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Recent Development and Applications of an Optical Method for Measurements of Thermophysical Properties[1]

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The experimental determination of thermophysical properties has been greatly improved by the introduction of laser technology. The laser beam is used for sensing and also for heating (or exciting) the specimen. The advantage of using a laser beam is most strongly felt in the measurement of the thermal conductivity or the thermal diffusivity, which are some of the most difficult properties to measure. Interesting features of new techniques for investigating various aspects of thermal conductivity in fluids and solids are reviewed. An optical method, the so-called forced Rayleigh scattering method, or the laser-induced optical-grating method, has been developed and used extensively by the present author's group. The method is a high-speed remote-sensing method which can also quantitatively detect anisotropy, namely, direction dependence of heat conduction in the material. It was used for determination of the thermal diffusivity and its anisotropic behavior for high-temperature materials such as molten salts, liquid crystals, extended polymer samples, and flowing polymer melts under shear. Interesting applications of the method were demonstrated also for thermal diffusivity "mapping" and microscale measurement.

KEY WORDS: flow anisotropy; forced Rayleigh scattering; molten salts; optical method; polymers; shear anisotropy; thermal conductivity; thermal diffusivity.

1. INTRODUCTION

Developments and applications of new materials are an essential part of new technologies, and information on the thermophysical properties of

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these new materials is a prerequisite. Even conventional materials are expected to be used under such extreme conditions as high or low temperatures, in strong magnetic fields, under microgravity conditions, under high pressures, and so on. They may have such various forms as stratified, anisotropic, microscale, ultrathin, mixed conditions, etc.

For these new materials, under extreme conditions or in different forms, measurements often have to be done in contact-free condition, in a very short time under transient conditions, or with a very weak signal. Therefore, the conventional techniques to measure thermophysical properties are often not applicable or not even available.

Therefore, development of a new measurement technique is extremely important and urgent. The present author's group at Keio University has developed a method which optically detects the decay of a laser-induced periodic temperature distribution in the sample, liquid or solid. Details are explained below. In recent years, Sengers and co-workers at Maryland University, Leipertz and co-workers at Friedlich-Alexander-Universitaet Erlangen-Nuernberg, and Nagasaka and co-workers at Keio University recently have made interesting and valuable contributions in the application of optical methods to fluids under unusual conditions [1–6]. Because of limited space, the details of these studies are not given here.

Among various properties, thermal conductivity or thermal diffusivity is among the most difficult to determine accurately. The main problems lie in the determination of heat flux, measurement of local temperature in the sample, prevention of thermal convection, and correction for radiation transfer. The advantages of the optical method are demonstrated most clearly for thermal conductivity and thermal diffusivity. The present paper intends to explain recent developments of optical methods by taking, as an example, the so-called forced Rayleigh scattering method by the present author's group.

2. ADVANTAGES OF AN OPTICAL METHOD

Measurement of thermal conductivity or thermal diffusivity requires the creation of a temperature distribution in a sample, and accurate detection of the temperature. From the temperature distribution we can determine the temperature gradient. From the change of temperature with time we can determine the heat flux. For steady state, we need to determine the heat flux from another source, for example, the electrical power supplied. Optical means can be used for both heating and sensing.

Advantages of various recent optical methods, such as the forced Rayleigh scattering method, are summarized as follows:

(1) Remote heating–remote sensing: This is a particularly important advantage for the measurement of such corrosive high-temperature fluids as molten salts.

(2) Local measurement: By means of a laser beam, measurement of local properties is possible. This feature is advantageous for determining the thermal diffusivity of a small heated spot on a sample. By scanning the beam, the two-dimensional distribution, namely a "thermal diffusivity map," can be determined. Furthermore, it is possible to measure a microscopic sample by focusing the laser beam.

(3) High-speed measurement: Since one run requires only 1 ms or less, measurement or checking of transition states is possible. Also, it is not necessary to stabilize the system.

(4) High sensitivity and small temperature rise: The local temperature increase can be minimal. The temperature rise at the measured spot is less than 0.1 K. This prevents radiation loss or permits measurement near critical conditions.

(5) Anisotropy measurement: Since the heating of the sample is performed by interference of two beams, heated stripes give rise to heat conduction in one direction. So it is possible to measure the thermal diffusivity in one direction selectively.

(6) Absolute reliability check: The principle is absolute and quite different from other conventional methods. No calibration is needed. The method can be used to check the absolute reliability of other methods.

3. PRINCIPLE AND APPARATUS FOR FORCED RAYLEIGH SCATTERING

3.1. Principle

Details of the principle and error analysis of ordinary forced Rayleigh scattering (FRS) are described in Ref. 7. Only an outline is presented here. The principle of the method is shown in Fig. 1. A heating laser beam is divided into two beams of a splitter. If the beams are focused on the sample surface, an interference fringe pattern is formed. When the beams are absorbed a sinusoidal temperature distribution is generated in the sample. It induces a distribution of the refractive index which acts as an optical grating for another probing laser beam with a different wavelength. If the probing laser beam is aimed at the heated area of the specimen, a diffracted beam whose intensity is proportional to the square of refractive index

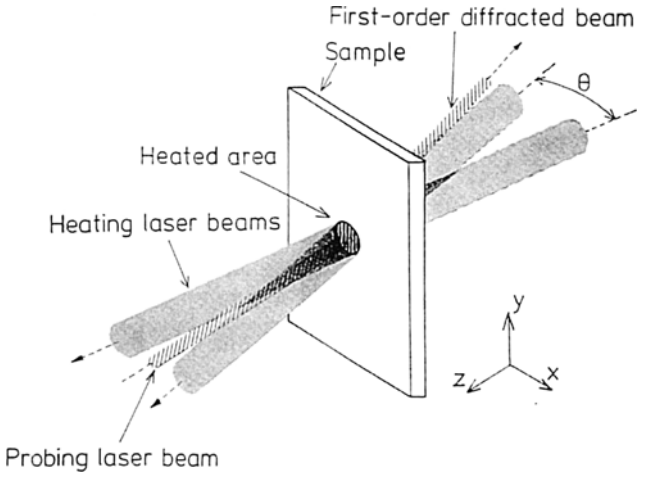

Fig. 1. Principle of the forced Rayleigh scattering method.

amplitude can be observed. If heating is terminated in a short time, an exponential decay of the excited temperature distribution takes place due to heat conduction in the specimen. Since the distribution of the refractive index also decays, the intensity I_1 of the diffracted beam relaxes with time t as shown in Eq. (1),

$$I_1 \propto \exp\left(-\frac{2t}{\tau}\right) \tag{1}$$

where τ is the time constant of the decay. That means we can measure the thermal diffusivity of the material by measuring the decay rate of the intensity of the diffracted beam. The working equation is derived by means of the theories of geometrical optics and of heat conduction [7]. From the time constant τ of relaxation, the thermal diffusivity a of the sample is calculated as

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

The grating period Λ in Eq. (2) is acquired from the wavelength of the heating laser λ_1, and the crossing angle θ between two heating beams as follows:

$$\Lambda = \frac{\lambda_1}{2 \sin(\theta/2)} \tag{3}$$

This is the principle of the forced Rayleigh scattering method. There are many factors such as three-dimensional conduction which may be affecting the accuracy of the measured thermal diffusivity. These factors have been extensively studied by Nagasaka and co-workers [7].

3.2. An Example of the FRS Experimental Apparatus

Each particular objective requires a different experimental setup. A single-mode argon-ion laser ($\lambda h = 514.5$ nm, 1.5 W) is employed as the heating laser. The continuous light of the heating laser is chopped by a rotating mechanical chopper into short pulses with which the heating pulse duration time can be changed from 40 to 1200 μs. The heating beams, divided into two beams of equal intensity by means of a beam splitter, cross in the sample to produce an interference pattern (the radius of the heating beam is approximately 1 mm at the sample). Typically, the range of fringe spacing of the gratings can be adjusted from about 35 to 90 μm, which corresponds to a range of beam crossing angles of about 0.84–0.33°. A He–Ne laser ($\lambda_p = 632.8$ nm, 5 mW), whose beam noise is sufficiently small to secure maximum reliability to determine τ, is used for probing the relaxation of the temperature distribution. The first-order diffracted light is detected in the homodyne scheme by a photomultiplier through a pinhole of 500-μm diameter and an interference filter. The output signal is recorded by a digital memory (12 bits, 5-μs sampling, and 1 k words) and is transferred to a microcomputer. The entire apparatus is arranged on an optical bench.

A sample cell is made of Pyrex glass. The thickness of the samples is usually adjusted from 0.8 to 1.2 mm by employing different thicknesses, of glass spacers which are glued onto glass cell walls with the aid of a SiO_2 cement prepared by a metal alkoxide hydrolysis process at about 150° C.

3.3. Experimental Verification of the FRS Method

As a test of the method, the thermal diffusivities of toluene and methanol have been measured near room temperature at atmospheric pressure. Both fluids had a stated purity of not less than 99%. The dye material employed was iodine for both toluene and methanol. The thermal conductivity of toluene derived from the measured thermal diffusivity and from literature data of the specific-heat capacity and the density was compared with the recommended correlation, which has an estimated accuracy of $\pm 0.6\%$. The sample results scatter within $\pm 3\%$. Although the precision of the FRS method is not better than that of the best conventional method such as the transient hot-wire method at the present stage, the former can be used for a variety of materials for which the latter cannot be applied.

4. APPLICATIONS OF FORCED RAYLEIGH SCATTERING

4.1. Absolute Reliability Check of Conventional Methods

In the previous section, the measured value of the thermal diffusivity was compared with existing data. However, the FRS method has been developed as an absolute method and can be used to check the reliability of many conventional methods of the thermal conductivity and the thermal diffusivity. Thermal conductivity is one of the most difficult properties to measure. The reasons for this difficulty are the heat losses due mainly to convection and radiation. Quantitative estimation of these losses is almost impossible. The best way to confirm the reliability of measured data of thermal diffusivity or thermal conductivity is to compare measured values obtained by different methods which are based on completely different principles and are capable of absolute measurements.

In this sense, the modern optical technique such as forced Rayleigh scattering is one of the most suitable ways to check the reliability of values measured by conventional methods such as the transient hot-wire method, the concentric-cylinder method, or the parallel-plate method.

4.2. Measurement of High-Temperature Melts

High-temperature fluids such as molten metals and molten semiconductor materials have really been fascinating targets of ambitious experimentalists, since these substances show scientifically interesting behavior, are important in industrial applications, and are often very difficult to measure. Sometimes no measuring method exists. Application of conventional methods which are well established at moderate temperatures requires complicated precautions at high temperatures. Indispensable requisites for reliable experimental measurement of transport properties are correct working equations, properly controlled fluid conditions, and experimental skill. The best method to measure the thermal diffusivity of high-temperature fluids is undoubtedly a contact-free or remote-sensing method. An optical method, that of forced Rayleigh scattering, has been developed and applied for many kinds of molten salts by the present author's group. Although the principle is not new, the method has been substantially improved by us and the new application to high-temperature melts has become possible. Experimental details and results were reported in a number of publications by Nagasaka and co-workers [8–16].

One typical example is shown here. It should be noted particularly, that the previous thermal-conductivity values of molten KCl scatter from about 0.4 to 1.2 $W \cdot m^{-1} \cdot K^{-1}$ as shown in Fig. 2, and their temperature coefficients are large and positive. In contrast, the present results are the

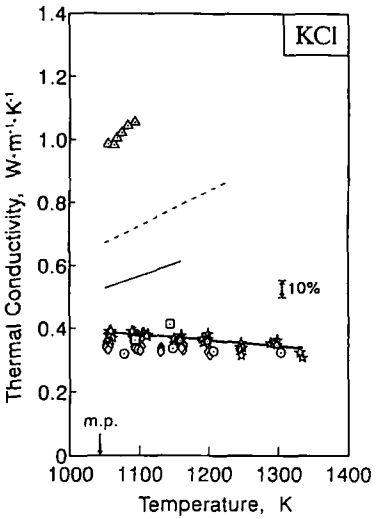

Fig. 2. Thermal conductivity of molten KCl. (---), Ref. 19; (⊡), Ref. 17; (△), Ref. 20; (——), Ref. 18; (⊙), Ref. 13; (◇), Harada; (☆) present author's group.

lowest of all and agree well with the data of McDonald and Davis [17] obtained with the transient hot-wire method using a platinum wire sheathed with a quartz tube. Moreover, the temperature dependence of the thermal conductivity is almost zero or weak negative. The data of Smirnov et al. [18], Fedrov and Machuev [19], and Polyakov and Gildebrandt [20] were all obtained by the steady-state concentric-cylinder method; it is thus considered that all these data are influenced strongly by convection, radiation, or other heat losses, all of which increase the apparent thermal conductivity. In the measurement of a high-temperature melt by FRS method, the overall uncertainty is estimated to be $\pm 7\%$.

It is concluded that the forced Rayleigh scattering method can be applied to high-temperature molten salts with a moderate accuracy which has never been accomplished by any other conventional methods.

4.3. Thermal Anisotropy Due to Deformation in Solid Polymers [21, 22]

In polymeric substances, anisotropy of various physical properties can be created by molecular orientation. Molecular orientation in polymeric materials is caused by various external forces, such as (a) deformation of the sample, (b) application of shear force in molten condition, and (c) stratification by other forces. An amorphous polymer is composed of

a great number of intertwined long molecules. If the intertwining of the molecules is completely random, the material does not show any anisotropy in its properties such as the thermal diffusivity, a. However, if molecular orientation does take place, for instance, by stretching of the material, the polymer shows an anisotropy of many physical properties. This is due to the fact that the atoms are covalently bonded along the chain molecules by a force much stronger than the van der Waals forces between the molecules. Accordingly, when a polymeric material is oriented, the mechanical strength along the orientation direction is much larger than that perpendicular to it. Therefore, it is expected that the orientation of polymer molecules also causes anisotropy of thermophysical properties such as thermal conductivity and thermal diffusivity. A unique feature of the forced Rayleigh scattering method is the capability to measure thermal diffusivity in any particular direction in anisotropic materials. Heat

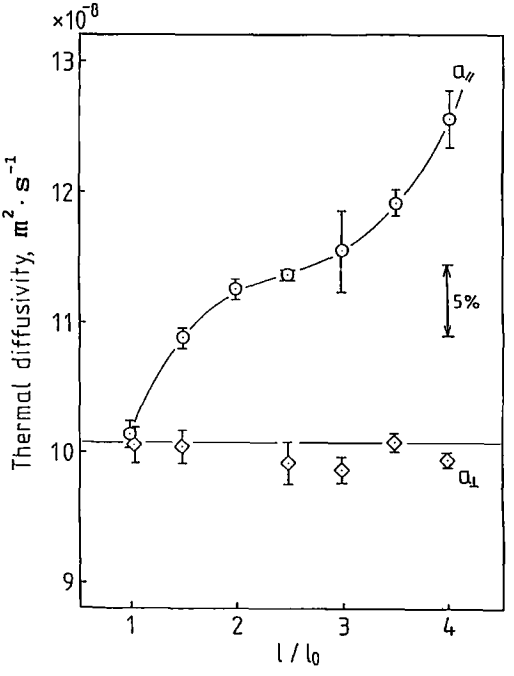

Fig. 3. Stretch ratio dependence of anisotropic thermal diffusivity due to molecular orientation (PMMA film). $a_{\parallel}$, thermal diffusivity along the flow direction; $a_{\perp}$, thermal diffusivity perpendicular to the flow direction.

conduction, in the process of relaxation of the temperature distribution, is considered to be one-dimensional in the x direction, perpendicular to the interference pattern. Therefore, the thermal diffusivity along this direction can be measured.

Figure 3 shows the dependence of the thermal diffusivity of PMMA on the stretch ratio, in the range $1 < l/l_0 < 4$. The variation of the thermal diffusivity along the stretch direction is shown, as is the variation perpendicular to the stretch direction. As shown, it was found that the thermal diffusivity in the stretch direction, $a_{\|}$, increases sharply with an increase in the stretch ratio, while the thermal diffusivity perpendicular to the stretch direction, $a_{\perp}$, shows little change. If the temperature of the anisotropic sample is raised, relaxation of molecular orientation may occur, and this would lead to a smaller difference between the thermal diffusivity of the stretched and that of the unstretched samples at higher temperature.

4.4. Thermal Diffusivity "Map"

An application of a high-speed optical method is the determination of the local distribution of thermal diffusivity. An interesting example is to measure this distribution by scanning the laser beam across the sample and then construct a "thermal diffusivity map" [23].

Normally when a polymer film is stretched strongly, the ends stretch more than the central part. And furthermore, the stretch direction at the ends is not the same as at the center. This means that the molecular orientation is not uniform along the film and there must be nonuniform distribution of the thermal diffusivity. Figure 4 shows a typical result of thermal diffusivity distribution on a film. The thermal diffusivity "map"

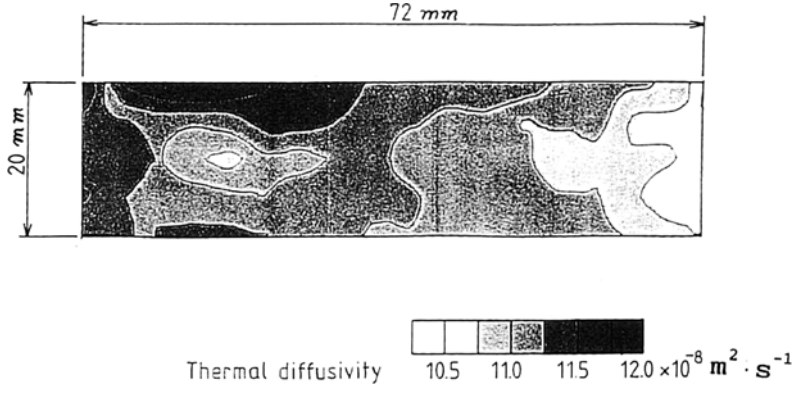

Fig. 4. An example of a thermal diffusivity "map" of a PVC film.

has successfully been obtained and a unique feature of the forced Rayleigh scattering method was demonstrated. The magnitude of nonuniformity caused by simple stretching was found to be 15%.

4.5. Thermal Anisotropy Due to Shear in Polymer Melts

Polymer materials can more easily be restructured in the molten state since the mobility of the molecules is higher. Therefore, molecular orientation takes place when a strong shear force is applied in the melt. Once molecular orientation is created, anisotropy of thermal as well as mechanical properties becomes significant. The quantitative relation between the flow pattern and the thermal anisotropy has been experimentally studied.

A series of *in situ* measurements of flowing polymer melts was performed [24]. Figure 5 shows a schematic diagram of the apparatus. The polymer sample was melted in a cylinder kept in an upright position and heated to the intended temperature. The cylinder was turned to the horizontal position and then the melt was extruded by a plunger through a gap 2 mm in width and 15 mm in height between two parallel glass plates. The flow condition was varied by adjusting the speed of the plunger.

In the flow of melt, the molecular orientation due to the shear force takes place in the direction of the flow and it causes anisotropy of the thermal diffusivity. Thermal diffusivity in the directions parallel to and

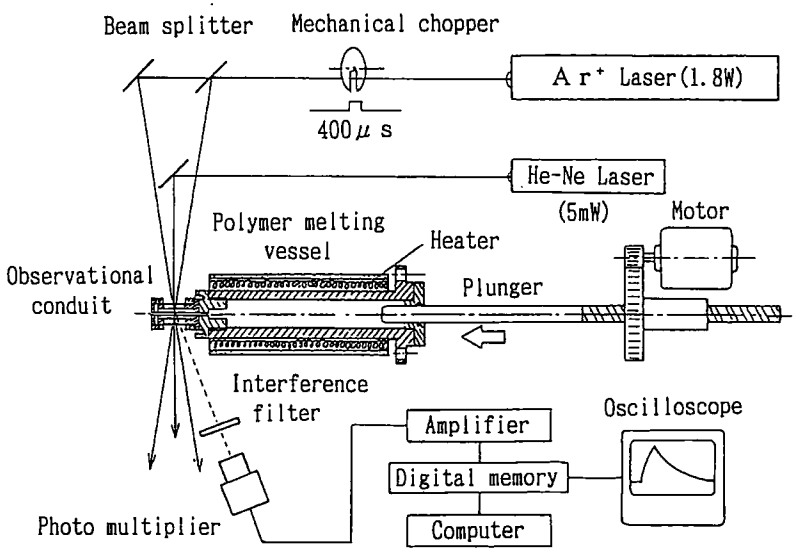

Fig. 5. Apparatus to measure the thermal diffusivity of a flowing polymer melt.

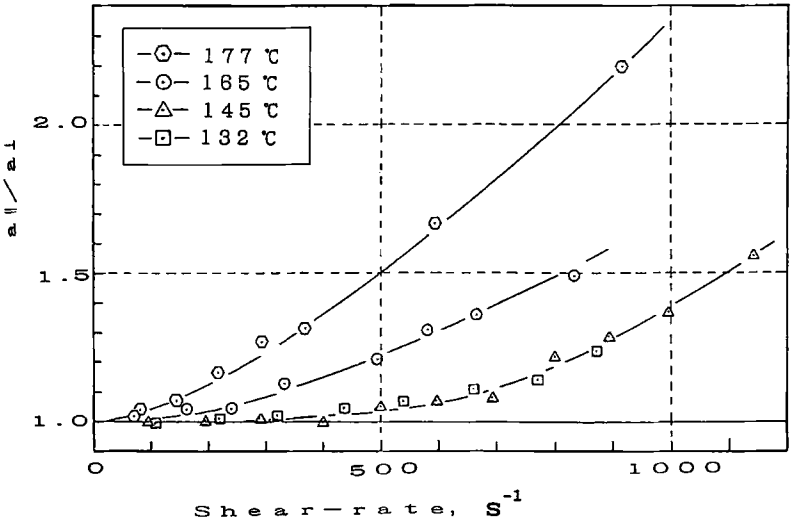

Fig. 6. Anisotropy of thermal diffusivity of molten polystyrenes as a function of the shear rate.

perpendicular to the flow axis was measured independently by varying the angle of the interference stripes (grating).

It is interesting that the thermal diffusivity parallel to the flow significantly increases with increasing mean velocity, while that perpendicular to the flow stays unchanged. This behavior is considered to be the result of molecular orientation and is consistent with the behavior of an extended polymer film studied earlier [22]. Anisotropy is characteristically expressed by the ratio of thermal diffusivities parallel to and perpendicular to the flow axis. The ratio is shown in Fig. 6 as a function of the shear rate at the wall, estimated by a simple hydrodynamic assumption. Anisotropy increases with increasing shear rate. The higher value at higher temperatures means that molecular orientation is more significant at increased mobility of the molecules due to lower viscosity at higher temperatures.

4.6. Transient Characteristics of Thermal Anisotropy in Polymer Melts [25–27]

To control and to simulate the process, reliable information is needed on transient and anisotropic behavior of thermophysical properties of polymer melts. Since theory has not been developed sufficiently, experimental studies have to be done. This section describes an experimental study of the transient behavior of thermal diffusivity of a molten polymer sample due to sudden

change of flow condition. The transient behavior of the thermal diffusivity and its anisotropy of flowing polymer melts under sudden loading of shear stress were measured. The aim of the study was to provide reliable information on transient characteristics and anisotropic behavior of the thermal diffusivity.

The results showed the existence of "overshoot" and relaxation phenomena for thermal diffusivity. This was shown for the first time for a thermal property, although similar phenomena had been known for mechanical properties of polymeric materials. The consistency with birefringence, a property closely related to molecular orientation, was also studied experimentally. The experimental system used in the present study is similar to that described in the earlier section. Sudden loading and unloading of shear stress were performed and controlled by means of a plunger with a driving system. Measurement of the thermal diffusivity in the transient state was made by means of an optical method over a small area, about 2 mm in diameter, through Pylex plates used as walls of the flow channel.

Measurements of the thermal diffusivity were made by forced Rayleigh scattering. The sample used in the present study was polystyrene which was

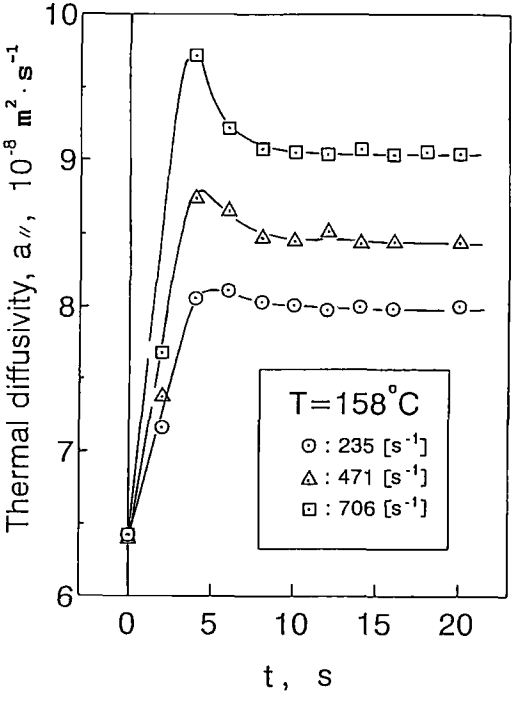

Fig. 7. Transient response of the thermal diffusivity of molten polystyrene after sudden loading of external force.

colored with a trace amount of dye to absorb the heating laser beam. The time dependence of the thermal diffusivity of the molten polymer layer was measured right after the sudden stop of steady flow and also after the sudden start of steady flow, from a still condition. At each measured point, 5 to 10 measurements were repeated and average values are shown in figures of experimental results. The experimental uncertainty of the measured data was estimated as $\pm 10\%$.

Figure 7 shows the transient response of the thermal diffusivity in the direction parallel to the flow direction. Symbols in this figure represent different magnitudes of shear rate in the flow near the wall. Details of calculations of shear rate and other factors are described in our earlier reports, mentioned before.

The existence of overshoot phenomena is clearly and quantitatively visible in Fig. 7. This is caused by transient behavior of molecular orientation. After sudden application of an external force, excessive orientation occurs at first and then the orientation gradually stabilizes to a well-balanced state of steady flow. The magnitude of overshoot is larger for a higher shear rate. At higher temperatures, the initial gradient toward overshoot is larger,

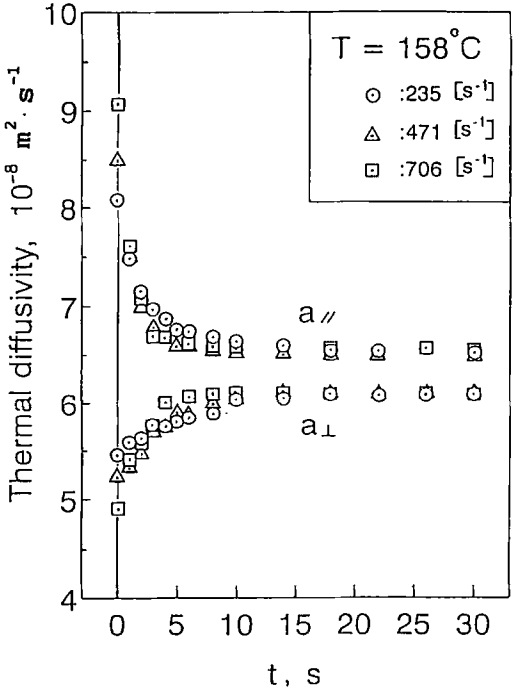

Fig. 8. Relaxation of the thermal diffusivity of molten polystyrene after stopping a steady flow.

and both the magnitude of overshoot and the final value under steady flow conditions are larger.

Figure 8 shows the relaxation of thermal diffusivity of a molten polymer after a sudden stop of steady flow. Here the thermal diffusivity in the direction parallel to and perpendicular to the flow is shown. They former is larger than the latter, and this is consistent with our previous results on a solid polymer film which has a molecular orientation due to one-dimensional extension. The relaxation with time is exponential as expressed by

$$\Delta a = \Delta_{a0} \exp(-t/\tau_0) \tag{4}$$

where Δ_a is the increase (or decrease) from the value at time t and Δ_{a0} is this increase (or decrease) at $t = 0$.

4.7. Microscale Thermal Diffusivity Measurement

Microscale heat transfer is an important subject, especially for such applications as cooling of computer chips, thermal control of micromachines, and diagnostics of biocells. Various computer simulations are widely used for these applications and require thermophysical property information. Microscale structures often have physical properties which are much different from those of bulk or larger-scale materials. They also show other interesting behavior, such as direction dependence, surface effects, and local migration of substance. However, quantitative information of thermophysical properties of microscale materials is lacking because of the total unavailability of proper measuring techniques. Especially microscale measurement of thermal conductivity or thermal diffusivity was impossible in the past, despite the strong need for the data.

We developed a new method for determining absolute values of the thermal diffusivity of a microscale sample. The sensing laser beam was focused by a lens system. In the first challenge [28, 29], we successfully showed that the thermal diffusivity of samples down to 100 μm could be determined on an absolute basis. Further development enabled measurements for sample size down to about 21 μm. Theory shows that the limit of measurement could be as low as 1.3 μm.

Although it is expected that the molecular orientation in polymeric materials causes mechanical as well as thermal anisotropy, the actual magnitude of the direction dependence of thermal diffusivity has not been known. A polymer fiber drawn through a nozzle from the molten condition has a remaining effect of molecular orientation in the direction of draw. In the study [30], measurements of the thermal diffusivity of a nylon fiber was made independently in both axial and radial directions. The fiber 650 μm in diameter was cut as shown in Fig. 9.

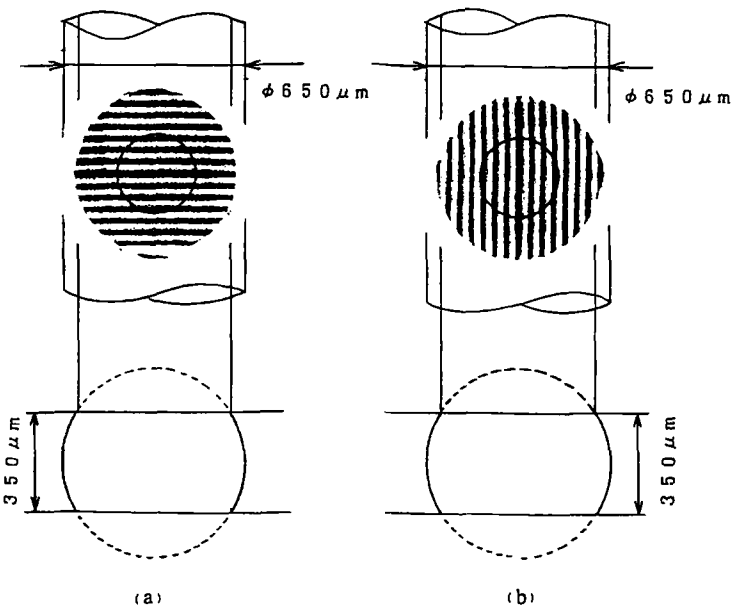

Fig. 9. Schematic view of a nylon 66 fiber. (a) Measurement of radial thermal diffusivity; (b) measurement of axial thermal diffusivity.

On the cut surface of a nylon fiber, fringe patterns were formed by the heating Ar^+ laser beams (see Figs. 9a and b). Heat conduction took place in the direction perpendicular to the fringes. Although the measured area was larger than that in the previous section (this condition corresponds to that of Raman–Nath diffraction), the effectiveness of microscale measurement could be demonstrated. Measured results of thermal diffusivity are as follows.

Axial $a = 1.21 \times 10^{-7}\ m^2 \cdot {}^{-1}$.

Radial $a = 0.57 \times 10^{-7}\ m^2 \cdot {}^{-1}$.

Note that the former is twice as large as the latter. This difference is supposed to be caused by molecular orientation in the axial direction. This qualitatively agrees with the effect in the case of an extended polymer film of a much larger size [22]. But the actual difference of the present sample, namely anisotropy, is much larger.

4.8. Measurement of Microscale Biomaterials

The behavior of a biomaterial depends on its temperature. Prediction and control of the temperature for cooling down and recovering are important for

processing, preservation, and treatment of various biomaterials in food/agricultural engineering and medical technology.

Difficulties in thermophysical property measurement of these materials are due to their complex structures, shape irregularity, self-protective reaction, etc. Therefore, the measurement of thermophysical properties has to be done in a short time with minimum disturbance. Optical methods are betters suited for this purpose than any other conventional methods.

One example is the measurement of the thermal diffusivity of a fish egg [31]. The core and the yolk have different properties, which have to be measured independently. By applying the microscale FRS method, local values of the thermal diffusivity have been measured, as shown in Fig. 10.

A minute amount of food coloring additives was used to color the sample to absorb the A^+ laser beam. After the preliminary test, it was found that the grating period should be smaller than about 50 μm for the present experimental setup. Measured results show different values for thermal diffusivity at the yolk and the core. The former shows a dependence on the grating period. This suggests the existence of some kind of photochemical effect due to laser beam on the sample, even though the beam intensity is not high and the temperature rise of the sample is small.

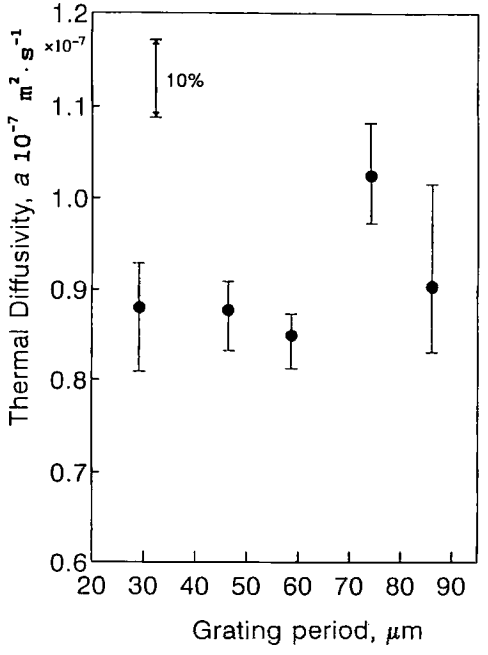

Fig. 10. Thermal diffusivity of the core of a salmon egg.

5. FUTURE OF OPTICAL TECHNIQUES

5.1. Development of Techniques

There are many possibilities for development of optical techniques for the measurement of thermophysical properties, especially transport properties.

One of the interesting directions is that of the next stage of microscale measurement. Difficulty lies in the wavelength of the heating or sensing beam. Further interesting developments will be the use of beams other than visible light, such as the X-ray and the neutron beam.

The use of extremely weak beams has interesting possibilities. Sensing techniques such as photon counting are progressing rapidly, and by accumulation of very weak light in a computer memory, measurement of thermal conductivity or diffusion coefficient will show interesting progress.

Features of optical sensing will be exhibited in remote sensing for extremely long-distance and ultrahigh-speed measurement. They will be combined with such techniques as imaging, scanning, and computer tomography.

5.2. Developments of Application

Optical techniques have advantages in applications for biomaterials under *in vivo* transient conditions. They are also suitable for detecting various types of material changes through measurement of, for instance, thermal diffusivity. Effects can be studied such as phase change, chemical reaction, surface deterioration, and fatigue due to internal stress. Quality control in production processes is an important possibility.

6. CONCLUSION

In the present review, the impressive progress of optical methods for measuring thermophysical properties has been illustrated, by introducing various recent applications of the forced Rayleigh scattering method. As mentioned in Section 5, there are numerous opportunities for optical methods, and it is expected that a new era of thermophysical property research is opening up.

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