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The measurement of thermal diffusivities for semi-infinite solids using the photothermal displacement method☆

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ABSTRACT

This paper develops a more accurate method of measurement for thermal diffusivity using photothermal displacement spectroscopy. In previous studies, thermal diffusivities for materials with a finite thickness have been determined by the angle of deformation and the phase difference, with regard to the relative positions of the pump and probe beams. In this study, a complete theoretical treatment and experimental measurement, through the photothermal displacement technique, have been conducted for semi-infinite solid materials. To verify the method developed, the measurement has been carried out for an alloy of pure copper and nickel with both finite and semi-infinite thicknesses. In the results of the study, the measured values for semi-infinite materials differed from those reported in the literature by at most 1% of the standard deviation, while the corresponding differences for finite materials were at most 3% of the standard deviation. Consequently, the model for semi-infinite solids is proposed as a highly accurate method for measuring thermal diffusivity.

1. Introduction

The photothermal technique is a very useful tool for measuring thermo-physical properties of solids, liquids, etc. As methods of measuring thermo-physical properties using the photothermal effect, photothermal radiometry, photothermal refraction, photothermal deflection, and the photothermal displacement method have been reported and developed by many researchers. Recently, there are lots of studies going on about measuring thermal characteristics of thin film, nano objects and layered structure, anisotropic materials using photothermal effect [1–9].

Among the above methods, the photothermal displacement method used in this study is useful for measuring the gradient and phase of the thermoelastic deformation that is produced by the absorption of light energy at the surface of materials during a period of heating. In 1983, Olmstead et al. [10] introduced a two-dimensional model wherein the pump beam was modulated as a sine wave and the magnitude and phase difference of the deformation gradient on the surface of material were calculated. By comparing theoretical results with experimental findings, Olmstead et al. [10] suggested the possibility of measuring thermal properties through the photothermal displacement method. In 1991, Li et al. [11] developed a two-dimensional model, wherein the pump beam was modulated as a square wave, and calculated the magnitude of deformation at one point. They defined the characteristic frequency as that modulation frequency at which the magnitude of deformation drops rapidly. In 1999, by applying the deformation gradient equation which had been presented by Olmstead et al., Ogawa et al. [12] determined the thermal diffusivity from the relationship between the phase difference of the deformation gradient and the change of modulation frequency at one point. In 2000, Lee et al. [13] proposed a "minimum phase method," which uses the relative position at the minimum phase difference and the length of thermal diffusion. However, this method has limitations in practice, because it is difficult to experimentally find the relative position at the minimum value of the phase difference.

Since previous studies have been conducted on samples with relatively small thickness, the error in the measuring the sample thickness causes relatively large uncertainty because of the sample preparation process such as surface polishing and removing the sample from the polishing mount after surface polishing. But the infinite sample does not need further jobs after polishing therefore it has lots of benefits. Also, in the case of materials for which the length of thermal diffusion is small, the adiabatic condition, between the back of the sample surface and ambient, results in theoretical error. Therefore, a model, which is not influenced by sample thickness, is proposed in this study by using semi-infinite solid materials. To verify the accuracy of the model,

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Nomenclature

a	radius of the pump beam (m)
f	modulation frequency (Hz)
J_o	zero-order Bessel function
J_1	first-order Bessel function
k	thermal conductivity (W/m K)
L	thickness of sample (m)
L_{th}	length of thermal diffusion (m)
P	power of the pump beam (W)
Q	heat source (W/m^3)
T	temperature (K)
t	time (s)
u	displacement (m)

Greek letters

Φ	deformation gradient (radian)
α	thermal diffusivity (m^2/s)
α_{th}	thermal expansion coefficient (1/K)
β	integration variable (Hankel variable)
ϕ	displacement potential
λ	optical absorption coefficient (1/m)
θ	phase difference (deg)
ν	Poisson's ratio
σ	stress (N/m^2)
ω	angular frequency (1/s)
ψ	love function

experimental measurements are carried out for a pure copper and nickel alloy.

2. Principle and theory

2.1. Principle

Fig. 1 schematically shows the principle of the photothermal displacement method. The method is based on the detection of the deformation of the sample surface that is produced by the absorption of energy from a modulated light beam (pump beam) that is incident upon the sample. The heating of the sample by the pump beam produces a temperature distribution and thermoelastic deformation of the sample, which can be measured by detecting the deflection of the probe beam that is reflected by the sample surface. Information on the thermophysical properties of the sample can be obtained from the phase difference between the deflected probe and the modulated pump beam. Thermoelastic deformation is affected by thermal and optical properties, such as thermal diffusivity, absorption coefficient, etc. Therefore, under the assumption that refraction by the air on the surface can be ignored, the difference between the incidence and reflection angles of a probe beam is proportional to the gradient of deformation, as shown in Eq. (1).

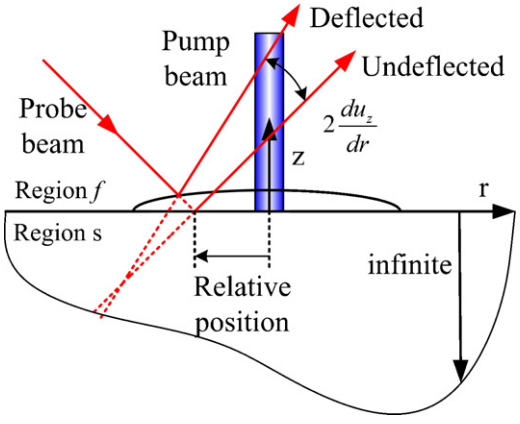

Fig. 1. The principle of the photothermal displacement method and the theoretical model.

$$\Phi = 2\frac{du_z}{dr}\bigg|_{z=0} \tag{1}$$

in (1), du_z/dr is the gradient of deformation.

Generally speaking, there are two ways of determining thermal diffusivity through photothermal displacement: using the angle of deformation from the surface of the specimen; and using the phase difference between the pump beam and the deflected probe beam. Since the pump beam is modulated, the gradient of deformation is modulated with the same frequency as that of the modulated pump beam. However, there is the time lag in the thermal diffusion of the pump beam energy; as a result, a phase difference occurs between the modulated pump beam and the deflected probe beam. The phase difference can be experimentally measured for unknown materials and also can be theoretically calculated for random thermal diffusivities using Eq. (2), which is given below. Then, thermal diffusivities can be determined by iteratively comparing the theoretical phase difference with the measured value. The phase difference (θ) can be expressed as:

$$\theta(r, f, a, \alpha) = \tan^{-1}\left[\frac{Im[\Phi]}{Re[\Phi]}\right] \tag{2}$$

Using the deformation and phase difference methods, the thermal diffusivity can be determined by comparing the experimental and theoretical results.

2.2. Theory

To obtain the gradient of deformation in Eq. (1), the temperature distribution for the specimen must be determined. For this, a two-dimensional solid model, which is infinitely long in the direction of r and z, is adopted as shown in Fig. 1 [13,14]. In the analysis of temperature, the conduction of heat is considered to be significant whereas the convection and radiation of heat are treated as being negligible. The governing equation is the 2 D heat-conduction equation with the heat source in cylindrical coordinates.

$$\nabla^2 T_i - \frac{1}{\alpha_i}\frac{\partial T_i}{\partial t} = -\frac{1}{k_i}Q_i \quad (i = f, s) \tag{3}$$

In Eq. (3), T is the temperature, k is the thermal conductivity, α is the thermal diffusivity, and Q is the heat source that is produced by a pump beam. The temperature and heat source are functions of the direction of the radius (r), the direction of the normal to the surface (z), and the time (t). The subscript f indicates the front gas of the specimen and the subscript s represents the domain of the specimen.

The heat flux and temperature are assumed to be zero at $z = \infty$ because the thermal effect of the pump beam cannot influence the specimen when z approaches infinity. Also, as the rise in temperature of the specimen by the pump beam is insignificant, the heat transfer by either convection or radiation is not considered [14]. Therefore, the boundary conditions are

$$k_f\frac{\partial T_f}{\partial z}\bigg|_{z=0} = k_s\frac{\partial T_s}{\partial z}\bigg|_{z=0}, \quad T_f|_{z=0} = T_s|_{z=0}, \quad \lim_{z\to\infty} T_f = \lim_{z\to-\infty} T_s = 0. \tag{4}$$

The heat source is the pump beam, which has a Gaussian distribution of intensity and is controlled by the modulation frequency (f). The radius of the pump beam is considered to be $1/e$ value of the

maximum intensity and the absorption coefficient (λ) is defined by the exponential law of light absorption. At the front gas region, light energy is not absorbed. Therefore the heat source is given by

$$Q_f = 0 \tag{5a}$$

and

$$Q_s = \frac{P\lambda_s}{4\pi a^2} e^{-r^2/a^2+\lambda_s z}[1+\cos(\omega t)] \tag{5b}$$

where P represents the energy absorbed into the specimen and is decided by the output power of the pump beam and the reflectivity of the specimen. The heat source, $Q(r,z,t)$, is comprised of a time-independent term and a time-dependent term that always oscillates with a given frequency. The time-independent term is not considered since it does not affect the phase of deformation; hence, only the time-dependent term is considered.

Due to the cylindrically symmetric geometry of the problem, the differential equations that describe the diffusion process can be solved by the Hankel transform method. The solution for the temperature distribution is obtained through an integral over the single variable β, which represents the Hankel variable in the radial direction.

$$T_f(r,z,t) = \frac{P\lambda_s e^{i\omega t}}{8\pi k_s} \int_0^\infty \beta \mathrm{d}\beta J_0(\beta r) \left(\frac{k_s\delta_s - k_s\lambda_s}{k_f\delta_f + k_s\delta_s}\right) \left(\frac{e^{-\beta^2 a^2/4}}{\delta_s^2 - \lambda_s^2}\right) e^{-\delta_f z} \tag{6a}$$

$$T_s(r,z,t) = \frac{P\lambda_s e^{i\omega t}}{8\pi k_s} \int_0^\infty \beta \mathrm{d}\beta J_0(\beta r) \left(e^{\lambda_s z} - \frac{k_s\lambda_s + k_f\delta_f}{k_f\delta_f + k_s\delta_s} e^{\delta_s z}\right) \times \left(\frac{e^{-\beta^2 a^2/4}}{\delta_s^2 - \lambda_s^2}\right) \tag{6b}$$

in (6a) and (6b), $J_0(\beta r)$ is the Bessel function of zeroth order and $\delta_i = (\beta^2 + i\omega/\alpha_i)^{1/2} (i = \mathrm{f}, \mathrm{s})$.

Assuming that the variation of the temperature field with time is small, the inertia terms in the thermoelastic equation may be neglected. Then, the thermoelastic equation with temperature $T(r,z,t)$ is given in [13,15] and reproduced below.

$$\nabla^2 \vec{u} + \frac{\nabla(\nabla \cdot \vec{u})}{1-2\nu} = \frac{2(1+\nu)}{1-2\nu} \alpha_{th} \nabla T \tag{7}$$

with stress-free boundary conditions

$$\sigma_{rz}|_{z=0} = 0, \sigma_{zz}|_{z=0} = 0, \lim_{z\to-\infty} \vec{u} = 0, \tag{8}$$

where ν is the Poisson ratio, $\vec{u}$ is the displacement vector, α_{th} is the thermal expansion coefficient, σ_{rz} and σ_{zz} are the two stress components, and T is the temperature inside the sample, which is previously obtained from Eq. (6). Owing to the geometry of the pump beam, the solution $u(r,z)$ can be expressed in cylindrical coordinates by introducing the displacement potential ϕ and the Love function ψ as follows:

$$\vec{u} = \nabla\phi + \frac{\{2(1-\nu)\nabla^2\psi - \nabla(\nabla\cdot\psi)\}}{(1-2\nu)} \tag{9}$$

where ϕ and ψ are expressed by the following differential equations,

$$\nabla^2\phi = \frac{1+\nu}{1-\nu}\alpha_{th} T \tag{10}$$

and

$$\nabla^4\psi = 0 \tag{11}$$

Note that to express the deflection angle as shown in Eq. (1), one needs only the normal component of the displacement (i.e., u_z at $z=0$) for calculating the gradient of the displacement in the r-direction as given below.

$$\left.\frac{\mathrm{d}u_z}{\mathrm{d}r}\right|_{z=0} = -\frac{(1+\nu)\alpha_{th} P\lambda_s}{4\pi k_s} \int_0^\infty \beta^2 d\beta J_1(\beta r) \frac{e^{-\beta^2 a^2/4}}{\delta_s^2 - \lambda_s^2} \times \left(\frac{1}{\lambda_s+\beta} - \frac{k_s\lambda_s + k_f\delta_f}{(\delta_s+\beta)(k_f\delta_f + k_s\delta_s)}\right) \tag{12}$$

in Eq. (12), $J_1(\beta r)$ is the Bessel function of the first order.

For a known radius of the pump beam, the phase difference is determined by integrating Eq. (12), which consists of a complex number and a rather intricate integral term that includes thermal diffusivity, etc. In terms of the thermal diffusivity and the modulation frequency, the length of thermal diffusion can be expressed as $L_{th} = (\alpha/\pi f)^{1/2}$.

3. Experiment

The experimental arrangement is shown in Fig. 2. The pump beam is obtained by the acousto-optic modulation of a continuous-wave laser beam that is delivered by an Ar+ laser (wavelength: 488 nm). Typically, 0.7 W of energy was used to heat the sample. The power of the beam decreases to 0.3 W when the beam approaches the surface of the sample after passing through the acousto-optic modulator, the lens, mirrors, and the collimator. Further, after accounting for the reflection from the surface of the sample, the absorbed energy eventually becomes about 0.2 W. Also, it is critically important that the spatial profile of the pump beam be Gaussian. The typical size of the spot of the pump beam on the sample is 55 μm in radius, based on the $1/e$ point of the intensity profile of the pump beam. The probe beam is provided by a He–Ne laser (wavelength: 633 nm), with a power of 5 mW and a beam diameter of 30 μm. The size of the beam on the sample surface is determined by the knife-edge method. Both the pump and the probe beam are expanded through collimators.

The pump beam is periodically modulated at a frequency (f) to create a steady, periodic, thermoelastic deformation in the sample. The deflection of the probe beam is measured by using a two-dimensional position sensor. An interference filter is fitted in front of the sensor to detect only the wavelength of the probe beam. The amplitude and the phase difference of the signal are measured by a lock-in amplifier, which is synchronized with the modulation frequency.

The position of the probe beam is aligned with the centerline of the spot of the pump beam. After the positions of the probe beam and the pump beam are aligned, the measurement is carried out by moving the probe beam along the centerline of the deformation. The probe beam is moved along the centerline from the center of the spot of the pump beam set the relative position of a probe beam by using an arrangement of mirrors and micro-positioners. The micro-positioners are controlled with an accuracy of ±0.5 μm. Experiments were carried out for samples of an pure copper and nickel alloy, which was polished using alumina paste.

4. Results and discussion

4.1. Critical thickness

The model for semi-infinite solids can be formulated by setting the thickness in the z-direction as being infinite for the purpose of theoretical analysis. However, for experimentation, we need to come up with a sample of finite thickness that can be considered as being physically similar to a semi-infinite solid. Therefore, the reasonable thickness for the sample is decided through a compari-

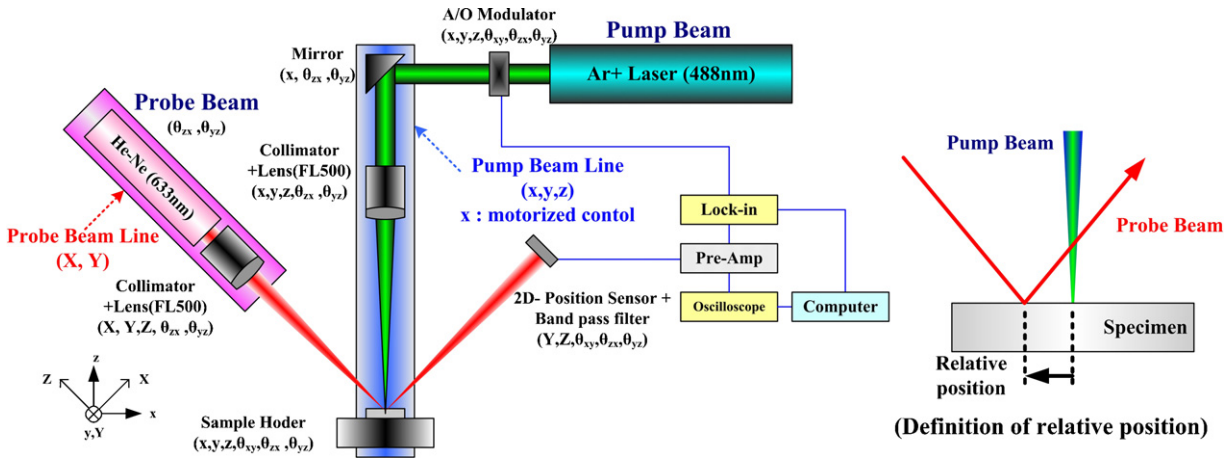

Fig. 2. Schematic diagram of the experimental setup.

son of the model for semi-infinite solids and the slab model [16,17]. This thickness was defined as the critical thickness.

Since the area of thermal diffusion varies with the length of thermal diffusion ($L_{th} = (\alpha/\pi f)^{1/2}$), the critical thickness is ascertained by varying the length of thermal diffusion. In this study, the range of the length of thermal diffusion is 100–400 μm. A length of 350 μm for thermal diffusion is used to investigate the effect of thickness. In case the sample thickness is greater than 12 mm, the result agrees with the curve for the semi-infinite solid model, with the relative error being less than 1%, as shown in Fig. 3. Accordingly, for this experiment, samples are prepared with a thickness exceeding 12 mm. Fig. 4 represents the amplitude of photothermal displacement signal for verifying of experimental setup and theoretical analysis, and it also shows that a good fit of the experimental result is obtained with the theoretical curve.

4.2. Experimental results

When the thermoelastic deformation in the sample surface is generated by the pump beam, we determine the thermal diffusivity through the value of the offset between the phase curves of the probe beam and the pump beam. The slope of the phase signal of the probe beam, with respect to the relative positions of the pump and probe beam, is quite sensitive to the thermal diffusivity of the sample. Even if the many parameters such as the probe beam, thermal diffusion length, modulation frequency and relative position which can influence the phase of the deflection angle exist, the modulation frequency and relative position have been known critical [7,8]. The measured and calculated phase difference curves are matched with each other in the region of between a quarter and three-quarters below the relative position of the minimum phase difference. The thermal diffusivity is determined when the standard deviation between the measured and calculated phase difference curves in Eq. (2) is minimized by using the bi-section method.

In the case of the slab model, thermal diffusivities are measured by using pure metal samples of 1 mm thickness. In the case of the model for semi-infinite solids, samples with thickness exceeding 12 mm are called semi-infinite solids, for experimentative purposes. The results, shown in Fig. 5, of the phase difference for semi-infinite solids confirm that experimental results agree very well with the theoretical analysis. The analysis is carried out under conditions wherein the frequencies of the modulated pump beam are 300 Hz and 400 Hz for pure copper, and 200 Hz and 300 Hz for nickel alloy (UNS N02201), respectively.

Table 1 shows the results on thermal diffusivity that are obtained by comparing the phase curve that is measured through experimentation with the curve that is obtained from theoretical analysis. The measured thermal diffusivities for semi-infinite materials agree with the values from the literature to within 1% percent of the standard deviation, while those for finite materials show about 3% [9,10].

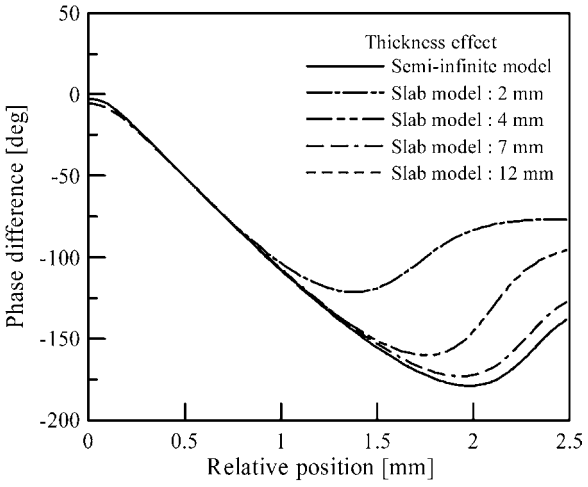

Fig. 3. Critical thickness for the semi-infinite solid model.

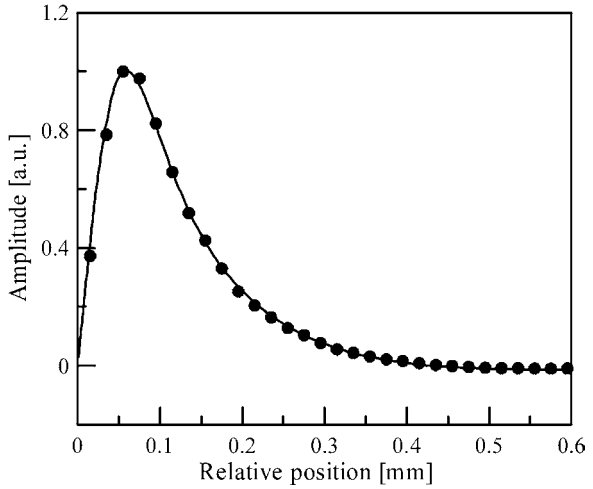

Fig. 4. Verification experiments for experimental setup and theoretical analysis.

Table 1
Comparison of thermal diffusivities from the literature [18,19] with those measured in the study (f: modulation frequency, L_{th}: thermal diffusion length, L: literature value, M: measured value).

Model	Material	f [Hz]	L_{th} [μm]	Thermal diffusivity (m^2/s) × 10^5		Relative error (%)
				L	M	
Slab	Silver	728	275	17.3	16.94	2.03
	Copper	763	220	11.6	11.13	4.05
	Zinc	185	275	4.4	4.26	3.18
	Iron	269	165	23	2.22	3.61
Serai-infinite	Nickel Alloy [UNSN02201]	200	176	1.947	1.96	0.69
		300	144	1.947	1,956	0,43
	Copper	300	351	11.6	11.656	0.48
		400	304	11.6	11.495	0.91

This result shows that the finite-thickness slab model yields relatively greater measurement error compared with the model for semi-infinite solids. Therefore, it can be said that the model for semi-infinite solids yields more accurate measures of thermal properties, as the method does not need to consider the thickness of samples.

Now, we compare the results of an analysis of uncertainty of the model for semi-infinite solids and the slab model with constant thickness. Through a parametric study of the slab model [17], it is verified that the sample thickness and the radius of the pump beam greatly impact upon the thermal diffusivity. On the other hand, in the model for semi-infinite solids, only the radius of the pump beam is the cause of error because sample thickness has no effect. The uncertainty is 1.7% in the slab model and 0.2% in the model for semi-infinite solids.

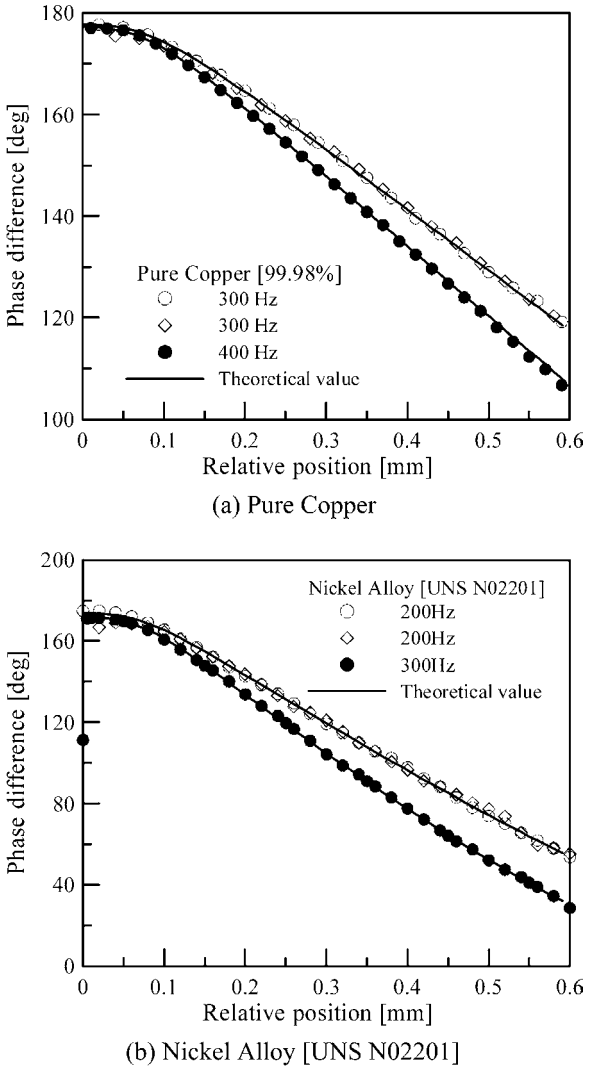

Fig. 5. Experimental results for the model for semi-infinite solids.

5. Conclusion

The photothermal displacement method is used for the quite accurate determination of the thermal diffusivity of materials, but most studies on the measurement of thermal diffusivity using the photothermal displacement method have considered samples with finite thickness. Since the thickness of samples causes many kinds of error in the measurement of thermal properties, a novel theoretical analysis and an associated experimental measurement, which are independent of sample thickness, were accomplished in this study. The main advantage of the approach, compared to classical methods, is that it can be applied on semi-infinite media, it is non-contact, and it only needs a small scanning spot on the sample surface. The contributions of the study are itemized below.

(1) Thermoelastic analysis of the sample surface has been conducted, using the results from thermal analysis for obtaining the gradient of the deformation that is caused by the photothermal effect. Thermal analyses are conducted using the two-dimensional cylindrical heat conduction equations and the Hankel transform. Then, the deformation gradient on the sample surface, through thermoelastic analysis, is obtained by applying the Hankel transform.
(2) The experiment for the sample of semi-infinite solids showed that thermal diffusivity can be accurately measured, i.e., to within 1% of the standard deviation compared with the values stated in the literature.
(3) An analysis of uncertainty for the model for semi-infinite solids and the slab model reveals that the former model is relatively more accurate. The reason is that in the model for semi-infinite solids, error factors such as the sample thickness and the assumption of adiabatic boundary conditions can both be excluded from consideration. Therefore, it is concluded that the model for semi-infinite solids suggested in this study will improve the reliability of measurement of thermal properties using the photothermal displacement method.

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