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Rapid Measurement of the Thermal Diffusivity of Flowing Polymer Melts[1]

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Use of polymer materials for precision parts in advanced technology requires precise control of production processes such as melting, casting, and cooling processes. To achieve this, a reliable knowledge of thermophysical properties is essential. In the present study, submillisecond measurement of the thermal diffusivity of a flowing polymer melt was performed. Laminar flow of polymer melt through a narrow gap between two parallel plates creates a shear stress in the flow layer which then results in molecular orientation of polymer molecules. Once this orientation takes place, anisotropy of the thermal diffusivity is expected to occur. The method used in the present study was that of the so-called forced Rayleigh scattering method, a high-speed optical method. Nagasaka and Nagashima have recently improved the method significantly and applied it to the measurements on various difficult materials including molten salts. Measurement of thermal diffusivity anisotropy of molten polystyrene was successfully performed. Dependence of the thermal diffusivity and its anisotropy on shear force in flowing polymer melt was studied.

KEY WORDS: anisotropy; forced Rayleigh scattering method; polymer melt; polystyrene; thermal diffusivity.

1. INTRODUCTION

Thermophysical properties of polymer materials are strongly dependent on molecular structure and therefore on molecular orientation. Studies on correlational characteristics between anisotropy of the thermal conductivity and molecular orientation have been requested, especially for detailed control of the casting-molding process of advanced polymer materials.

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Advanced applications of polymer materials include optical instruments, high-precision parts, and fine fibrous materials. Molecular orientation can be created in polymer materials mainly by two kinds of processes. The first is mechanical deformation of solid materials and the second is application of shear force in molten state.

The present authors' group studied variations of the magnitude and the anisotropy of the thermal conductivity due to molecular orientation in a stretched polymer film [1], in an extended two-dimensional film [2], in excited liquid crystals [3], in an extruded thin polymer rod [4], in a molded spiral-flow polymer [5], etc. However, the ever-most-demanded information was data on molten polymer at actual flowing conditions. This information is essential for control and simulation of molding processes. Obviously, *in situ* measurement of the thermal conductivity of flowing fluid has been considered one of the most difficult tasks and no successful measurements have been found in the literature.

In the present paper, rapid measurement of the thermal diffusivity of a molten polymer in the actual flowing condition is reported. This was made possible by means of a rapid optical method which was greatly improved by Nagasaka and Nagashima [6, 7] in recent years.

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

2. PRINCIPLE OF THE METHOD AND APPARATUS

Details of the principle of the method and also the apparatus used for measurements on solid samples were described in the literature [7]. Here, an outline of the principle and the modification of the apparatus for polymer melts is described briefly as follows.

A short pulse of two heating laser beams hits the surface of a thin sample and creates a periodic distribution of temperature as shown in Fig. 1a. This distribution corresponds to the distribution of the refractive index. In the case of the present study, an argon ion laser was used for heating and two beams split by mirrors met on the sample with an angle of about 1°. This means about 50 stripes of refractive index distribution appeared within the laser-beam diameter of about 2 mm. These stripes acts as an optical grating to a probing beam which has a different wavelength. A He–Ne laser was used for probing. This means a diffracted beam can be detected if we set the detector shown schematically in Fig. 1b. We use a

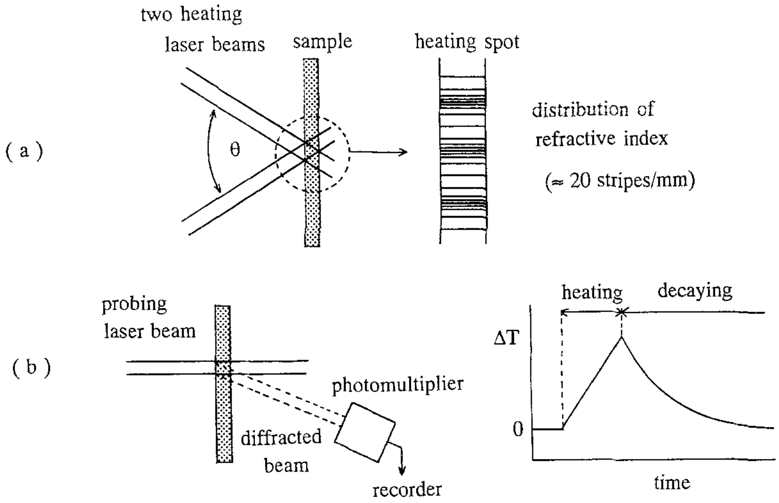

Fig. 1. Principle of the forced Rayleigh scattering method.

photomultiplier and the output signal was recorded in a digital memory. After the termination of a short heating period, the periodic temperature distribution in the sample equalizes due to heat conduction. Thus the analysis of this decaying process gives quantitative information of the thermal diffusivity of the sample. Actually, the thermal diffusivity a is calculated by the following equation:

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

where τ is the time constant of the decay and Λ is the grating period. The grating period Λ is calculated by the equation

$$\Lambda = \frac{\lambda_h}{2\sin(\theta/2)} \approx \frac{\lambda_h}{\theta} \tag{2}$$

where λ_h is the wavelength of the heating laser beam and θ is the crossing angle of the heating laser beams.

Another feature of the method is that it enables us to measure selectively the thermal diffusivity in the direction perpendicular to the temperature fringes.

In the present measurement, the heating period was about 400 μs and the total time needed for heating and decaying process was shorter than

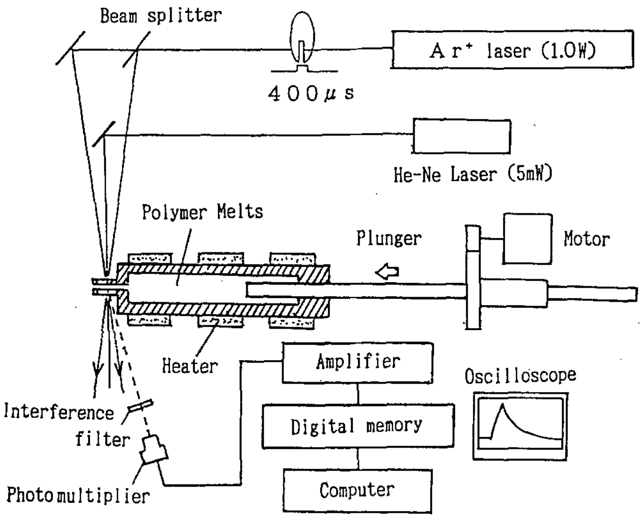

Fig. 2. Schematic diagram of the experimental apparatus.

1 ms. Various factors affecting the accuracy of the method were studied theoretically and experimentally and are described in detail in Ref. 6.

Figure 2 shows a schematic diagram of the apparatus. The heating laser beam was cut to a short pulse by a chopper and split into two beams by a beam splitter. 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. The optical system and the recording-calculating system are also shown in Fig. 2.

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

3. RESULTS AND DISCUSSION

A total of 34 measurements was performed in the temperature range from 132 to 177°C and the mean velocity range from 17 to 220 $\mathrm{mm \cdot s^{-1}}$. Every 1 of the 34 points is an average of 10 repeated measurements under the same flow conditions, and the scatter in these 10 data is less than 10%.

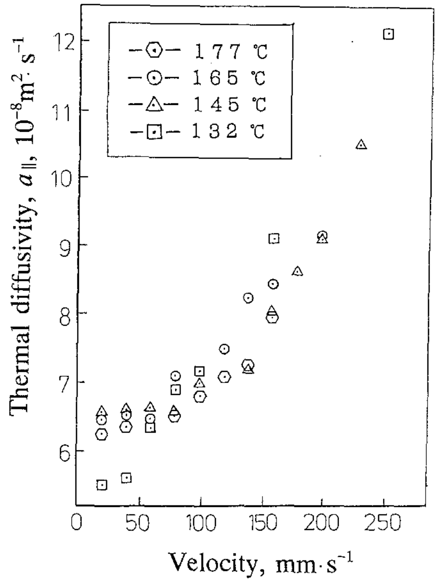

Fig. 3. Dependence of the thermal diffusivity parallel to the flow $a_{\parallel}$ on the mean velocity.

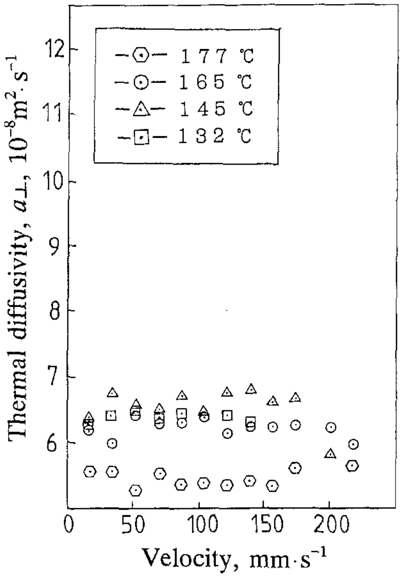

Fig. 4. Dependence of the thermal diffusivity perpendicular to the flow $a_{\perp}$ on the mean velocity.

Polystyrene samples were colored by adding a trace amount of red dye. The estimated uncertainty of the measured thermal diffusivity is about ±12%.

Figures 3 and 4 show the measured thermal diffusivity of flowing polystyrene melt in the directions parallel and perpendicular to the flow axis, respectively. It is interesting that the thermal diffusivity parallel to the flow $a_{\parallel}$ significantly increases with increasing mean velocity, while that perpendicular to the flow $a_{\perp}$ 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 previously [8]. Anisotropy as characteristically expressed by, for example, the ratio of thermal diffusivities parallel to and perpendicular to the flow axis. The ratio is shown in Fig. 5 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 with an increase in the mobility of the molecules due to lower viscosity at higher temperatures.

In order to find a more generalized correlation, we treat to express the ratio as a function of the Reynolds number Re defined as follows:

$$\mathrm{Re} = \frac{\rho U^{2-n} h^n}{K} \tag{3}$$

where ρ is the density, U is the mean velocity, n is the power index, h is the distance between the two parallel walls, and K is the consistency index.

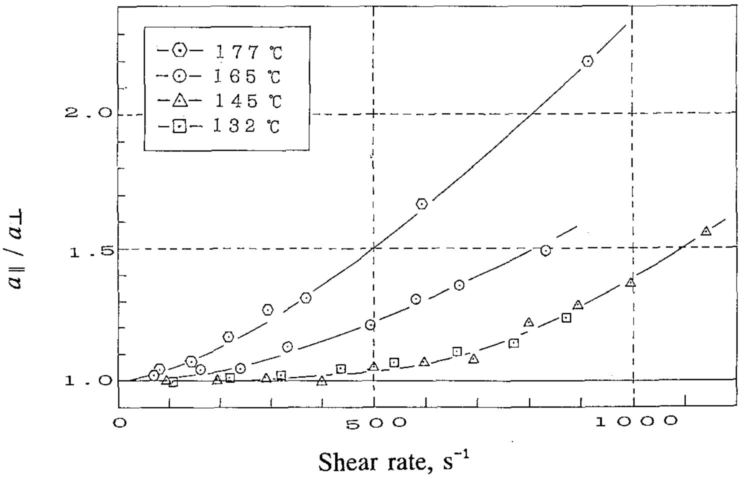

Fig. 5. Dependence of the thermal diffusivity ratio on the shear rate.

Following the Ostwald-de-Waele model for a non-Newtonian fluid [9], the relation between U and the pressure gradient is expressed as

$$U=\left(-\frac{dP}{dx}\right)^{1/n}\frac{n}{(2n+1)K^{1/n}}\left(\frac{h}{2}\right)^{(n+1)/n} \tag{4}$$

where dP/dx is the pressure gradient. In Eq. (4), the pressure gradient is assumed to correspond to the gradient of the shear rate, namely,

$$\frac{d\tau}{dy}=\frac{dP}{dx} \tag{5}$$

where τ is the shear stress.

The mean velocity U was determined experimentally and the pressure gradient was calcullated. Therefore, we can see the relation between the ratio of the thermal diffusivities and the shear stress gradient as shown in Fig. 6. The effect of temperature is shown more clearly in Fig. 6 than in Fig. 5.

The ratio is shown in Fig. 7 as a function of the Reynolds number defined in Eq. (3). It is clearly shown that all the experimental data can be correlated as a function of the Reynolds number within the experimental encertainty.

Although the molecular orientation is essentially a three-dimensional phenomenon, the behavior in a narrow parallel channel, such as that used

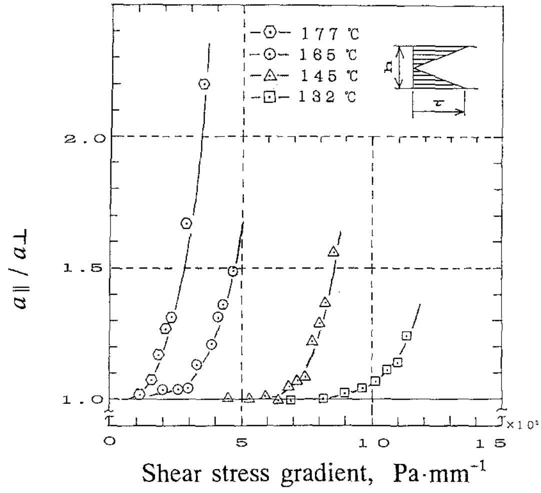

Fig. 6. Dependence of the thermal diffusivity ratio on the shear stress gradient.

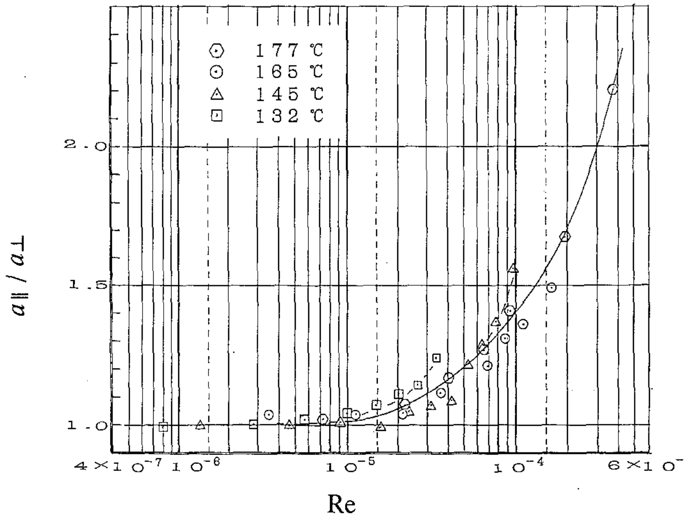

Fig. 7. Thermal diffusivity ratio as a function of the Reynolds number as defined by Eq. (3).

in the present study, can be assumed to represent the pattern of the molecular orientation and the anisotropy of thermophysical properties as a function of averaged flow characteristics.

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