MEASUREMENT OF TRANSPORT PROPERTIES OF THIN FILMS AND COATINGS

by

Andrej STANIMIROVIĆ and Kosta MAGLIĆ

Original scientific paper
UDC: 536.629.7=20
BIBLID: 0354–9836, *1* (1997), 2, 91–106

Free-standing and substrate-mounted thin films have been widely used for microelectronic and optical devices. In particular, CVD diamond, as a new material with ideal combination of properties (electrical isolator, excellent heat conductor, hardest known material) becomes important both in heat spreading and surface strengthening applications. Thermal barrier coatings are quickly gaining widest application in heat management applications.

In devices and systems design, demand for accurate thermophysical property functions of these materials, especially their thermal conductivity and thermal diffusivity, has been increasing. A variety of methods have been applied, modified and developed to measure transport properties of thin films and CVD diamond. Paper reviews methods of measuring thermal conductivity and thermal diffusivity of thin films and CVD diamond and discusses their advantages and disadvantages.

Measurements of thermal diffusivity of tantalum, tungsten and nickel thin films at our laboratory applying AC calorimetric technique are reported.

INTRODUCTION

Importance of thin films and coatings has increased substantially over the last decade. To mention but a few applications of thin films: CVD diamond layers as heat spreaders for integrated electronics or cutting die coatings, in optical components of high power laser applications, use of thin films as ceramic thermal barriers for thermally stressed mechanical parts (jet turbines, *e. g.*), *etc.* Degradation of device performance due to internally generated heat is becoming a limitation with increasing level of integration of microelectronic devices, as are increasingly severe conditions of optical device use, or stringent performance requirements in adverse environments in mechanical applications. Thermal characterisation of thin films or thin film multi-layered structures poses specific problems. Several techniques and their appropriateness for various types of applications will be reviewed in other sections of the text.

THIN FILMS/COATINGS

In thermal management applications, the most important property of a thin film/coating is its thermal conductivity. In principle it can be obtained by measuring heat flux, temperature gradients and sample geometry, but most frequently it is measured by determining experimentally thermal diffusivity and then computing thermal conductivity from diffusivity, density and specific heat, provided the latter two are known with sufficient accuracy. Thermal diffusivity and thermal conductivity are related by the well known equation $\lambda = a \cdot Cp \cdot \rho$, where λ is thermal conductivity, a is thermal diffusivity, Cp is isobaric specific heat and ρ is the density. As accurate determining of unidirectional thermal flux, either in longitudinal or radial direction tends to be a difficult task, especially with thin films and coatings, methods directed at measuring thermal conductivity are more an exception than a rule. Thermal diffusivity in turn may be determined from time-space dependence of specimen temperature under transient conditions, and temperature increase due to perturbation causing transient can be very small. It is therefore generally a lot more convenient to measure thermal diffusivity than thermal conductivity. Beside thermal conductivity and thermal diffusivity, properties of thin film/coating that may be of interest, depending on application, include thermal optical properties, crystallographic properties and hardness.

For the purpose of this paper, thin film is a free standing layer of a material with uniform thickness. A coating is one or several thin film layers adhering to a thick substrate. Depending on the material, thickness of the film or coating may be from 5 μm to 2 mm.

REVIEW ACCORDING TO PROPERTIES, MATERIALS AND MEASURING TECHNIQUES

PROPERTIES AND MEASURING TECHNIQUES

Techniques of measuring thermal transport properties can be broadly divided into stationary-state and transient techniques.

In stationary-state thermal conductivity techniques, specimen has to be held under adiabatic conditions in order to measure input heat flux accurately, and to ensure that this flux gives rise to corresponding temperature gradient, depending on thermal conductivity of the material.

In transient techniques, stationary temperature field in the specimen is perturbed and thermal diffusivity of the material determines the dynamics of relaxation. Conversely, thermal diffusivity of the material can be derived analysing the dynamics of the relaxation. In pulse techniques, heating is applied during a very short interval, and thermal diffusivity is determined from the history of either rear or front sample surface.

In thermal wave (periodic) techniques, the specimen is subjected to periodic heating which results in periodic changes of temperature along the specimen. Thermal diffusivity is determined from either amplitude decrement or phase shift of temperature waves propagating along the sample. Step (or monotonic) heating technique represents variation of the first technique, with a longer, or very long pulse duration. Analysing temperature history along the sample in the a quasi-stabilised regime of heating permits obtaining its thermal diffusivity. It should be noted that the step heating technique can also give thermal conductivity.

MATERIALS AND MEASURING TECHNIQUES

Depending on the film thickness and its application, thin layer of material can be either a free-standing film or a layer on a substrate. Free-standing films can be divided into many sub-categories, but metallic films (foils) and diamond films etc. are of primary interest for this presentation. Although thickness of diamond sample may be of the order of 1mm and may seem too large for a thin-film classification, it is nevertheless considered here. CVD grown diamond has multi-crystalline structure typical of other thin films. From the thermal management point of view, coatings can have two roles: to enhance heat conduction and spreading, or to impede heat flow. Diamond coatings serve the former purpose, while the latter is achieved by various thermal barrier coatings.

Free-standing diamond films of sufficiently large size can be measured by stationary-state techniques, giving thermal conductivity as a result. Free-standing thin metallic films can be measured only using thermal diffusivity techniques. Depending on application (heat spreading or thermal insulation), in-plane and/or perpendicular heat conductivity of a coating may be of interest. In measuring perpendicular heat conductivity (normal to surface), thermal diffusivity pulse, or high frequency thermal wave techniques prevail, due to the need for high spatial resolution of measurement. In-plane (parallel to surface) thermal diffusivity of a coating may be also measured using both groups of methods, provided diffusivity of the coating be significantly higher than diffusivity of the substrate.

TEMPERATURE RANGE AND MEASURING TECHNIQUES

Depending on application, thermal properties of interest may lie in near room temperature region or in high temperature region. In microelectronics, principal interest lies in the room temperature thermal conductivity. For thermal barriers on the other hand, thermal properties at high temperatures bear highest importance.

PROBLEMS IN THIN-FILM THERMAL CHARACTERISATION

Measurement of thermal transport properties of thin films and coatings presents challenges not encountered when measuring bulk properties with larger specimens. Due to morphological differences, thermal conductivity of a film may be significantly less than that of bulk material having the same nominal composition.

Additionally, thermal conductivity of thin film may exhibit anisotropy and inhomogeneity. The film may, and often has different in-plane and perpendicular thermal diffusivity and conductivity.

Stationary-state methods require adiabatic conditions which are more difficult to attain with thin film miniature specimens. Heat capacity of a thin film sample is extremely small, and even a very fine thermocouple (with 25 μm diameter wires) becomes major heat sink.

As the film gets thinner and its thermal conductance larger, interfacial thermal contact resistance between the film and the substrate becomes more important.

In order to derive thermal diffusivity of one of the two-layers, one has to know very accurately all properties of one layer and specific heat and density of the investigated layer.

When applying thermal wave technique to a thin film specimen, one has to make sure that the frequency of excitation be sufficiently high to avoid reflection from specimen boundaries. Conversely, the specimen has to be of sufficiently large dimensions.

Contact measurement of thin films' temperature using thermocouples is much more difficult than with the bulk material, and thermocouple attachment becomes more critical as the thickness of the film decreases. Cements of various sorts do not guarantee lasting, let alone thermally good contact. If the measurement is relative (what will be discussed later), poor thermocouple attachment can be tolerated.

Contactless (infrared) temperature detection solves the previous problem but introduces its own. Due to chromatic aberration, visible light travels differently (if at all) through IR optics, rendering positioning of the IR optics difficult.

When measuring thermal diffusivity, thermal conductivity information is obtained by computation. This makes knowing Cp and ρ mandatory, which may not be easily obtained – especially ρ in the case of thin films.

If the specimen material is transparent and/or translucent, its surfaces have to be coated to make measurement meaningful. At the same time, this coating may affect the measurement and alter the results.

TECHNIQUES

STATIONARY-STATE TECHNIQUES

Axial heat flow method [1] is a steady state technique used for bulk thermal conductivity measurement, which is directly applicable to CVD diamond specimens (slabs) as well as free-standing thin films provided they are of appropriate size. In this method one end of a bar-shaped specimen is heated by a wound heater, and the other is kept at approximately constant temperature. The temperature profile along the specimen is determined using several thermocouples. A more elegant technique involves formation of a graphitic microheater on the surface of an end of a diamond sample [2]. Namely, using a Nd:YAG laser it is possible to "scribe" on the electrically isolating diamond surface, locally converting diamond into graphite and thus producing a microresistor with well-defined and miniature geometry. Golden leads are then bonded to contact pads to pass electrical current and heat the sample. This convenience has been implemented in several techniques of thermal conductivity and diffusivity measurement: in the axial heat flow (thermal conductivity) method, and in converging wave and modulated thermal wave thermal diffusivity methods.

For coatings, several attempts have been made at measuring thermal conductivity/diffusivity using resistive heaters and temperature sensors deposited on the film. Cahill's DC technique [3] is based on two parallel metallic strips deposited on the surface of the sample. When a current pulse is passed along one strip it acts both as a heater and also as an electroresistive temperature sensor. The other strip acts solely as an electroresistive temperature sensor at some distance from the first. Knowing the heat supplied, temperatures of strips and their separation, one can directly compute thermal conductivity. Chief difficulty arises from strip/sample adhesion problems, which worsen with increasing temperature and in adverse environments.

NONSTATIONARY-STATE TECHNIQUES

Rear-surface flash technique

The flash method of determining thermal diffusivity described first by Parker *et al.* in 1961 [4] relates temperature rise of the rear sample surface, whose front surface has been irradiated by a short burst of light energy. Its thermal diffusivity, a, is determined from the expression: $a = 0.139 \cdot L^2/t_{1/2}$, where L is the sample thickness, and $t_{1/2}$ the time for the rear surface temperature to reach half of its maximum value [5]. Later developments of this method take into account departures of real experiment from the model and analyse the whole rising curve as well as the significant part the sample temperature decay curve, contributing to better accuracy throughout the widest temperature range. These developments are critically reviewed in [6, 7].

Attempt to measure thermal diffusivity of a thin film using rear-surface flash technique, however, poses significant difficulties [8]. Experimental equipment has to provide very short pulse time and commensurably fast IR detector, sample thickness, also, has to be measured with high accuracy. Usually Q-switched lasers have to be utilised with pulse duration of the order of 1 μs. Q-switching degrades laser pulse energy, but this is of only minor concern. More serious is the likelihood of damaging sample surface as irradiation energy is very densely packed. Suitable IR detectors are liquid nitrogen cooled InSb and HgCdTe above 0 °C, and PbS above 250 °C. Typical temperature rise times of 1ms dictate data acquisition rates of the order of 1Msamples/s.

The sample size and preparation also become critical. With such thin samples, their transparency and/or translucency in the visible or infrared (IR) wavelength region become commonplace, of special concern being the possibility of direct laser irradiation of IR detector, which can lead to its permanent damage. Although methods to account in mathematical model for transparency/translucency effects have been devised, coating of the sample is a more common way of dealing with these problems. To prevent laser irradiation from penetrating beyond the front surface of a transparent sample and also from irradiating the detector from within the sample, the sample is conveniently coated with gold having very low emissivity and absorptivity. To intensify light absorption on the front surface and IR radiation on the rear surface, a second layer of boron or titanium is added. This imposes a need however to consider whether significant thermal resistances have been introduced in this way in series with the sample, particularly if the sample is thin itself, and of high diffusivity.

Original Parker's method is applicable only to isotropic and homogeneous materials. Several variants of the technique have been described which were developed for measuring both in-plane (parallel to sample surface) [9–12] and perpendicular (through the sample plane) [13] thermal diffusivities. Ohta *et al.* [14] recently reported a variant of line heating method for deriving data on both in-plane and perpendicular thermal diffusivities using the sample in the form of film or ribbon, illuminated with a pulsed laser. This line is flashed onto the sample and temperature variation is observed at the rear surface using IR detector. By changing the offset between the line and the detector it is possible to obtain data on both directional thermal diffusivities.

Front surface flash (back-scatter) techniques

As their name states, front surface or back-scatter variants of the pulse technique derive thermal diffusivity from the dynamics of cooling of the surface irradiated by the energy pulse. There are several advantages peculiar to front-surface flash variants which make them suitable for thin film/CVD diamond thermal diffusivity measurements. Only one side of the specimen need be accessible, temperature excursions are significantly larger than of the rear surface and the technique is more sensitive to near surface (coating) thermal characteristics than to those of the bulk (substrate).

"Classical" front surface flash technique was extensively studied by Leung and Tam [15]. They have applied the method to a wide variety of materials. As for thin films, they have made an extensive mathematical analysis considering sample shaped as a thin slab and as an infinite solid. They have also found radiation losses and two-dimensional effects to be negligible when measurement is performed at room temperature.

Converging wave technique

With an aim of measuring radial (in plane) thermal diffusivity, Cielo *et al.* [16] developed a front surface flash method involving an annular convergent temperature wave. An annular shaped laser pulse is applied to the sample surface, while the temperature change at the centre of the annulus is monitored using an IR detector. Propagation of the thermal wave in the sample has been studied employing a finite difference numerical model taking into account radiative and convective heat losses. Method also permits heating of the annular region sinusoidally and measuring sample thermal diffusivity on the basis of phase delay between temperature variations in the annulus and at the centre.

Many experimental techniques basically similar to front surface flash method have been developed in recent years to measure thermal properties of thin films and CVD diamond. Among these, transient thermoreflectance [17] and transient surface thermal grating [18] methods appear to be suitable for characterizing thin film materials in situ.

Picosecond thermoreflectance

The surface of the thin film is heated by a picosecond pulse. The chosen wavelength is such that heating laser pulse be absorbed in a thin layer of a few tens of nanometers from the thin film surface, establishing thus a steep temperature distribution in a microscopic distance as the initial condition. The heat diffuses toward the inside of the thin film and the temperature of the film surface decreases in a time scale of a picosecond to nanosecond. Temperature decreases faster in more thermally diffusive materials. Observation of temperature change on a picosecond time scale is achieved by measuring the change of reflectance by probe picosecond laser pulses. For typical metals reflectivity changes are of the order of 10^{-5} K^{-1}. The delay of a probe laser pulse after the pump laser pulse is achieved by splitting the laser beam in two and changing the length of optical path the probe beam traverses between the picosecond laser source and thin film specimen. A difference of optical length of 1 mm corresponds to 3.33 ps delay of probe laser pulse. By changing the delay the whole temperature decay signal is recreated.

Transient surface thermal grating

Transient thermal grating is also a contactless technique, which uses two interfering laser beams to form an interference pattern on the thin film surface. This pattern heats the surface unevenly and produces a transient thermal image on it. There are two methods of detection of transient thermal effects: IR detection measures thermal image diffusion and laser beam probing measures surface displacement resulting from heating.

For IR detection [19] the thin film surface is coated with titanium, enhancing thus both heat absorption and IR emission. The temperature profile of the grating is then monitored in reference to position and time using a high resolution IR detector. The exponential time decay of the grating is related inversely to the diffusivity in the plane parallel to the surface.

Localised heating applied by Jauregui and Matthias [18] produces undulations of the surface caused by thermal expansion. Degree of thermal expansion, which is related to thermal diffusivity, can be detected by a probe laser beam reflected from the surface of the sample. The deflection of the probe beam is detected by a detector array.

THERMAL WAVE TECHNIQUES

Modified Angstrom's techniques

In 1861 Angstrom conducted his historical experiments to measure thermal diffusivity of copper and iron [20]. Samples were long metal bars with one end enclosed in a chamber through which he alternatively passed cold water and steam. Several thermometers were mounted in wells along the bar. Decay of temperature variations along the bar enabled Angstrom to determine thermal diffusivities of samples and, using known specific heats and densities, to calculate their thermal conductivities. Angstrom's original concept has been modified considerably since then as reviewed by Phylippov [21], including also range of very high temperatures reviewed by De Coninck and Peletsky [22].

In applying temperature wave method to thin film samples both modulated heat supply and temperature wave detection have been attempted in a variety of ways, giving rise to a multitude of techniques collectively referred to as modulated thermal wave techniques. Techniques reviewed in the following paragraphs are named after some distinctive feature peculiar to each respective variant, other aspects may be shared with another variant or variants. For example, photoacoustic technique owes its name to the particular method of detecting temperature waves in the sample. Modulated heat, on the other hand, can be supplied in various ways, using *e. g.* laser light, halogen lamp, microheaters or concentrated sunlight.

AC calorimetry

This is a modern version of Angstrom's technique [23-32]. It does not require a complicated measurement system and provides reliable results of in-plane thermal diffusivity. It has been shown recently that the method is applicable to two-layered specimens also [23].

A part of a rectangular thin film sample, either a free standing film or coating on the substrate is subjected to sinusoidally modulated light irradiation. The source is usually a laser with a beam scanned transversely across a portion of the sample surface, the temperature waves emanating from this irradiated region; when a halogen lamp is used part of sample surface is covered by a movable mask, the uncovered region being the source of thermal or temperature waves. These thermal waves propagate along the sample, and a fine thermocouple is used to pick up temperature signal at some distance from the irradiated region. A characteristic of the method is that position of the specimen's irradiated part changes relatively to the temperature sensor whose position is kept constant.

By analysing the decay curve of the temperature waves, one can determine thermal diffusivity. If measurements are made in vacuum, i.e. in absence of convective and gas conduction heat losses, information about thermal diffusivity can be derived from either wave amplitude decay or phase delay along the sample. Otherwise, to compensate for heat losses, one has to measure and incorporate both in thermal diffusivity calculation [29].

Photothermal radiometry

Photothermal radiometry [33] is a variant of AC calorimetry where an IR detector is used to pick up information on thermal waves propagating along the specimen. The technique is entirely contactless.

Thermal wave interferometry

This method is based on interference of two thermal waves: the one carrying incoming heat supply (usually laser light radiation), and the thermal wave reflected from the opposite surface of a thin sample, or a coating-substrate boundary. Interference of these two waves generates thermal image on the irradiated surface which is picked up by an IR detector. Measurements reported by Almond *et al.* [34] and Frederikse *et al.* [35] have been primarily directed at measuring thermal diffusivities of coatings. A modulated argon ion laser light was used as modulated heat source, and the thermal emission from the coating surface was detected by an IR detector and fed to lock-in amplifier referenced to modulation frequency. Aamodt *et. al* [36] have reported the work on a technique named time-resolved infrared radiometry. They applied stepped heat pulse to a coated sample surface and recorded the resulting temperature change as a function of time. Characteristic times and amplitudes can be attributed to internal thermal reflections that return heat to the surface. Characteristic times depend on layer thickness and thermal diffusivity - one can be determined when the other is known. The amplitude provides information on the thermal mismatch (contact resistance) between the layers. The method has been used to study disbonding of zirconia thermal barrier coatings.

Photoacoustic method

When a thin film sample contained in a gas-tight cell is periodically heated by a modulated light source, periodic heat flow from the sample causes a temperature variation in a thin layer of gas at the gas-sample boundary. The resulting piston-like behaviour of this layer induces periodic overpressure of gas in the cell which is detected as a sound, this phenomenon being called the photoacoustic effect. The photoacoustic signal (its amplitude and phase picked up by a microphone), contains information about thermal conductivity and diffusivity of the sample. Measurement of photoacoustic signal as a function of modulation frequency of the incident light enables one to calculate thermal conductivity and diffusivity of thin film material [37].

Photothermal deflection – mirage effect

When a thin sample is held in gas (*e.g.* air) and subjected to periodic heating, part of the heat is transferred to a thin layer of surrounding gas which expands upon heating. If a laser light beam passes through this layer its path is slightly distorted or bent (well known mirage effect) and the amount of distortion is proportional to the temperature of the gas, and in turn, of the sample [38]. This is yet another way of detecting thermal waves in a thin film sample. The distortion of laser beam path can easily be detected using na array of photodetectors.

3 ω method

This simple thermal wave method has been developed by Cahill *et al.* [39]. A line heater/resistance thermometer about 1mm long and 5 μm wide is deposited on an electrically non-conducting film or coating. An ac current of angular frequency ω is passed along the heater producing a temperature wave of frequency 2 ω. The temperature wave diffuses into the sample radially and its amplitude decays exponentially with radial co-ordinate. The amplitude of the wave is inversely proportional to the conductivity of the sample and to the logarithm of the reciprocal angular frequency. The signal at 3 ω, measured with a lock-in amplifier, is proportional to AC temperature in the specimen. By measuring amplitude as a function of angular frequency some distance from the heater one can determine sample material thermal conductivity without the need to know its specific heat. Depending on the frequency and thermal diffusivity, the method can probe the sample to the depth between 10 and 1000 μm.

IMPLEMENTATION CONSIDERATIONS

A significant international effort has been invested within this decade to enable accurate determination of thermal transport properties of thin films, coatings and CVD diamond, primarily for application needs. In spite of multitude of available methods reviewed in foregoing, it is not possible to establish those which give reliable results for particular groups of materials or sample geometries. So far two inter-laboratory (round-robin) measurement campaigns on thermophysical characterisation of thin films, coatings and CVD diamond have been carried out [40, 41]. The results of the second round-robin were analysed at the 13th Symposium on Thermophysical Properties in Boulder, Colorado in June 1997 [41]. This analysis showed that certain methods and variants tend to group results (mainly stationary techniques), but it also showed that differences between particular groups are significant, especially with thermal wave techniques. It indicates clearly that difficulties in measuring materials from the group of thin films, coatings and CVD diamond outlined in paragraph 4, can not be overcame easily. Research of measurement methods for thermophysical characterisation of thin films, coatings and CVD diamond stays therefore as the primary interest for international thermophysics community.

Before starting research on thin films/CVD layers for application needs of the VINČA Institute, facilities in our thermophysics laboratory included laser pulse apparatus for thermal diffusivity measurement [6] which has been in use since late sixties. This apparatus employs a ruby laser with maximum energy of 30 J per pulse of about 1 ms duration. Retrofit of this apparatus to accommodate thin film specimens would, however, ask for substantial investments. Although sufficiently fast IR detector is available and is used in measurements routinely, this millisecond-pulse laser would have to be Q-switched to shorten the pulse into microsecond range. Moreover, the presently used data acquisition system is inappropriate for this application. Its maximum sampling rate is approximately 40 kSamples/s, and in thin film measurements a data acquisition system with sampling rates of the order of 1 MSamples/s would have to be employed. All this, and additional effort to set up the new apparatus adds up to a considerable amount of time and equipment investment. Also, this equipment would allow thermal diffusivity measurement transverse to the layer only, while information most frequently sought is in-plane property.

Modified Angstrom's method on the other hand is comparatively simple for experimental implementation, and permits relatively quick setting up of the apparatus and its subsequent use. The property measured is in-plane thermal diffusivity, which characterises efficiency of heat spreading in the direction parallel to the surface, orthogonal to crystal growth, which is the primary objective of our research. In selecting the method for thin film transport properties measurement at our lab, preference was therefore given to the modified Angstrom's method.

APPARATUS AND MEASUREMENT TECHNIQUE

An experimental setup made for measurements at near-ambient temperature using the technique of AC calorimetry [30] is shown schematically in Fig. 1.

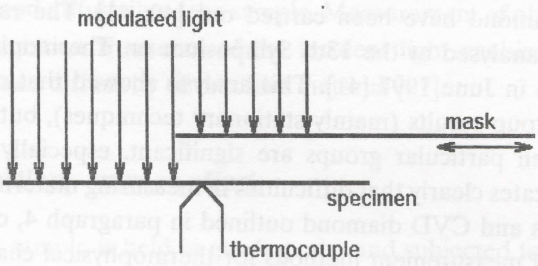

Figure 1. Schematic view of AC calorimetric measurement of thermal diffusivity

In initial experiments, a halogen lamp as a heat source was employed for irradiation of a part of the specimen surface. Modulation of the source has been generally accomplished by controlling the output current of a stabilised power supply using a low-frequency generator, operating at frequencies ranging between 0.25 and 1 Hz. In later experiments which applied to very thin foils (gold), a low-power continuous-wave laser was used. Periodical variations of temperature along the specimen were detected by a 25 or 50 μm diameter K-type thermocouple. Thermocouple was either spot-welded to the specimen surface or attached to it using silver paste, depending on the thickness and hardness of specimen material. In order to enable phase delay (shift) measurement, variation of incident light energy has been recorded using a photodetector.

Signal attenuation and the phase shift in measurements with different distances between the heated zone and the temperature detector were obtained by performing experiments with a mask shielding incident light in different positions relative to the sensor. Both modulated incident irradiation signal from the photodetector and the signal from the thermocouple were processed by a PC-based data acquisition system. Since lock-in amplifier was not available, all data and signal processing had to be done off-line, subsequent to the acquisition of signals. Measurements being made in air with heat losses present, the latter had to be accounted for using the technique proposed by Gu *et al.* [29]. This method requires information both on the amplitude decrement and the phase shift of temperature waves.

When the halogen lamp has been replaced by an on-off modulated low power HeNe laser (with a vibrating galvanometer mirror to generate irradiation area in a form of a strip), this modification eliminated unwanted specimen heating during the measurement. This arrangement was, however, limited to thinnest metallic foils. Then the halogen lamp could be used to heat the whole specimen in a controlled fashion, enabling measurements in the range between room temperature and 100 °C. Experimental results obtained for tantalum, nickel and tungsten foils at room temperature [42,43] were in agreement with recommended data for the bulk of these three metals [44] within few percent, as shown in the following table.

Table 1. Thermal diffusivity of Ta, Ni and W at room temperature

Material	Measured data [42,43] [$cm^2 s^{-1}$]	Recommended data [44] [$cm^2 s^{-1}$]
Tantalum, Ta	0.25	0.247
Nickel, Ni	0.66	0.662
Tungsten, W	0.25	0.235

DISCUSSION

Sample type, size, temperature and atmosphere around the specimen suggest the most suitable measurement method, along with desired accuracy of measurement and the necessary data which have to be already known. To a free standing thin film sample all the techniques described can in principle be applied.

Of all the techniques of measuring thermal diffusivity reviewed in foregoing, rear surface flash technique is by far the most established. However, as the heat pulse travels through the thickness of the sample, for measuring thermal diffusivity of a coating on a substrate with any degree of accuracy, the thickness of this layer must be known with high accuracy and thermal resistance of the coating must be significant. Temperature excursion within this thin layer may also be significant. All this restricts usefulness of the technique.

The most convenient way of determining thermal diffusivity of a coating is to use a modulated thermal wave technique. If the thickness of the coating is known accurately, the method of choice would be thermal wave interferometry. However, if the boundary is irregular or the thickness is uneven, some front surface flash method may be more appropriate.

In terms of apparatus complexity, the simplest method is AC calorimetry. Applicability of the technique to two-layer samples has recently been established. Accuracy of this method is under evaluation in most recent round-robin measurements conducted world-wide, in comparison to rear surface laser flash technique and the heated bar steady state technique.

ACKNOWLEDGMENT

Reported research is a part of scientific programme of the Ministry of Science and Technology of Serbia. Support is gratefully acknowledged.

REFERENCES

[1] Laubitz, M. J., Axial Heat Flow Methods of Measuring Thermal Conductivity, in *Compendium of Thermophysical Property Measurement Methods, Vol. 1: Survey of Measurement Techniques* (Eds. K. D., Maglić, A. Cezairlyan, V. E. Peletsky), Plenum Press: New York and London, 1984, pp. 11–60

[2] Worner, E. , Wild, C., Sebert, W. M., Koidl, P., Thermal Conductivity Measurements on CVD Diamond Using Graphitic Microheaters Prepared by Laser Scribing, presented at 13. Symposium on Thermophysical Properties, Boulder, 1997

[3] Cahill,D. G., Fischer, H. E., Klitsner, T., Swarts, E. T., Pohl, R. O., *J. Vac. Sci. Technol. A, Vac. Surf. Films 7* (1989), 3, pp. 1259–1266

[4] Parker, W. J., Jenkins, R. J., Butler, C. P., Abbott, G. L., Flash Method of Determining Thermal Diffusivity, Heat Capacity and Thermal Conductivity, *J. Appl. Phys. 32* (1961), 9, p. 1979

[5] ***Standard Test Method for Thermal Diffusivity of Solids by the Flash Method, *ASTM Standard E* 1461–92, (1992)

[6] Taylor, R. E., Maglić, K. D., Pulse Method for Thermal Diffusivity Measurement, in *Compendium of Thermophysical Property Measurement Methods, Vol. 1: Survey of Measurement Techniques,* (Eds., K. D. Maglić, A. Cezairlyan, V. E. Peletsky), Plenum Press: New York and London, 1984, pp. 305–336

[7] Maglić, K. D., Taylor, R. E., The Apparatus for Thermal Diffusivity Measurement by the Laser Pulse Method, in *Compendium of Thermophysical Property Measurement Methods, Vol. 2: Recommended Measurement Techniques and Practices*, (Eds. K. D. Maglić, A. Cezairlyan, V. E. Peletsky), Plenum Press: New York and London, 1992, pp. 281–314

[8] Remy, B., Maillet, D., Andre, S., Laser Flash Diffusivity Measurement of Diamond Films: Some Experimental Specificities, presented at 13. Symposium on Thermophysical Properties, Boulder, 1997

[9] Donaldson, A. B., Radial Conduction Effects in the Pulse Method of Measuring Thermal Diffusivity, *J. Appl. Phys. 43* (1972), pp. 4226–4228

[10] Chu, F. I., Taylor, R. E., Donaldson, A. B., Thermal Diffusivity Measurements at High Temperatures by the Radial Flash Method, *J. Appl. Phys. 51* (1980), pp. 336–341

[11] Amazouz, M., Moyne C., Degiovanni, A., Measurement of Thermal Diffusivity of Anisotropic Materials, *High Temp. High Pressures, 19*, (1987), pp. 37–41

[12] Batsale, J. C., Degiovanni, A., Extension de la méthode "flash" à deux cas particuliers: Les matériaux anisotropes et les liquides, *Proc. Recontre Société Française des Thermiciens 88*, Limoges, CPM-14-1, 1988

[13] Chu, F. I., Taylor, R. E., Donaldson, A. B., Flash Diffusivity Measurements at High Temperatures by the Axial Heat Flow Method, in: *Proc. Seventh Symposium on Thermophysical Properties, ASME 1977* (Ed. A. Cezairlyan), ASME, New York, 1977, pp. 148–154

[14] Ohta, H., Shibata, H., Waseda, Y., *Rev. Sci. Instrum, 60* (1989), 3, pp. 317–321

[15] Leung, W. P., Tam, A. C., *J. Appl. Phys. 56* (1984), 1, pp. 153–161
[16] Cielo, P., *J. Appl. Phys. 56* (1984), 1, pp. 230–234
[17] Taketoshi, N., Baba,T., A. Ono, Development of a Thermal Diffusivity Measurement Sysytem With a Picosecond Thermoreflectance Technique, *High Temp. – High Press., 29* (1997), pp. 59–66
[18] Jauregui, J., Matthias, E., *J. Appl. Phys. A, Solids Surf., 54* (1992), pp. 35–39
[19] Graebner, J. E., Jin, S., Kammlott, G. W., Herb, J. A., Gardiner, C. F., *Nature*, (1992), p. 658
[20] Angstrom, A. J., *Ann. Phys. Chem. (Pogg. Ann.)* 114 (1861), pp. 513–530
[21] Phylippov, L. P., Temperature Wave Techniques, in *Compendium of Thermophysical Property Measurement Methods, Vol. 1: Survey of Measurement Techniques,* (Eds., K. D. Maglić, A. Cezairlyan, V. E. Peletsky), Plenum Press: New York and London, 1984, pp. 337–366
[22] De Coninck, R., Peletsky, V. E., Electron Bombardment Modulated Heat Input Method, in *Compendium of Thermophysical Property Measurement Methods, 1: Survey of Measurement Techniques (Eds. K. D.* Maglić, A. Cezairlyan, V. E. Peletsky), Plenum Press: New York and London, 1984, pp. 367–429
[23] Gu, Y. Q., Yu, L. X., Hatta, I., Thermal Diffusivity of Diamond/Si Composite Films Measured by an AC Calorimetric Method. II. Characterization of Deposited Diamond Layers, *Int. J. Thermophys, Vol. 18* (1997), 2, pp. 525–534
[24] Hatta, I., Gu, Y. X., Yu, L. X., Thermal Diffusivity of Diamond/Si Composite FilmsMeasured by an AC Calorimetric Method. I. Edge Effects, *Int. J. Thermophys, 18* (1997), 2, pp. 515–524
[25] Maesono, A., Tye, R. P., Gardner, R. L., Methods to Measure Relevant Thermal Characteristics of Thin Films of CVD Diamond and Other Highly Conducting Materials, *High Temp., High Pressures, Vol. 25* (1993), pp. 329–336
[26] Hatta, I., Kato, R., Maesono, A., Development of AC Calorimetric Method for Thermal Diffusivity Measurement I. Contribution of Thermocouple Attachment in a Thin Sample, *Jpn. J. Appl. Phys., 25* (1986), 6, pp. L493-L495
[27] Hatta, I., Kato, R., Maesono, A., Development of AC Calorimetric Method for Thermal Diffusivity Measurement I. Sample Dimension Required for the Measurement, *Jpn. J. Appl. Phys., 26* (1987), 3, pp. 475–478
[28] Gu, Y., Hatta, I., Effect of Edge Sample in AC Calorimetric Method for Measuring Thermal Diffusivity of Thin Films with High Thermal Diffusivity, *Jpn. J. Appl. Phys., 30,* (1991), 5, pp. 1137–1138
[29] Gu, Y., Tang, X., Xu, Y., Hatta, I., Ingenious Method for Elliminating Effects of Heat Loss in Measurements of Thermal Diffusivity by AC Calorimetric Method, *Jpn. J. Appl. Phys., 32*, Pt. 2 (1993), 9B, pp. L1365–L1367
[30] Hatta, I., Sasuga, Y., Kato, R., Maesono, A., Thermal Diffusivity Measurements of Thin Films by Means of an AC Calorimetric Method, *Rev. Sci. Instr., 56* (1985), 8, pp. 1643–1647
[31] Kato, R., Maesono, A., Hatta, I., Measurement of the Thermal Diffusivity of Films Using Advanced AC Calorimetric Method, *Proceedings,* Fourth Asian Thermophys. Properties Conference, Tokyo, 1995
[32] Kato, R., Maesono, A., Hatta, I., Development of AC Calorimetric Method for Thermal Diffusivity Measurement, V. Modulated Laser Beam Irradiation, *Jpn. J. Appl. Phys., 32*, Pt. 1 (1993), 8, pp. 3656–3658
[33] Feldman, A., N., Balzaretti, M., A Modification of Angstrom's Method that Employs Photothermal Radiometry to Measure Thermal Diffusivity: Application to CVD Diamond, presented at 13. Symposium on Thermophysical Properties, Boulder, 1997
[34] Almond, D. P., Patel, P. M., Pickup, I. M., Reiter, H., *NDT Int., 18* (1985),1, pp. 17–24
[35] Frederikse, H. P. R., Ying, X. T., Feldman, A., *Proceedings,* Materials Research Society Symposium, *142* (1989), pp. 289–294
[36] Aamodt, L. C., Spicer, J. W. M., Murphy, J. C., *J. Appl. Phys. 68* (1990),12, pp. 6087–6098
[37] Akabori, M., Sawajiri, O., Nagasaka, Y., Thermal Diffusivity of Thin Films on Substrate by the Photoacoustic Method, presented at 13. European Conference of Thermophysical Properties, Lisboa, 1993
[38] Bertolotti, M., Liakhou, G. L., Voti, R. L., Paoloni, S., Sibilia, C., Yakovlev, V. P., New Compact Setup for Thermal Diffusivity Measurement by Photothermal Deflection Technique, presented at 13. Symposium on Thermophysical Properties, Boulder, 1997
[39] Cahill, D. G., Lee, S. M., Heat Transport Measurements of Thin Films and Interfaces Using the 3ω Method, presented at 13. Symposium on Thermophysical Properties, Boulder, 1997
[40] Feldman, A., Round Robin Thermal Conductivity Measurements on CVD Diamond, *Proceedings,* Third Intl. Conf. on Applications of Diamond Films and *Related Materials,* 1995, pp. 627–630
[41] Graebner, J., Discussion of Diamond Round-Robin II, held at 13. Symposium on Thermophysical Properties, Boulder, 1997

[42] Stanimirović, A. M., Maglić, K. D., Perović, N. Lj., Vuković, G. S., Measurement of Thermal Diffusivity of Thin Films by AC Calorimetric Method, presented at 14. European Conference of Thermophysical Properties, Lyon, 1996
[43] Maglić, K. D., Stanimirović, A. M., Thin Film Thermal Diffusivity Measurement (in Serbian), *Proceedings* XLI ETRAN, *Vol. 1,* 1997, pp. 171–173
[44] Touloukian, Y. S., Powell, R. W., Ho, C. Y., Nicolaou, M. C., Thermophysical Properties of Matter, Vol. 10: Thermal Diffusivity, IFI/Plenum, New York, 1973

Authors address:

A. Stanimirović, Dr. K. Maglić
Laboratory for Thermal Engineering and Energy
VINČA Institute of Nuclear Sciences
P. O. Box 522, 11001 Belgrade, Yugoslavia

Paper submited: December 25, 1987
Paper revised: February 28, 1998
Paper accepted: March 9, 1998