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Cross-plane Thermal Conductivity and Thermal Diffusivity Measurement in Thin-films by One-dimensional Thermal Wave Method

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Abstract—**A novel approach for cross-plane thermal conductivity and diffusivity measurement in thin-films by combining steady state comparative-longitudinal heat flow method (SCHF) and the Angström method is reported. A thin specimen was sandwiched between two identical metal cylinders; thermal wave was introduced from the lower cylinder and crossed through the specimen then passed to the upper cylinder where steady heat flux is enforced. There were four equally spaced thermocouples on both upper and lower cylinders to record the temperature change along the cylinder column. By solving the one-dimensional (1D) heat conduction equations, thermal diffusivity and thermal conductivity can be deduced simultaneously. Thermal diffusivity and thermal conductivity of Polytetrafluoroethylene (PTFE) films with different thickness were measured, and the results are in a good agreement with reported standard value.**

Keywords-thermal conductivity; thermal diffusivity; PTFE; SCHF; thermal wave method;

I. INTRODUCTION

The thermal conductivity (κ) and thermal diffusivity (α) of thin-films are important thermophysical properties for both industry applications and fundamental science. In general, thermal conductivity is measured by steady state conditions and thermal diffusivity is obtained by dynamic state measurements. The test methods including guarded-hot-plate method[1], the laser flash technique[2], hot wire technique[3], 3ω method[4], periodic heat flux method (such as Angström method)[5], *et al*. However, most of those methods can only obtain one thermophysical property (κ or α), while the other property is often calculated according to the relationship: $\alpha = \kappa/\rho C$, where ρ is the density, C is the specific heat. However, the calculated results possibly have large uncertainties because the values of ρ and C are often acquired from the literatures and are considered as constant. Actually, those values vary rather largely at different solid states (such as compacting materials and porous materials) and composition. It is attractive to develop new test methods that could simultaneously measure these thermophysical properties.

The steady state thermal measurement according to ASTM standard[6] is an effective method to measure cross-plane thermal conductivity. Several specimens (Such as Kapton[7], Nafion[8], porous metal[9], *et al*.) have been investigated by this technique. This method is also called steady state comparative-longitudinal heat flow method (SCHF), and the typical equipment mainly consists of two cylinders that are made of a standard material with known thermal conductivity.

In this paper, a modified SCHF method by introducing a periodic heat flux is employed for measuring thermal conductivity and thermal diffusivity simultaneously. The thermal conductivity can be obtained from the component of steady heat flux by the SCHF method, and the thermal diffusivity can be determined from the periodic heat flux according to the Angström method[5,10-11]. The thermophysical properties of PTFE films are measured to verify the feasibility of this method.

II. EXPERIMENTAL

A. Apparatus

As shown in Fig.1, two copper solid cylinders (length: 50mm; diameter: 20 mm) were placed vertically at where thin

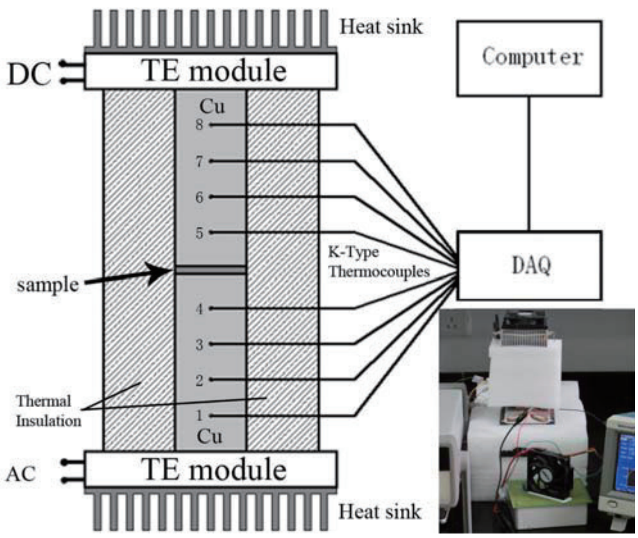

Figure1. Schematic and photography (insert) of experimental apparatus

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film sample was sandwiched between the upper rod and the lower rod. In order to ensure good thermal contacts between the thin film sample and two cylinders, a weight which generates 30 KPa pressure was subjected onto the top of the upper cylinder. In order to satisfy the 1D heat transport condition, cylinders are wrapped with multilayered thermal foams to minimize heat loss. Each cylinder equips four K-type thermocouples of which the spacing distance is 10 mm, and there are totally eight thermocouples (the sequence refers to Fig.1) to record the temperature changes along the cylinders. The bottom of upper cylinder and the top of lower cylinder are separately in contact with a thermoelectric (TE) module assembled with a fan cooler. The specimen with the same cross-section area of copper rods is sandwiched between cylinders in a symmetrical manner. High thermal conductivity grease (~ 6 $Wm^{-1}K^{-1}$) is used to ensure good thermal contact between cylinders and specimen.

In our experiment, the upper TE module was powered with a direct current (DC) source and served as a plate heater. A steady axial heat flux was injected and flowed from upper cylinder to lower cylinder. The lower TE module is connected with an alternating current (AC) source. When AC current signal is input to TE module, its surface can generate heat and draw heat alternatively. It can produce a periodic heat and generate almost harmonic temperature oscillation [10]. The thermal wave (in mHz) from lower TE module propagates across the cylinders and specimen and the thermocouples record the temperature changes.

B. *The measurement method*

Theoretical fundamentals for this experimental technique are described by 1D steady and transient heat conduction equations:

$$q = -\kappa A \frac{dT}{dx} \tag{1}$$

$$\frac{\partial^2 T}{\partial x^2} = \frac{1}{\alpha}\left(\frac{\partial T}{\partial t}\right) \tag{2}$$

where q is heat flux; A is the area; T is the temperature; x is the distance along the axial direction of cylinders; t is time. Here, heat loss is neglected to simplify the calculation. According to the Angström method, the cylinder is mathematically considered as a semi-infinite rod and the temperature at the end of rod varies in sinusoidal form with time as[11]:

$$T = T_0 + T_A \sin(\omega t + \varphi) \tag{3}$$

where T_0 is the initial temperature (or equilibrium temperature), T_A is the amplitude of temperature oscillation, φ is phase value. Then, the temperature distribution along cylinders can be deduced as[11]:

$$T(x,t) = T_0 + c_1 e^{-a_1 x} \sin(\omega t - b_1 x + \beta_1) \tag{4}$$

where c_1 and β_1 are constants, ω is angular frequency ($\omega=2\pi f$, f is frequency), a_1, b_1 are coefficient of temperature attenuation and coefficient of phase difference respectively and they are defined as:

$$a_1 = b_1 = \sqrt{\omega / 2\alpha} = \sqrt{\pi f / \alpha} \tag{5}$$

Comparing (3) and (4), it is found that a_1 and b_1 are slope in $\ln T_A$-x relationship and slope in φ-x relationship, respectively. Thus, if the amplitude decrement or phase delay along the specimen is known, the thermal diffusivity can be calculated according to (5).

Equation (4) also shows the nature logarithm of temperature amplitude and the phase value has linear change along the specimen. In our apparatus, the thermocouples record the temperature change of copper rods, so the amplitude and the phase value of specimen can be deduced by extrapolating the linear relationship in each cylinder. Then, given a specimen with thickness of L, the thermal diffusivity can be obtained from amplitude method:

$$\alpha = \pi f L^2 / [\ln(T_{A2} / T_{A1})]^2 \tag{6}$$

or from phase method:

$$\alpha = \pi f L^2 / (\varphi_1 - \varphi_2)^2 = \pi f L^2 / \Delta\varphi^2 \tag{7}$$

where T_{A1} and φ_1 are temperature amplitude and phase value on the upper specimen surface respectively, T_{A2} and φ_2 are temperature amplitude and phase value on the lower specimen surface respectively, $\Delta\varphi=\varphi_1-\varphi_2$ is the measured phase difference (or total phase difference) between specimen.

It is important to take the boundary-inducted interference, diffraction, and reflection of thermal wave in our considerations[12]. The reflection coefficient (R_T) of the thermal wave was defined as a ratio of amplitudes of reflected and incident waves is given as [5]:

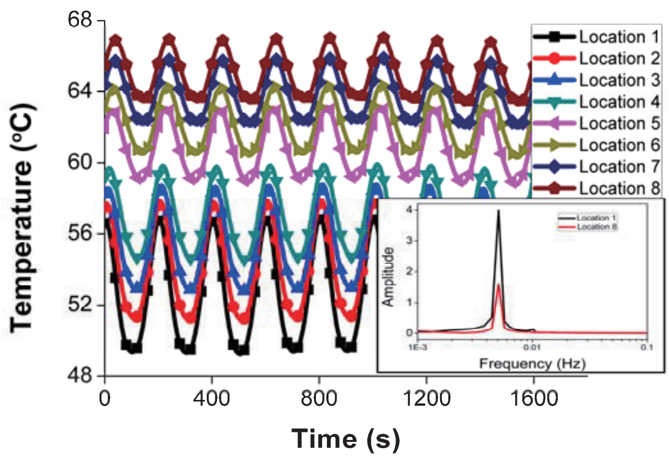

Figure 2. Temperature fluctuation at each thermocouple measuring location without specimen. Insert is FFT of temperature fluctuation at location 1. DC voltage of 8 V was placed on the upper TE module and AC voltage with peak-to-peak voltage of 10 V and frequency of 5 mHz was applied on the lower TE module.

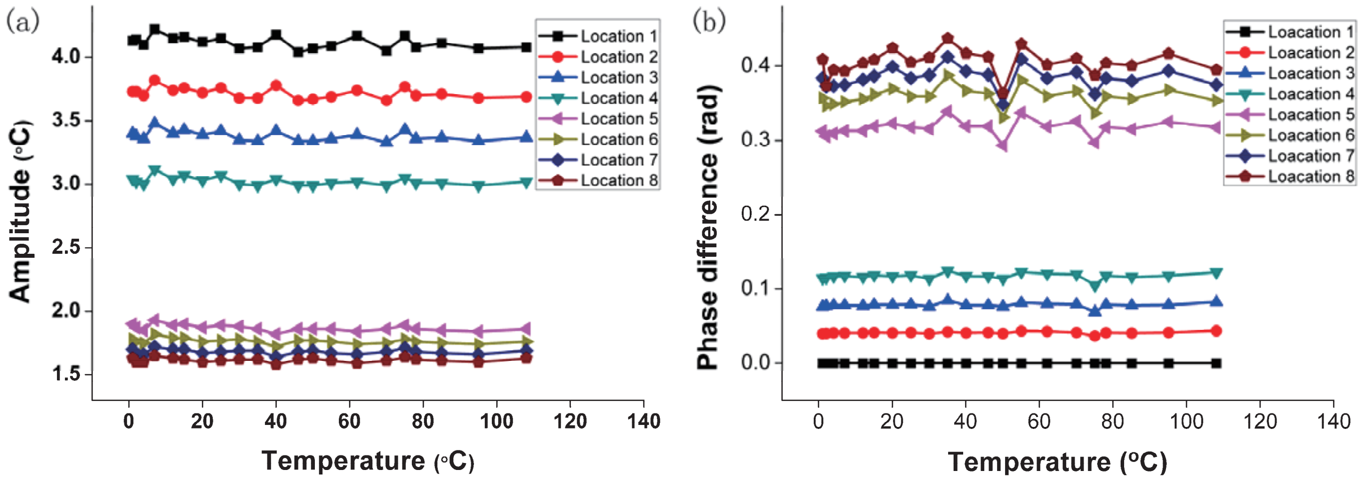

Figure 3. Tendency of (a) amplitude of temperature oscillation and (b) phase differenc under different temperature at the contact surface between upper TE module with cylinder. The temperature was controlled by varying the voltage of upper TE module. AC voltage with peak-to-peak voltage of 10 V and frequency of 5 mHz was applied on the lower TE module.

$$R_T = (\varepsilon_1 - \varepsilon_2)/(\varepsilon_1 + \varepsilon_2) \tag{8}$$

where ε_1 and ε_2 are thermal effusivities (ε) of adjacent media, $\varepsilon = \sqrt{\kappa\rho C}$. Take the ideal boundary of copper and PTET for example, the R_T is calculated at around 27% by using (8) and it may have strong impact on the amplitude of thermal wave. It indicates that the amplitude method may be not suitable in our method. However, for phase method, the boundaries and thermal grease also have influence on the actual phase delay on the specimen ($\Delta\varphi_s$). If considering the phase delayed by the boundaries and thermal greases as φ_c, the total phase difference $\Delta\varphi$ is the sum of $\Delta\varphi_s$ and φ_c. From (7), we can deduce the relationship between $\Delta\varphi$ and the effective thermal diffusivity of specimen (α_s) as:

$$\Delta\varphi = \varphi_c + \Delta\varphi_s = \varphi_c + L\sqrt{\omega/2\alpha_s} \tag{9}$$

Assuming specimens with different thicknesses are measured and φ_c is constant under the variety of L, κ_s can be expressed as the gradient of total phase difference $\Delta\varphi$ to L as:

$$\alpha_s = \omega/2[d\Delta\varphi/dL]^2 = \pi f/[d\Delta\varphi/dL]^2 \tag{10}$$

In SCHF method[6-9], a steady heat flux produces linear temperature change along the cylinder, so heat flux densities of the upper cylinder (q_1)and that of lower cylinder (q_2) may be computed with known values of thermal conductivity (κ_{Cu}=401 $Wm^{-1}K^{-1}$ at 300K [13]) and temperature gradient $(dT/dx)_{1,2}$ detected by thermocouples as:

$$q_{1,2} = -\kappa_{Cu} A (dT/dx)_{1,2} \tag{11}$$

The heat flux through the specimen (q_s) is evaluated by the average value of q_1 and q_2. The temperature drop $T_{s1}-T_{s2}$ (T_{s1} and T_{s2} are the temperature on the upper specimen surface and that on the lower specimen surface respectively) across the specimen also can be deduced by extrapolating the temperature gradient in each cylinder. Then, the total thermal resistance (R_t) is obtained as:

$$R_t = 2R_c + R_s = (T_{s1} - T_{s2})/(q_s A) \tag{12}$$

where R_c is contact thermal resistance between the specimen and rods, $R_s = L/\kappa_s A$ is thermal resistance of specimen, κ_s is effective thermal conductivity of specimen. Assuming specimens with different thickness are measured and R_c is constant under the variety of L, κ_s can be expressed as the gradient of total thermal resistance R_t to L as:

$$\kappa_s = 1/[(dR_t/dL)A] \tag{13}$$

To ensure φ_c and R_c constant, it is necessary to carefully control the coating thickness of thermal grease as constant.

As mentioned above, the upper TE module inputs a steady heat and the lower TE module generates a periodic heat in our apparatus. In other words, the upper TE module causes a steady temperature gradient and the lower TE module induces a thermal wave along the cylinders and specimen, and the thermal wave only produces temperature oscillation without changing the temperature gradient. The equilibrium temperature $T_0(x)$ in (4) is linear variation along the cylinder in this case. Therefore, the temperature detected by each thermocouple can be expressed in sinusoidal form as (3). T_0 is extracted as steady-state component of heat flux to determine the thermal conductivity by the SHCF method; T_A and φ are used as periodic component of heat flux to identify the thermal diffusivity. By combining the features of Angström method and SCHF method and measuring specimens with different thickness, the effective thermal diffusivity and thermal conductivity may be determined simultaneously according to (10) and (13).

III. Results and Discussion

Control tests without specimen were conducted to ensure accurate measurement. As shown in Fig.2, the temperature fluctuating situation at all measuring locations consisted well with sinusoidal form wave. FFT (inserted) analysis demonstrated that it had two main frequency signals at 5 mHz and 10 mHz of which the fundamental frequency (5 mHz) signal was dominated. The amplitude at 5 mHz was about 30

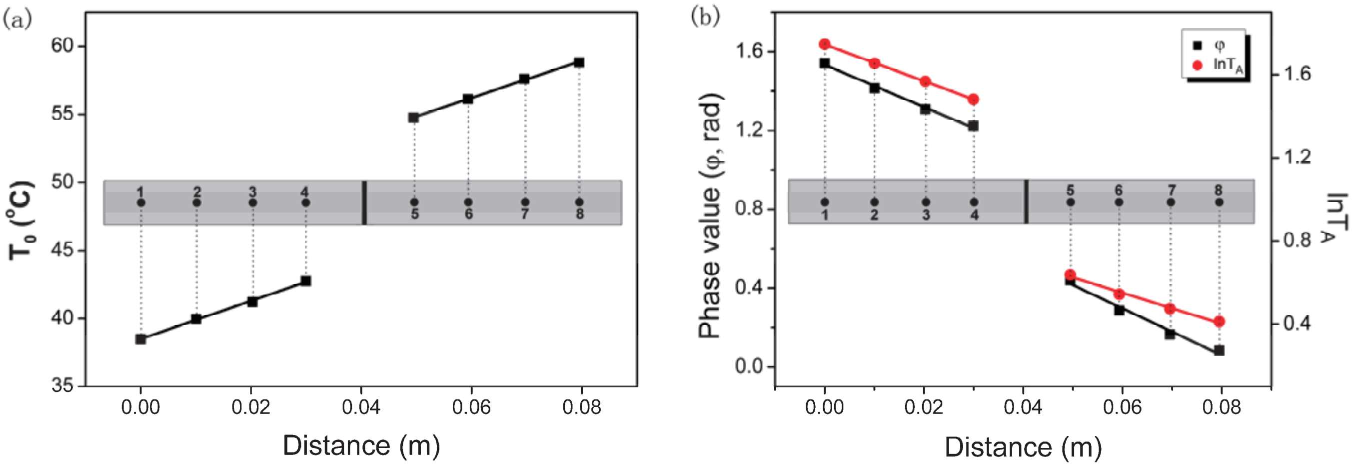

Figure 4. Distribution of (a) equilibrium temperature T_0 and (b) phase value φ and nature logrithm of temperature amplitude lnTA along the cylinders. Location 1 was considered as origin point. The specimen was PTFE with thickness of 112 μm. The upper TE module was applied by 10V DC voltage and served as plate heator. The lower TE module was input by AC voltage with peak-to-peak voltage of 10 V and frequency of 5 mHz.

times larger than that at 10 mHz. It indicates that AC current drives the TE module to output harmonic thermal wave with the same frequency and the fluctuation intensity decrease gradually along the propagation direction. The weak double frequency signal was generated by the Joul heat (I^2R, I is current, R is resistance) in TE module and it may be ignored in our method. However, the Joul heat effect can be eliminated by carefully adjusting the bias of input AC current[10].

In order to investigate the effect of heat flux generated by the upper TE module, we recorded the variation of amplitude and phase of temperature change by adjusting the applied voltage to the upper TE module without specimen. The value of amplitude and phase delay was extracted by fitting temperature fluctuation with equation (3). The results are shown in Fig.3. The phase differences were defined as the phase delays at each measuring point relative to location 1. As we can see from Fig.3, both the amplitude and phase differences at each thermocouple measurement point are almost maintained at a constant value at different temperature and the fluctuant features are not changed with different heat flux. It indicated the thermal wave motion is independent on the temperature gradient along the cylinder. The constant component in (4) was effective steady state parameter that could be used to evaluate specimen's thermal conductivity according to SCHF method, and the fluctuation component may be considered to determine the thermal diffusivity by Angström method.

The equilibrium temperature, temperature amplitude and phase difference were obtained by fitting temperature oscillation with (3). The slopes of their linear relationship along the copper rods were computed by linear least square method. A serious of specimens with different thickness was considered. Take 112-μm-thick PTFE film for example (shown in Fig.4), the average temperature (Fig.4a) was 54 ℃, the equilibrium temperature drop on PTFE films was 9.5 K and the amount of heat through this specimen was around 7.1 W, so the total thermal resistance was 1.34 K/W for 112-μm-thick PTFE film. The heat loss could also be evaluated at 2.61% by the formula $|q_1 - q_2|/q_1$. It showed our apparatus were well thermal isolated and the results were receivable.

From Fig.4b, the thermal diffusivity of copper rods could be evaluated; and the amplitude attenuation and the total phase delay induced by specimen might be extrapolated according to (4). In this case, the total phase delay is 0.54 rad, and the thermal diffusivity of upper rod and lower rod is calculated as

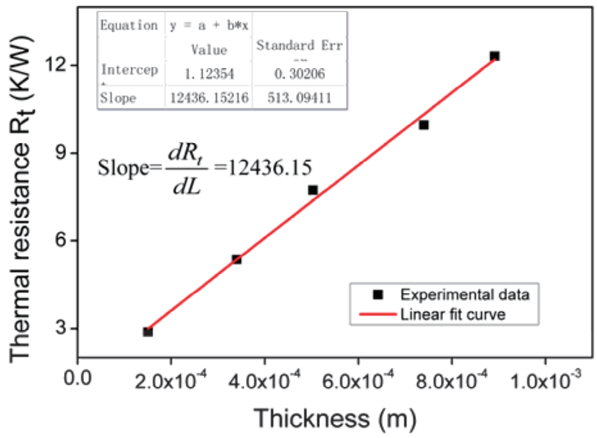

Figure 5. Thermal resistance vs. thickness of specimens

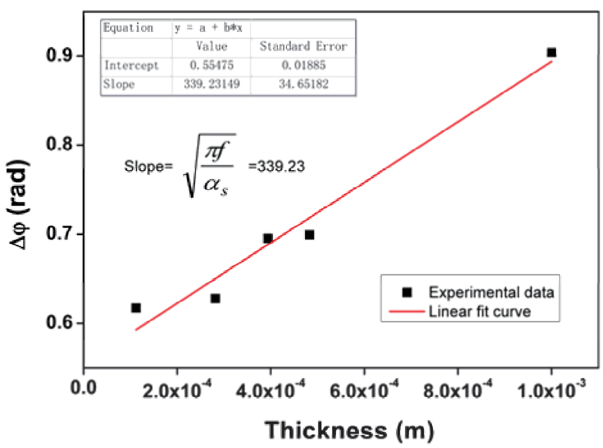

Figure 6. Phase difference vs. thickness of specimens

1.169×10^{-4} m^2/s and 1.117×10^{-4} m^2/s by phase values, while 2.04×10^{-4} m^2/s and 2.85×10^{-4} m^2/s by values of lnT_A. The results of copper thermal diffusivity from the phase method consisted well with the reference value of copper (1.14×10^{-4} m^2/s[14]) , but from the amplitude method the results were twice larger than the reference value. As shown in Fig.4b, the slope of lnT_A in upper and lower rods was smaller than corresponding slope of phase delay, but the slopes should be identical theoretically. The reason may be caused by the reflection of thermal wave at the boundaries, and the oscillation amplitude may be enhanced by the reflected wave that actually leads to temperature buildup in the lower cylinder while there is no influence in the phase delay. It indicates that only phase value is effective to evaluate the thermal diffusivity of specimens and the copper rods can be used as standard substance to examine the test accuracy.

The effective thermal conductivity and diffusivity of PTFE films were evaluated according to (10) and (13) by testing specimens with different thickness. The average temperature of specimens were in the range of 50 ℃-60℃. As shown in Fig.5 and Fig. 6, the thermal conductivity and diffusivity of PTFE films were 0.257 Wm^{-1}K^{-1} and 1.365×10^{-7} m^2/s.

IV. Conclusion

In this paper, we report the technique for measuring cross-plane thermal conductivity and diffusivity in thin-films with combining steady state comparative-longitudinal heat flow method (SCHF) and the Angström method. Thin specimens are sandwiched between two identical copper cylinders. A series of thermocouples detect the temperature fluctuation in copper rods. From the linear relationship between the temperature drop and the phase difference with the distance along the copper cylinders, the equilibrium temperature drop and the phase difference on specimens can be extrapolated. The thermal conductivity and thermal diffusivity are deduced from these temperature drop and phase difference on specimens, respectively. By the slopes of the total thermal resistance and total phase difference versus thickness of specimens, the effective thermal conductivity and diffusivity of the specimen can be obtained. PTFE films were tested and the effective thermal conductivity and diffusivity of PTEF films were determined as 0.257 Wm^{-1}K^{-1} and 1.365×10^{-7} m^2/s in the range of 50 ℃- 60 ℃.

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