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Experimental Investigation of the nonuniform heating effect in laser flash thermal diffusivity measurements

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ABSTRACT

Nonuniform heating effects of laser flash thermal diffusivity measurements were experimentally investigated. Spatial profiles of pulsed laser beams were monitored using a TV camera system. Direct laser beams from a Nd-glass laser were irregular because of multi-mode oscillation. Nonuniform heating error of larger than 5% was observed when thermal diffusivity of a disk shaped specimen of POCO AXM-5Q1 graphite was measured under irradiation by this irregular direct beam. A new technique was established to convert irregular direct beams into spatially uniform beams by using a step index optical fiber of large core diameter. The nonuniform heating error was reduced to the order of 1% when the specimen was irradiated by this uniform beam.

INTRODUCTION

It is generally accepted that the laser flash method is standard and most common to measure thermal diffusivity of solid materials (Righini and Cezairliyan, 1973). However, the uncertainty of thermal diffusivity values obtained by the laser flash method stays 5-10% at best because of several error factors. One of the largest is the nonuniform heating effect in which the front surface of a specimen is nonuniformly heated by a laser beam since spatial profiles of the pulsed laser beams used for the laser flash method are generally irregular because of multi mode oscillation. Nonuniform heating deviates heat flow inside of the specimen from one-dimensional heat flow on which the standard mathematical model of the laser flash method is founded.

Analytical calculation of the nonuniform heating effect is made by Watt (1966) and McKay and Schriempf (1976). Based on their solutions, the nonuniform heating error for specimens of various dimension has been calculated under various energy distribution of heating by Azumi et al. (1981) and Baba et al. (1986b).

From the experimental point of view (Taylor, 1975), there have been two major technical difficulties to solving the nonuniform heating effect. The first is the measurement of spatial profiles of pulsed laser beams. The second is a technique to obtain a spatially uniform laser beam with enough energy to make thermal diffusivity measurements by the laser flash method.

The National Research Laboratory of Metrology (NRLM) has been studying the nonuniform heating effect experimentally. One approach in NRLM is development of a thermographic technique by which history of temperature distribution is observed. This technique was successfully applied to measurements of one-dimensional profiles of high energy pulsed laser beams (Baba et al. 1986a). Another application of the thermographic technique was observation of lateral heat flow in a specimen when front surface of the specimen was heated by a nonuniform beam from a YAG laser (Arai et al. 1987). Recently, NRLM has established a new technique to convert irregular multi mode pulsed laser beams into spatially uniform beams using a step-index optical fiber of large core diameter (Baba et al. 1989). Mechanism to improve laser beam profile is multiple reflection of travelling light through the optical fiber. Spatial profile of the improved laser beam was monitored by using a TV camera system. Now we can use the improved laser beam as a pulse heat source of the laser flash method and the nonuniform heating effect can be reduced.

In this report, the nonuniform heating error of the laser flash thermal diffusivity measurements is experimentally investigated under nonuniform heating by direct laser beams and under uniform heating by improved laser beams. Experimental results are analyzed by calculation based on the differential equation of heat conduction referring to the monitored laser beam profile as the initial condition.

ANALYTICAL CALCULATION

McKay and Schriempf (1976) gave general analytical solution of temperature distribution within the specimen after the front surface is heated by a laser beam of arbitrary energy distribution. Since the beam after transmitting through an optical fiber is axially symmetric (Baba et al. 1989), only axially symmetric solutions are discussed in this report. We consider the disk shaped specimen of r in radius and d in thickness which is held adiabatic to the environment. When axially symmetric beam of radial energy distribution, $I(z)$, is absorbed at the front surface of the specimen, we define temperature distribution along the rear surface of the specimen as the function $T(z,t)$ where $0<z<r$ and $t>0$. The energy distribution function $I(z)$ is expanded to a series of 0-th order Bessel functions:

$$I(z)=1+\sum_{i=1}^{\infty} c_i J_0(\mu_i z) \tag{1}$$

where averaged energy density and radius of the specimen are normalized as unity. The expansion coefficient c_i is given by

$$c_i = \frac{\int_0^1 I(z) J_0(\mu_i z) z dz}{J_0^2(\mu_i) \int_0^1 I(z) z dz}. \tag{2}$$

The constant μ_i is the i-th positive root of $J_1(z)=0$ where $J_1(z)$ is the 1-st order Bessel function. Then $T(z,t)$ is expressed as:

$$T(z,t) = T_0 [1 + \sum_{i=1}^{\infty} c_i J_0(\mu_i z) \exp(-(\frac{2p\mu_i}{\pi})^2 \frac{t}{t_0})] \cdot [1 + 2\sum_{n=1}^{\infty} (-1)^n \exp(-n^2 \frac{t}{t_0})] \tag{3}$$

where $T_0=Q/C$: the equilibrium temperature increase, Q is total energy absorbed by the specimen, C is heat capacity of the specimen, $p=d/(2r)$: thickness to diameter ratio of the specimen, $t_0=d^2/(\pi^2\alpha)$: characteristic time of thermal diffusion and α is thermal diffusivity of the specimen. The nonuniform heating effect is expressed in the first parenthesis and the second parenthesis corresponds to the normal solution under the uniform heating.

Because irregularity of shorter range can be reduced more easily than that of longer range by using an optical fiber, the 1-st order term of the Bessel function expansion is particularly important. If temperature is measured at the center of the specimen ($z=0$) and the higher order terms than second are truncated, function (3) is simplified as follows:

$$T(0,t) = T_0 [1 + c_1 \exp(-(\frac{2p\mu_1}{\pi})^2 \frac{t}{t_0})] \cdot [1 + 2\sum_{n=1}^{\infty} (-1)^n \exp(-n^2 \frac{t}{t_0})]. \tag{4}$$

The standard algorithm to calculate thermal diffusivity from the laser flash method is the $t_{1/2}$ method (Parker et al., 1961) and thermal diffusivity, α, is represented by the following equation:

$$\alpha = \frac{0.1388 d^2}{t_{1/2}} \tag{5}$$

where $t_{1/2}$ is the time delay when the temperature of the rear surface reaches one half of the maximum temperature increase, T_{max}, after the front surface was heated by the laser pulse. If measurements are made under the ideal condition with no heat losses under uniform heating, T_{max} is equal to T_0.

When temperature is measured at the center, the temperature history of any combination of p and c_1 can be simulated based on equation (4) after normalizing T_0 and r as unity and the apparent thermal diffusivity value, α, is calculated by the $t_{1/2}$ method. Figure 1 shows apparent thermal diffusivity as a function of p in log scale with

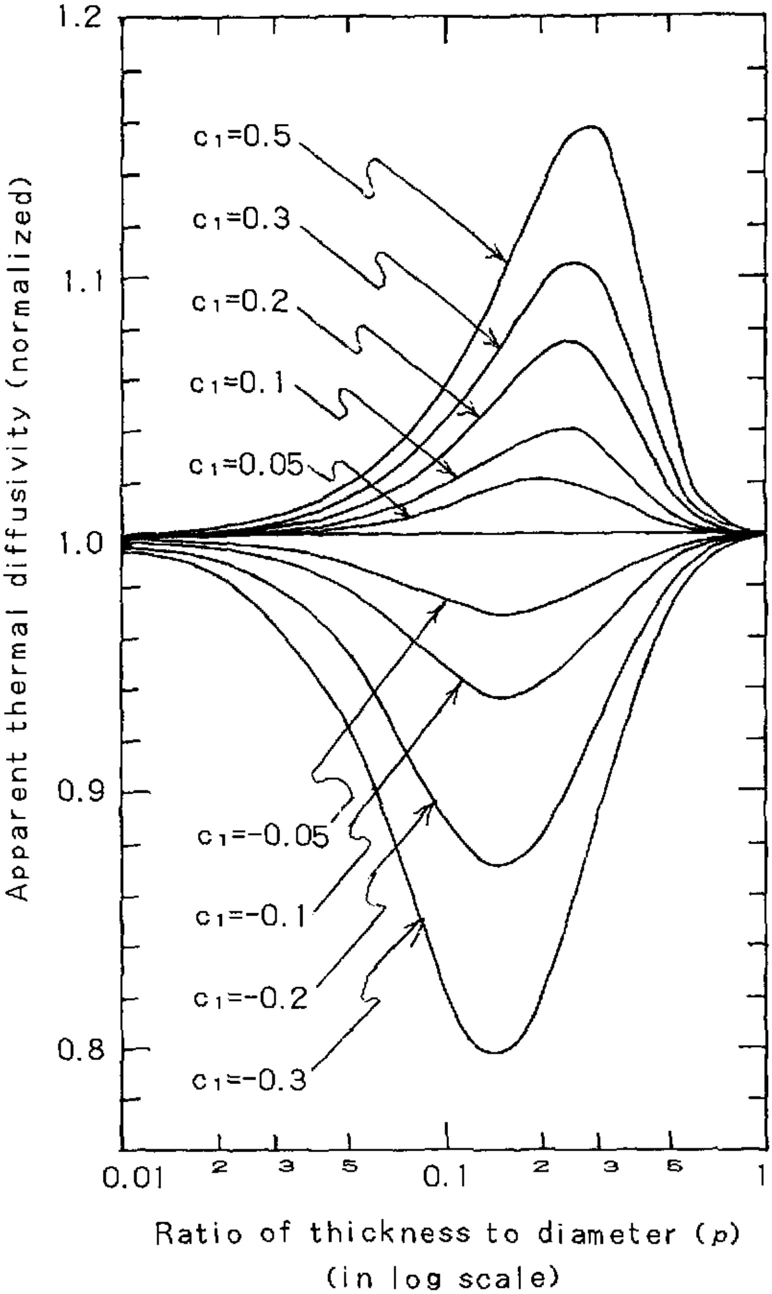

Fig.1. Calculated apparent thermal diffusivity when temperature is measured at the center of the specimen as a function of thickness to diameter ratio "p" with a parameter "c_1".

a parameter c_1. When the coefficient c_1 is positive, the beam has a hot center and when the coefficient c_1 is negative, the beam has a cold center. All curves have maximum (c_1>0) or minimum (c_1<0) between p=0.15 and p=0.3, where the nonuniform heating effect is largest under irradiation of the laser beam of the same profile.

Examples of calculated temperature increase curves are shown in Fig.2. These three curves are for the specimen of p=0.3. The left one is under hot center heating (c_1=0.25), the middle one is under uniform heating (c_1=0) and the right one is under cold center heating (c_1=-0.25). The hot center heating gives 8.5% higher and the cold center heating gives -9.3% lower than the thermal diffusivity value "unity" assumed for calculation. It should be noted that it is very difficult to tell the difference of the shape among three curves although they give considerably different apparent thermal diffusivity values. Thus, even if we do not see a hump (in the case of hot center) nor monotonic increase of temperature (in the case of cold center), we cannot exclude a possibility of the nonuniform heating error of the order of 10% involved.

EXPERIMENTAL INVESTIGATION

Experimental setup

Figure 3 shows a functional diagram of a laser flash thermal diffusivity measurement system developed in NRLM. In this series of measurements, the system was operated only at room temperature in order to suppress radiation heat losses from the surface of the specimen. A Nd-glass laser was used as a pulsed heat source. The pulse duration was about 700μs and the maximum energy per pulse was 17J under multi-mode oscillation. A step index optical fiber of 1mm in core diameter and 3m in length (DIAGUIDE ST1000H) was used to improve the spatial profile of the laser beam. Several points of the optical fiber were locally bent to homogenize density of

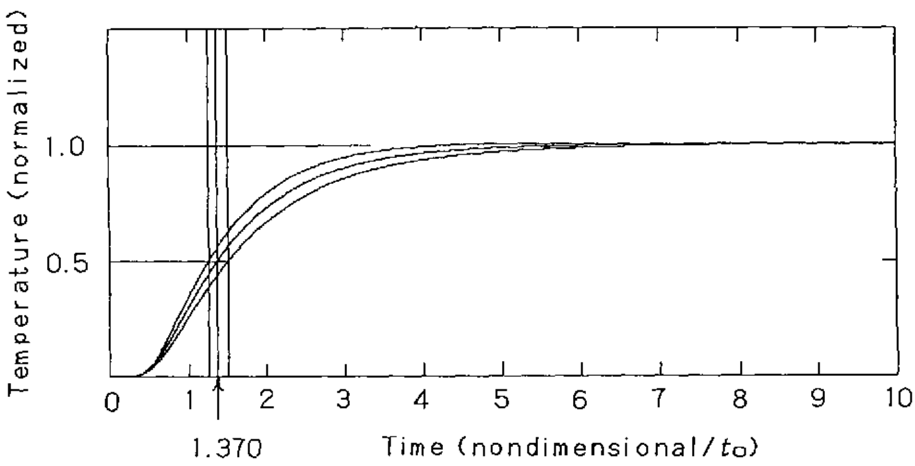

Fig.2. Calculated temperature increase curves for thickness to diameter ratio p=0.3. Left: hot center heating (c_1=0.25), Middle: uniform heating (c_1=0), Right: cold center heating (c_1=-0.25).

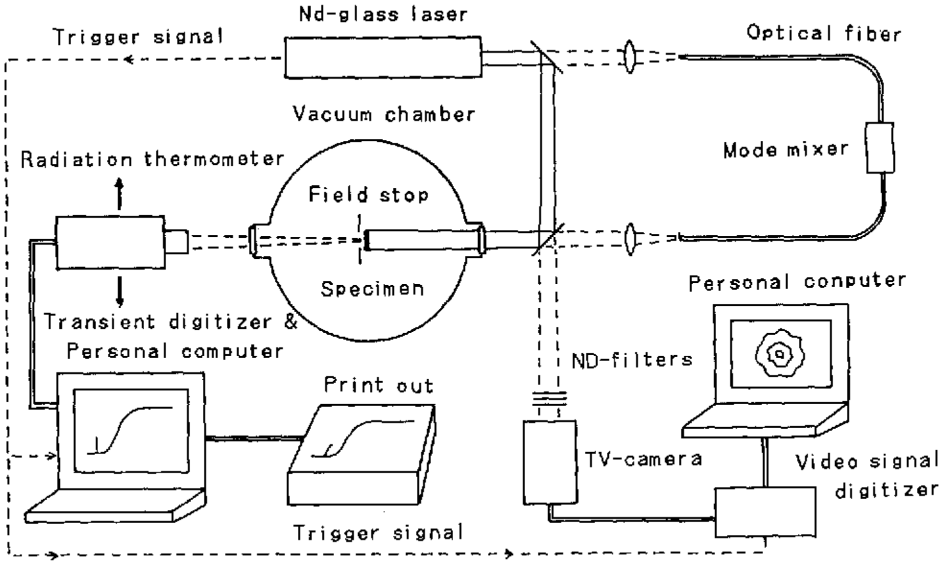

Fig.3. Functional diagram of the laser flash thermal diffusivity measurement system in NRLM equipped with a fiber optics to improve beam profiles and with a TV camera to monitor beam profiles.

light intensity inside the core. Thus, uniform irradiance was obtained at the output end of the step-index optical fiber and a real image of the output end was projected on the front surface of the specimen. The image was optically magnified to cover the entire specimen with a flat area.

When the beam profile was monitored, the laser beam was reflected by a mirror, attenuated by neutral density filters and the real image of the output end of the optical fiber was projected onto the detective area of a Super-CHALNICON E5476 tube of Toshiba mounted in a TV camera. Video signal was digitized and processed in an NEC PC-9801V personal computer. Temperature history at the rear surface of the specimen was measured with an infrared radiation thermometer designed and assembled in NRLM. The observed wavelength by the InSb infrared detecter was 3-4.5μm. Target size was 2mm in diameter. The response time is 10μs with temperature resolution of higher than 0.1K. This radiation thermometer was fixed on an adjustable stage and the target can be shifted along the rear surface of the specimen.

The analogue signal from the radiation thermometer was digitized and stored into a transient-wave memory, Model TMR-120 of Electronica Co Ltd., with a capacity of 16K words of 12bits. The data stored in the transient-wave memory was transferred to an NEC 9801-DA personal computer and saved onto a floppy disk. The saved transient temperature distribution data can be analyzed, displayed on CRT and printed out during and/or after the measurement. The apparent thermal diffusivity value based on the $t_{1/2}$ method was calculated from the measured temperature increase curve in real time.

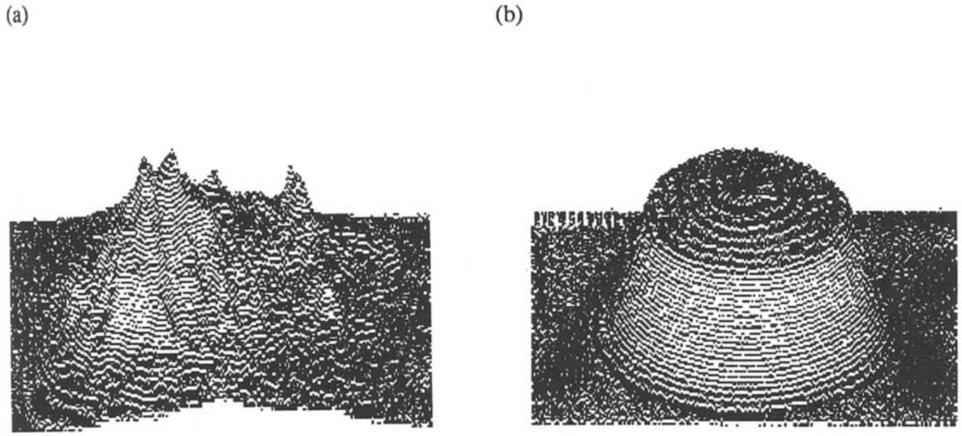

Fig.4. Three-dimensional profiles of an irregular direct laser beam (a) and an improved laser beam by using an optical fiber (b).

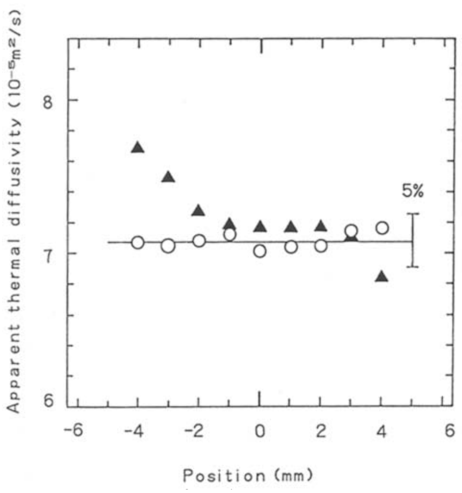

Fig.5. Distribution of apparent thermal diffusivity values of POCO AXM-5Q1 graphite.
▲ : nonuniform heating by a direct beam
○ : uniform heating by an improved beam

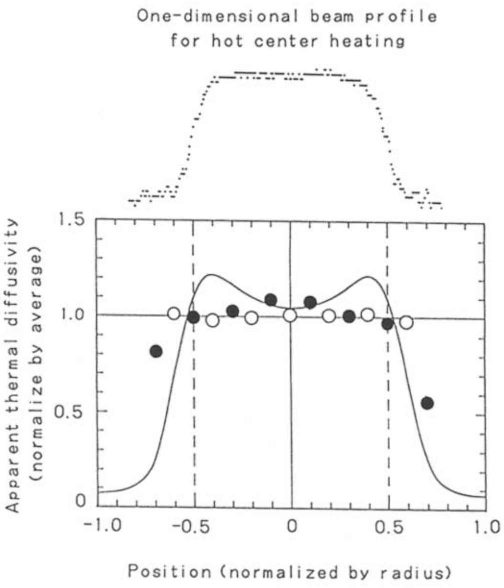

Fig.6. Distribution of apparent thermal diffusivity values of glassy carbon.
● : hot center heating
○ : uniform heating

Experimental results

Local apparent thermal diffusivity values of a POCO AXM-5Q1 graphite of 12.7mm in diameter and 1.73mm in thickness were measured under nonuniform heating by an irregular direct beam and under uniform heating by an improved beam after transmitting through an optical fiber. Spatial profiles of the irregular direct beam and the improved beam were measured with the TV camera system and their three dimensional display are shown in Fig.4.

Local thermal diffusivity values were obtained with 1mm interval by shifting the radiation thermometer. Figure 5 shows distribution of local thermal diffusivity values where closed triangles correspond to nonuniform heating by the direct beam and open circles correspond to uniform heating by the improved beam. The average apparent thermal diffusivity $\overline{\alpha}$ is $7.230 \times 10^{-5} m^2/s$ and the standard deviation σ is 3.22% under the nonuniform heating whereas $\overline{\alpha}$ is $7.077 \times 10^{-5} m^2/s$ and σ is 0.72% under the uniform heating. This result demonstrates that nonuniform heating error of the order of 5% is unavoidable as far as irregular beams direct from high energy pulsed lasers under multi mode oscillation are used for pulse heating. This error can be reduced to less than 1% if uniform beams after transmitting through an optical fiber are used as pulsed heat sources for the laser flash method.

Comparison with calculation

A glassy carbon of 10.0mm in diameter and 1.015mm in thickness was used as a specimen to obtain data to be compared with theoretical calculation. Spatially uniform beam, of which one-dimensional energy distribution was shown in the upper part of Fig.6, was projected onto the central part of the front surface of the specimen with 5mm in diameter for hot center heating. The same beam was magnified and projected onto the entire front surface of the specimen with 10mm in diameter for uniform heating.

Figure 7 shows temperature history measured at the rear surface of the specimen under the hot center heating. When temperature is measured close to the center, the temperature history curve has a maximum and decrease gradually as curve A (0.5mm from the center) and curve B (1.5mm from the center). As the measurement point shifts apart from the center, the slope of decrease becomes gentler as curve C (2.5mm from the center). When the measurement point is close to edge of the specimen, the temperature monotonically increases as curve D (3.5mm from the center). Thus, the shape of temperature history curve is quite different each other dependent on the measurement point. The apparent thermal diffusivity values determined by the $t_{1/2}$ method are $6.33 \times 10^{-6} m^2/s$ by curve A, $5.92 \times 10^{-6} m^2/s$ by curve B, $5.69 \times 10^{-6} m^2/s$ by curve C and $3.25 \times 10^{-6} m^2/s$ by curve D.

Figure 8 shows temperature history measured at four different points at the rear surface of the specimen under the uniform heating. In contrast with Fig.7, four temperature history curves have essentially the same shape. The apparent thermal diffusivity values determined by the $t_{1/2}$ method are $5.96 \times 10^{-6} m^2/s$ by curve E (the center), $5.99 \times 10^{-6} m^2/s$ by curve F (1mm from the center), $6.05 \times 10^{-6} m^2/s$ by curve G (2mm from the center) and $5.78 \times 10^{-6} m^2/s$ (3mm from the center) by curve H. The average

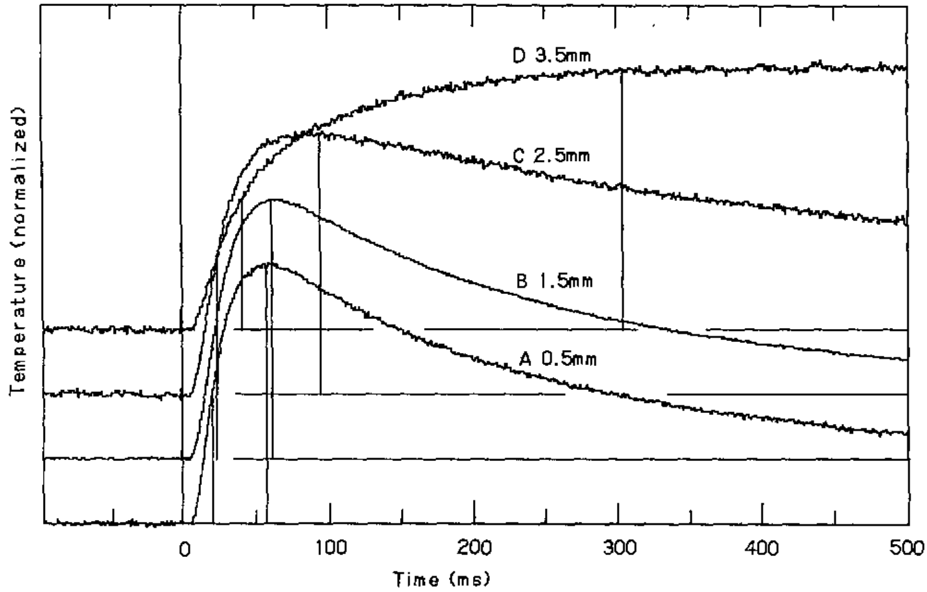

Fig.7. Temperature history curves at different observing points when a glassy carbon specimen was measured under hot center heating.

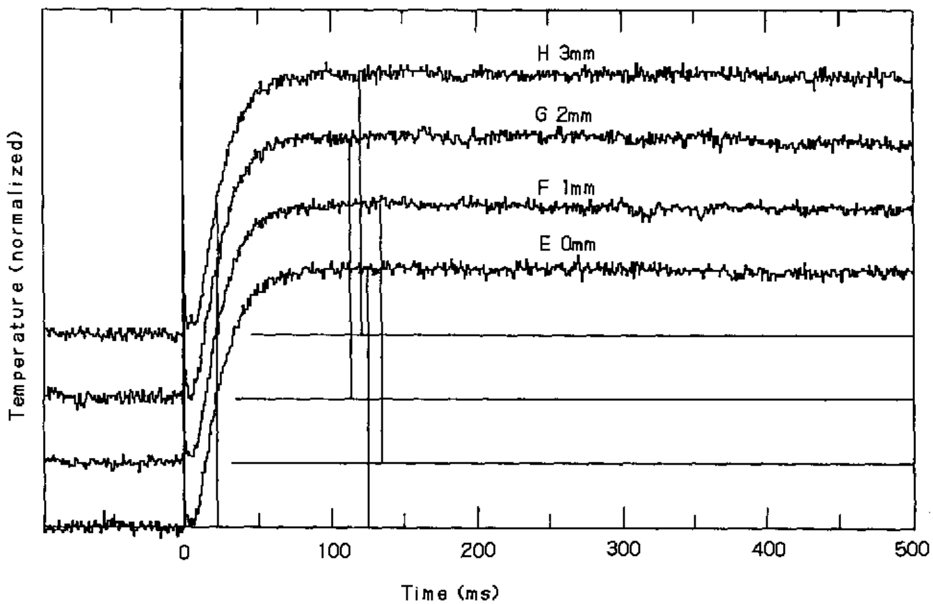

Fig.8. Temperature history curves at different observing points when a glassy carbon specimen was measured under uniform heating.

value of the thermal diffusivity under the uniform heating, $\bar{\alpha}$, is $5.89\times10^{-6} m^2/s$ for totally 27 measurements.

Distribution of local apparent thermal diffusivity values are plotted in Fig.6. Values under the hot center heating were represented by closed circles and values under the uniform heating were represented by open circles. The distribution under the hot center heating is compared with the calculation. If we normalize the radial coordinate z by the radius (r=5mm), the energy distribution of the improved laser beam projected on the central part is approximated by the function $I(z)=1$ for $0<z<0.5$ and $I(z)=0$ for $0.5<z<1$. When $I(z)$ is expanded as an infinite series of Bessel function as equation (1), the coefficient c_i is given as:

$$c_i=\frac{4J_1(\frac{\mu_i}{2})}{\mu_i J_0^2(\mu_i)}. \tag{6}$$

Then temperature increase curves are calculated by equation (3) with substituting p=0.1015.

The calculated distribution of local apparent thermal diffusivity values is represented by a solid line in Fig.6 and close to the closed circles in general except that the calculated curve has a local minimum at z=0 but the experimental result has a maximum at z=0. This difference can be explained by two assumptions on which theoretical calculation is founded. The first is that calculated value is at point "z" with infinitesimally small area whereas in experiment the radiation thermometer observes averaged temperature over finite target size of 2mm in diameter. Then experimental result should correspond to some average over approximately 2mm in z-axis and the concave dependence in the central area is diluted. The second is that the real energy distribution function I(z) does not change stepwise as assumed. This must also reduce the concave shape of the calculated result.

CONCLUSION

It is demonstrated that nonuniform heating error of the order of 5% is unavoidable as far as direct beams from high energy pulsed lasers under multi mode oscillation are used as pulse heat sources for the laser flash thermal diffusivity measurements. Improvement of laser beam profiles using an optical fiber is the promising solution to reduce the nonuniform heating error to the order of 1%. Details about principle and procedure to get uniform beams by using optical fiber and the technique to monitor the obtained laser beam profile are described in the reference (Baba, Hong and Ono, 1989). Beams of uniformity better than 5% can be obtained by using this optical fiber technique without special skill. The maximum limit of the nonuniform heating error can be estimated by referring to Fig.1. For example when c_1=0.05 (hot center of 5% nonuniformity) maximum error is approximately +2% and when c_1=-0.05 (cold center of 5% nonuniformity) maximum error is approximately -3%.

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