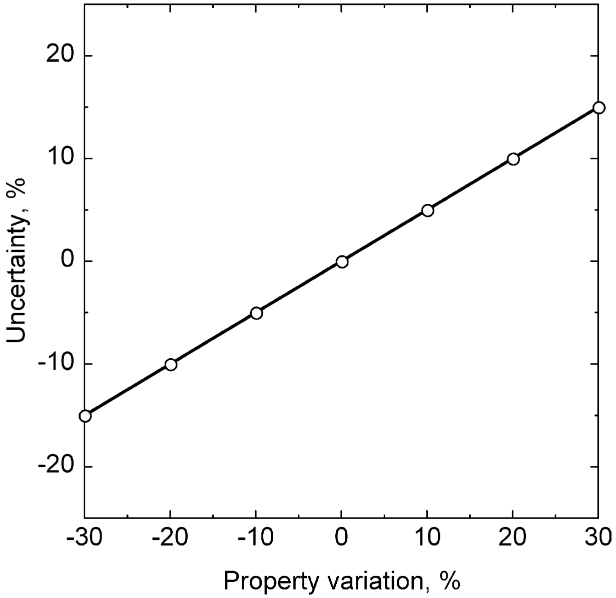

Fig. 3. Analytical results of the uncertainty factor due to property variations during a single measurement time period on the measured value.

4. EXPERIMENTAL

A schematic diagram of the experimental setup is shown in Fig. 4. A continuous-wave CO_2 laser with a wavelength of 10.6 μm serves as a heating source. The beam is converted into a short pulse by a mechanical chopper (MC), and is then divided into two beams of equal intensity by a beam splitter (BS). Part of the heating pulsed beam is sampled by a partial reflection mirror (PM, 1%) and is detected by using a LN_2-cooled HgCdTe detector (MCT) for a timing trigger for data acquisition. A He–Ne laser with a wavelength of 632.8 nm is utilized as the probing laser. The time-dependent diffracted beam is detected by a photomultiplier tube (PMT) through pinholes (PH1 and 2) and an interference filter (IF). The signal from the PMT is sent to a computer via an 8-bit analog-to-digital converter so that the data can be analyzed. In this study, the grating period Λ is determined by measuring the first-order diffraction angle θ_1,

$$\Lambda = \frac{\lambda_p}{\sin\theta_1}, \tag{8}$$

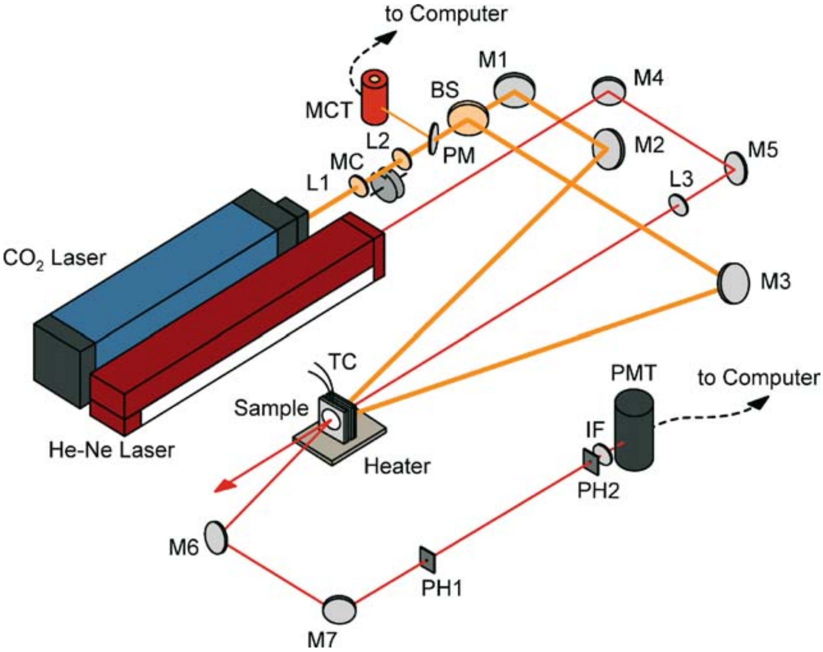

Fig. 4. Schematic of the present experimental setup. A signal from MCT is used as a timing trigger for data acquisition of the diffracted beam detected by PMT.

where λ_p is the wavelength of the probing laser. The measurement area can be changed by adjusting the lens (L3), which in the present study was fixed to an approximate diameter of 650μm. In the case of measuring a solid, the sample is directly measured. And in the case of treating a liquid, we utilize a sample cell constituted with anti-reflection coated ZnSe windows, an aluminum housing, and a fluorocarbon rubber spacer that defines the sample thickness. The sample temperature is controlled by an electric plate heater attached to the cell and is monitored by a K-type thermocouple (TC). This experimental system can perform a series of thermal-diffusivity measurements at intervals of 10 ms; in other words, we can measure the thermal diffusivity at a rate of 100 data per second. This rate has primarily been achieved by a sampling scheme of heating pulse timing and by employing a high-speed analog-to-digital converter.

The grating period selected in this study was around 30–100 μm, meaning that the decay time of heat conduction of the temperature rise from the interference pattern of heating beams was less than 1 ms. After a lapse in the relaxation time, a temperature rise remains in the sample, which is due to the Gaussian intensity distribution of the heating beam. Therefore, there is a need for some time interval so that the macroscopic temperature rise will adequately decay. The period necessary for this temperature decrease to occur can be determined by calculating the three-dimensional heat conduction problem, and this means the time resolution of continual measurements for the thermal diffusivity by the FRSM. Consequently, the time is determined by the thermal diffusivity of a sample, the absorption coefficient of the sample at the wavelength of the heating laser, and the Gaussian radius of the heating beam [4]. From this analysis, we can determine the theoretical limit of the time resolution of continual measurements, making it possible to carry out continual measurements at proper time intervals.

5. EXPERIMENTAL RESULTS

As a verification of the present experimental setup, we carried out continual measurements for the thermal diffusivity at various repetition intervals. The sample is a polymethylmethacrylate plate which has a thickness of 2 mm. The repetition rates were 10, 50, 100, 200, 500, and 1000 ms, and all of these experiments were conducted at room temperature (approximately 298 K) and under atmospheric pressure. Fig. 5 shows a typical example of the signal data taken from the measurements and the fitting curve. The signal-to-noise ratio was high enough to perform a nonlinear fit with sufficient convergence. Measurement results of the thermal

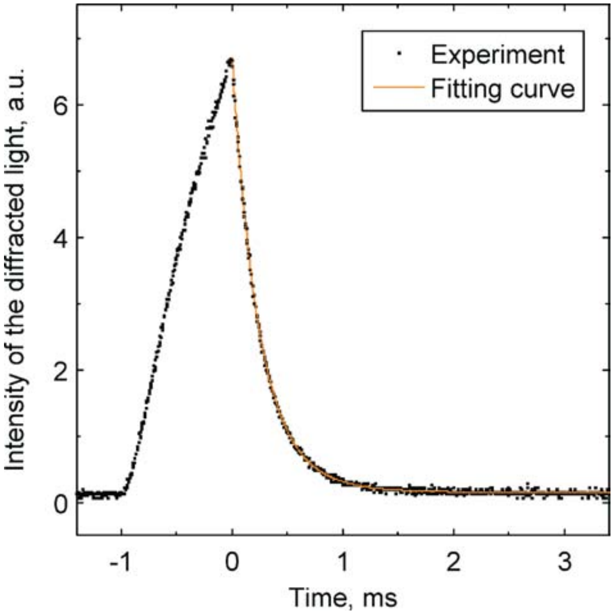

Fig. 5. Typical signal and fitting curve from real-time measurements of a polymethylmethacrylate plate.

diffusivity are illustrated in Fig. 6. The grating period of the laser heating was set to 40.1 μm. Except in the case of a repetition rate of 10 ms, the thermal-diffusivity data have good reproducibility. Only at a rate of 10 ms, a gradual decrease on the measured values was observed. Due to the temperature rise caused by the Gaussian intensity distribution of the heating laser, the time required to adequately decay in the temperature change of a polymethylmethacrylate plate is calculated to be approximately 50 ms, using reported values of the thermal diffusivity [8] and absorption coefficient [9]. In the case of continual measurements with the highest repetition frequency, the excited temperature rise still remains in the sample plate when the next measurement starts. A gradual decrease in the measured value is then attributed to the negative temperature dependence of the thermal diffusivity in the sample. From these measurement results, the validity of the calculation model regarding the time resolution of continual measurements is demonstrated, and the possibility of the experimental system to perform real-time thermal-diffusivity measurements with good stability is indicated.

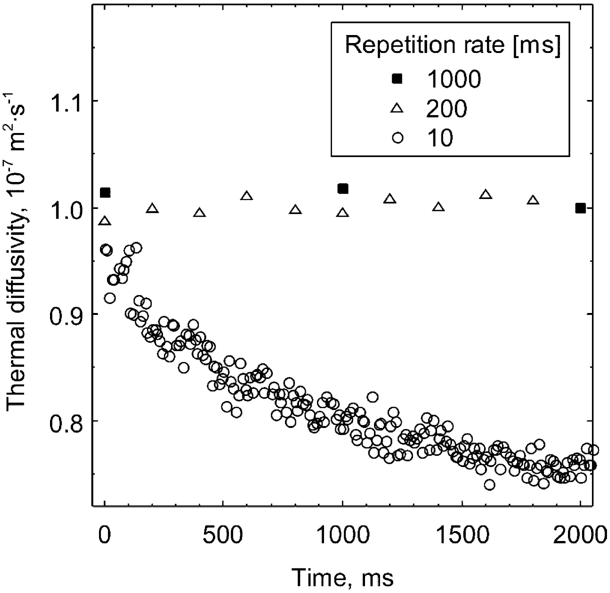

Fig. 6. Experimental results of continual measurements of the thermal diffusivity with various repetition rates. The range of the measurement time interval is from 10 to 1000 ms.

6. CONCLUSIONS

A measurement system for in-plane thermal diffusivity in real-time and at a high repetition rate up to 100 data per second by the forced Rayleigh scattering method (FRSM) has been developed. In order to evaluate the influence quantity of property variations during a single measurement time on the measured value, a theoretical model to describe the uncertainty factor was established. According to experimental results from continual measurements of a polymethylmethacrylate plate with various repetition rates, the validity of the FRSM to be a reliable monitoring technique for the thermal diffusivity that can be applied to a rapid process or phenomenon was demonstrated.

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REFERENCES

1. J. O. M. Flores, J. M. Y. Limón, F. J. E. Beltran, and J. G. Hernandez, *Rev. Sci. Instrum.* **74**:814 (2003).
2. W. A. Wakeham, A. Nagashima, and J. V. Sengers, eds. *Measurement of the Transport Properties of Fluids*, *Experimental Thermodynamics*, Vol. 3 (Blackwell Scientific Pubs., London, 1991), pp. 111–192.
3. D. P. Almond and P. M. Patel, *Photothermal Science and Techniques*, *Physics and its Applications 10* (Chapman & Hall, London, 1996), pp. 199–214.
4. M. Motosuke, Y. Nagasaka, and A. Nagashima, *Int. J. Thermophys.* **25**:519 (2004).
5. H. J. Eichler, P. Gunter, and D. W. Pohl, *Laser-Induced Dynamic Gratings*, *Springer Series in Optical Sciences*, Vol. 50 (Springer-Verlag, Berlin, 1986), pp. 84–89.
6. Y. Nagasaka, T. Hatakeyama, M. Okuda, and A. Nagashima, *Rev. Sci. Instrum.* **59**:1156 (1988).
7. W. R. Klein, B. D. Cook, and W. G. Mayer, *Acustica* **15**:67 (1967).
8. B. M. Suleiman and V. Bohac, *Macromol. Symp.* **170**:329 (2001).
9. R. T. Graf, J. L. Koening, and H. Ishida, *Appl. Spectrosc.* **39**:405 (1985).