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Plastics — Determination of thermal conductivity and thermal diffusivity —

Part 3: Temperature wave analysis method

Plastiques — Détermination de la conductivité thermique et de la diffusivité thermique —

Partie 3: Méthode par analyse de l'oscillation de la température

Reference number
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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

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The main task of technical committees is to prepare International Standards. Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

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ISO 22007-3 was prepared by Technical Committee ISO/TC 61, *Plastics*, Subcommittee SC 5, *Physical-chemical properties*.

ISO 22007 consists of the following parts, under the general title *Plastics — Determination of thermal conductivity and thermal diffusivity*:

— *Part 1: General principles*

— *Part 2: Transient plane heat source (hot disc) method*

— *Part 3: Temperature wave analysis method*

— *Part 4: Laser flash method*

Introduction

Thermal-transport properties of plastics are indispensable not only in the plastics industry but also in other fields. Plastics are used in various manufacturing processes in new application areas, such as nanotechnologies, and in the biomedical industry. Accurate but simple small-scale measurements are required which can be performed quickly.

High sensitivity and excellent temperature resolution are peculiar to the modulation techniques used for the measurement of thermal-transport properties. Temperature wave analysis is a method of measuring the thermal diffusivity of thin specimens and is also suitable for use with small specimens.

Plastics — Determination of thermal conductivity and thermal diffusivity —

Part 3: Temperature wave analysis method

1 Scope

This part of ISO 22007 specifies a temperature wave analysis method for the determination of the thermal diffusivity of thin films and plates of plastics in the through-thickness direction. The method can be used on plastics in either the solid or molten state, and having either an isotropic or an orthotropic structure.

The method covers values of the thermal diffusivity, α, in the range $1{,}0 \times 10^{-8}\ m^2 \cdot s^{-1} < \alpha < 1{,}0 \times 10^{-4}\ m^2 \cdot s^{-1}$.

Measurements can be performed either in air or in another atmosphere, e.g. an inert gas, at atmospheric pressure or at other, reduced or elevated, pressures, or under a vacuum, at a variety of temperatures.

2 Normative references

The following referenced documents are indispensable for the application of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 472, *Plastics — Vocabulary*

ISO 22007-1, *Plastics — Determination of thermal conductivity and thermal diffusivity — Part 1: General principles*

ISO 80000-5, *Quantities and units — Part 5: Thermodynamics*

ISO/IEC Guide 98-3, *Uncertainty of measurement — Part 3: Guide to the expression of uncertainty in measurement (GUM:1995)*

3 Terms and definitions

For the purposes of this document, the terms and definitions given in ISO 472, ISO 22007-1 and ISO 80000-5 and the following apply.

3.1
temperature wave
temperature oscillation produced by a power-modulated heat source

3.2
phase shift
$\Delta\theta$
phase difference of the temperature wave between the front and rear surfaces of a specimen

NOTE A delay is defined as a negative phase shift.

4 Symbols and units

Symbol	Meaning	Unit
A	slope of a plot of phase shift, $\Delta\theta$, versus the square root of the angular frequency, ω, of the temperature wave	$s^{1/2}$
C	heat capacity per unit volume	$J/(m^3 \cdot K)$
d	thickness of specimen	m
f	frequency of temperature wave	Hz
k	the quantity $(\omega/2\alpha)^{1/2}$	
α	thermal diffusivity	m^2/s
λ	thermal conductivity	$W/(m \cdot K)$
ω	angular frequency of temperature wave	rad/s
ω_c	angular frequency that satisfies the condition $kd = 1$	rad/s

5 Principle

5.1 Temperature wave analysis is a method of measuring thermal diffusivity in the through-thickness direction of a thin, flat specimen by measuring the phase shift of a temperature wave between the front and rear surfaces of the specimen.

5.2 Electrical resistors, sputtered or contacted on both surfaces of the specimen, are used, one as a heater to generate the temperature wave by a.c. Joule heating and the other as a thermometer to detect the temperature wave.

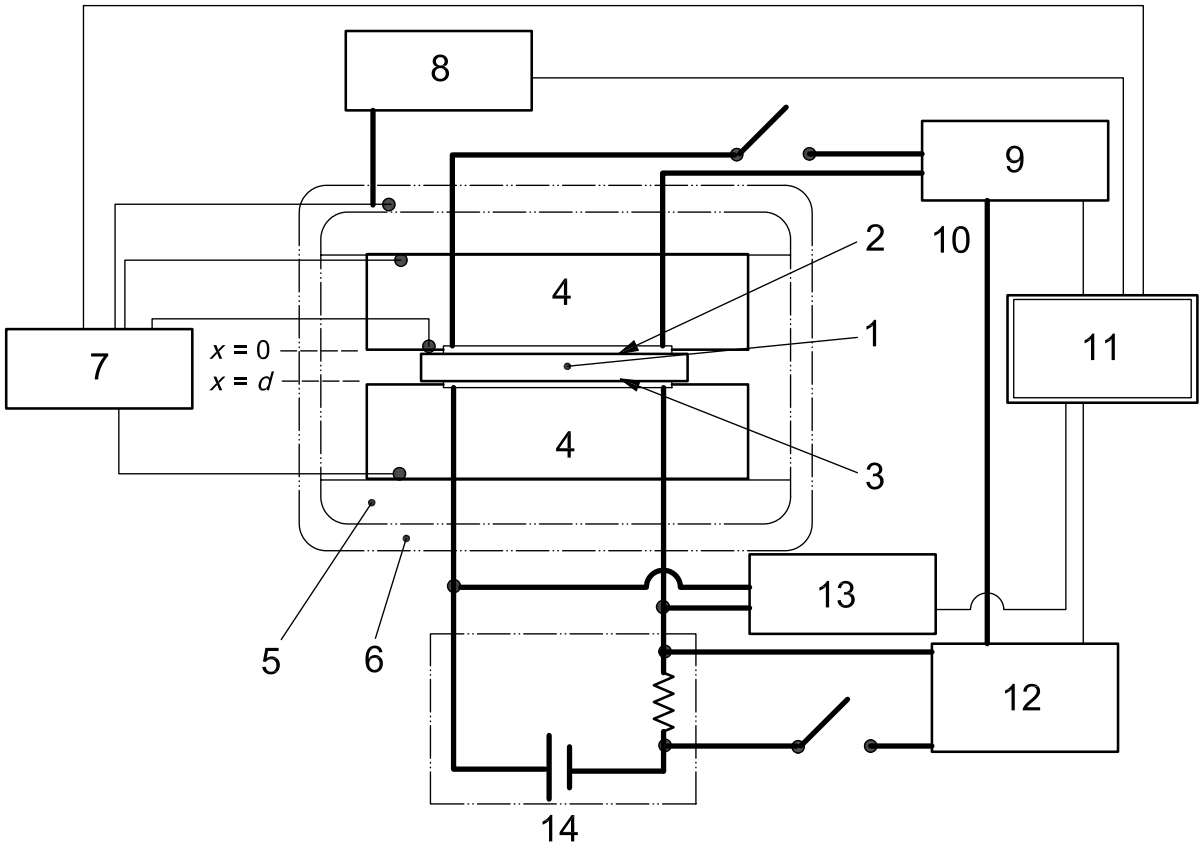
5.3 This method involves analysis of the phase shift of the temperature wave, which is propagated through the specimen, as a function of the square root of the angular frequency of the temperature wave.

NOTE Further details of the theoretical background are given in Annex A and the references in the Bibliography.

6 Apparatus

6.1 General

The apparatus shall be designed to determine the thermal diffusivity as described in Clause 5 and shall consist of the following main components. An example of a suitable apparatus is shown in Figure 1.



Key

1	specimen	6	constant-temperature enclosure	11	personal computer
2	heater	7	thermometer	12	lock-in amplifier
3	sensor	8	temperature controller	13	digital multimeter
4	backing plate	9	function synthesizer	14	bias current circuit
5	specimen holder	10	reference signal		

Figure 1 — Schematic diagram showing an example of a suitable apparatus

6.2 Constant-temperature enclosure

The temperature range of the constant-temperature enclosure shall be appropriate to the materials to be tested.

It shall be possible to control the enclosure temperature such that the specimen temperature does not change by more than $\pm$ 1 K throughout the duration of the measurement.

6.3 Heater and sensor elements

The heater element used to generate the temperature wave by passing alternating current through an electrical resistor attached to the front surface of the specimen is assumed to be located at $x = 0$ (see Figure 1).

The sensor element used to detect the temperature wave by measuring the oscillation of the resistance of an electrical resistor attached to the rear surface of the specimen is assumed to be located at $x = d$.

The heater and sensor should preferably be sputtered directly onto opposite surfaces of the specimen in order to achieve high sensitivity and quick response. The heat capacities of the heater and sensor should be negligible.

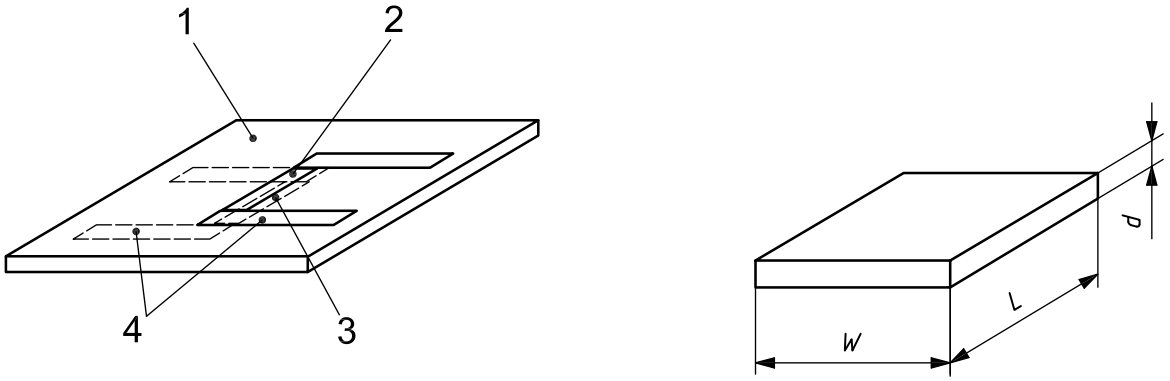
NOTE 1 An example of heater and sensor elements sputtered directly onto the front and rear surfaces of a film specimen is shown in Figure 2 a).

NOTE 2 An example of a heater and sensor set-up that can be used for liquid samples is shown in Figure 2 b). As direct sputtering is not possible in this case, a set of pre-sputtered specimen-backing plates is used.

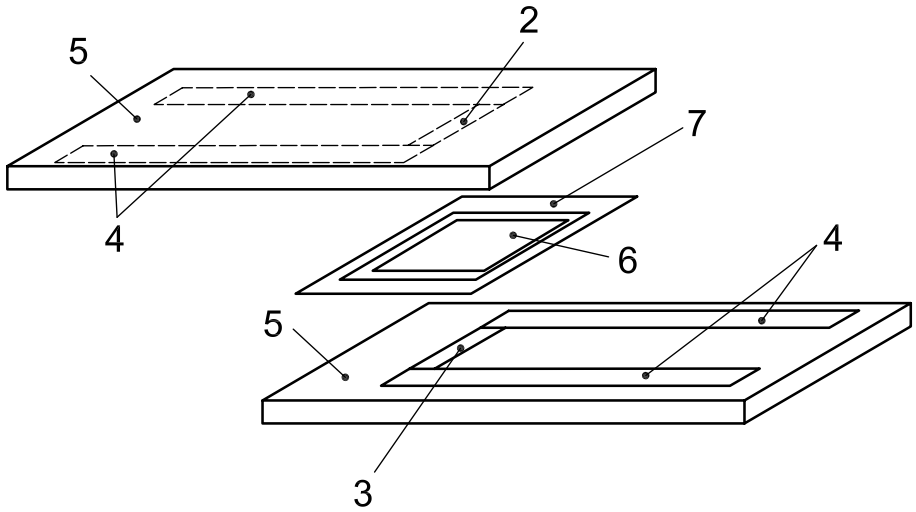
NOTE 3 Typical dimensions of a heater and sensor set-up deposited by sputtering on the specimen are 1 mm wide by 5 mm long.

Metal-layer leads are sputtered or connected using conductive paste or solder to each end of the heater and sensor elements. Alternatively, the leads can be sputtered onto the backing plates [see Figure 2 c)]. Electrical contacts from the lead layers to electric cables for connection to the power supply and measurement devices can be provided using conductive paste or solder.

Direct sputtering of the heater and sensor onto the front and rear surfaces of the specimen is recommended for good thermal contact. As an alternative, the heater and sensor can be attached to the specimen surface under constant load.

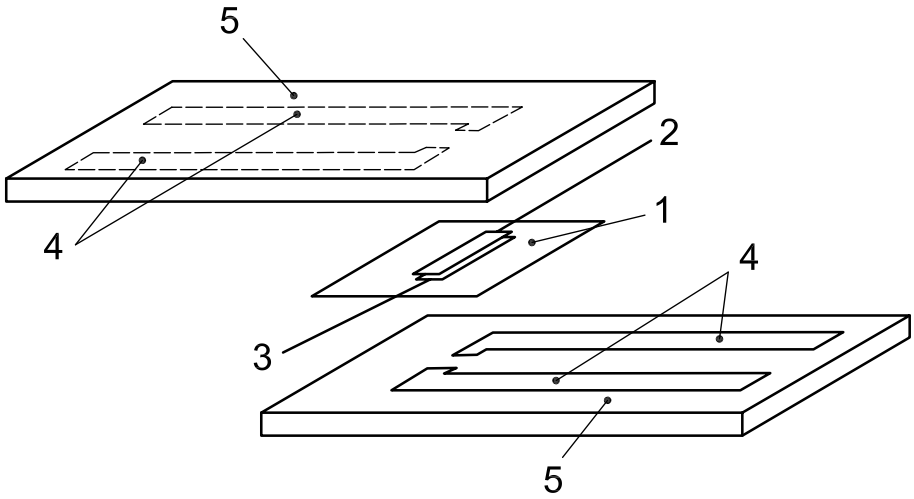


a) Heater and sensor elements and leads sputtered directly on the front and rear surfaces of a film specimen (Examples of dimensions — Specimen: $W = 10$ mm, $L = 10$ mm, $d = 100$ µm; heater: $W = 1$ mm, $L = 5$ mm; sensor: $W = 1$ mm, $L = 5$ mm)



b) Liquid specimen inserted between the backing plates on which the heater and sensor elements and leads are sputtered (Examples of dimensions — Spacer: $W = 10$ mm, $L = 10$ mm, $d = 100$ µm; heater: $W = 1$ mm, $L = 5$ mm; sensor: $W = 1$ mm, $L = 5$ mm; backing plate: $W = 30$ mm, $L = 25$ mm, $d = 2$ mm)

Figure 2 (*continued*)



c) Heater and sensor elements sputtered directly on the specimen, with the leads on the backing plates
(Examples of dimensions — Specimen: $W = 10$ mm, $L = 10$ mm, $d = 100$ µm; heater: $W = 1$ mm, $L = 5$ mm; sensor: $W = 1$ mm, $L = 5$ mm; backing plate: $W = 30$ mm, $L = 25$ mm, $d = 2$ mm)

Key

1	specimen	4	leads	7	spacer
2	sputtered heater (on front)	5	backing plate		
3	sputtered sensor (on rear)	6	specimen (liquid)		

Figure 2 — Examples of sets of heater and sensor elements

6.4 Heating circuit

The power applied to the heater shall be adjusted so as to avoid a specimen temperature rise of more than 1 K.

6.5 Measurement circuit

A bias electric current is supplied to the sensor via the layer leads for the measurement of the oscillation of the electric resistance of the sensor. A d.c. source that can supply an electric current to the sensor in the range 1 µA to 10 mA is used.

It is recommended that the accuracy of measurement of the oscillation frequency be better than 50 ppm.

6.6 Phase-shift measurement device

The oscillation of the electrical resistance of the sensor, observed as an oscillation of the voltage between the ends of the leads to the sensor, shall be measured to determine the phase shift of the temperature wave across the thickness of the specimen. The phase shift between the heater and the sensor shall be measured by a set-up such as that shown in Figure 1. The recommended accuracy for measurement of the phase shift is $\pm$ 0,01°.

6.7 Devices for measuring the specimen temperature

Thermocouples may be fixed onto the backing plates and also attached to the specimen if possible. When the temperature coefficient of the electrical resistance of the sensor is known, the temperature of the sensor can be determined by measuring its electrical resistance.

7 Test specimen

7.1 Dimensions

The specimen shall be a thin film, typically between 10 µm and 500 µm thick. It is usually 10 mm wide by 10 mm long. In principle, for specimens having a flat area larger than the area of the heater and sensor elements (see Figure 2 for examples of dimensions), the permitted thickness range for reliable measurement can be determined from the thermal diffusivity and the temperature wave frequency utilized for the measurement, using the condition $kd > 1$ or $\omega > \omega_c$ (see Annex A or References [1] to [3] for details).

Liquid specimens are inserted between backing plates, using a quantity of liquid sufficient to cover an area larger than that of the heater and sensor elements on the backing plates [see Figure 2 b)]. The thickness shall be kept constant with a spacer, the thickness of which shall be measured in accordance with 7.2.

NOTE A practical example of the frequency-thickness relationships required for acceptable measurements is shown in Annex C.

7.2 Thickness

The specimen thickness shall be measured by means of a suitable calibrated instrument, such as a mechanical micrometer or an optical, electronic, capacitive or inductive gauge. It shall be measured before the specimen is placed in the specimen holder. The variation in the thickness of the specimen should preferably be less than 1 % of the mean thickness or 1 µm, whichever is the smaller.

7.3 Specimen-backing plates

The thickness of the backing plates used should preferably exceed that of the specimen. It is also desirable that the backing-plate material and specimen have similar thermal properties (i.e. similar values of the parameter λk) in order to obtain a wide linear range in the plot of phase shift versus the square root of the angular frequency of the temperature wave (see Figure 3 and Annexes A and D).

NOTE The backing plates are not necessarily limited to solid materials: liquids, gases (including air) or a vacuum could be used.

8 Procedure

8.1 Measure the thickness of the specimen.

8.2 In the case of a solid film, sputter a thin metal layer on each of the surfaces of the specimen to form the heater and sensor. Sputter the metal leads and insert the specimen between the backing plates.

8.3 If the test is performed on a liquid, insert the specimen between backing plates onto which the heater and sensor have already been attached and which are separated by a spacer of known thickness.

8.4 If the heater and sensor cannot be sputtered onto the specimen, mechanical loading of the assembled device can be used to ensure good electrical contact between the leads and the heater and sensor and good thermal contact between the heater and sensor elements and the specimen.

8.5 Place the assembled device containing the specimen in the constant-temperature enclosure. Connect the heater to the power source and the sensor to the phase detector.

8.6 Raise or lower the temperature of the enclosure to the test temperature at not more than 10 K/min.

8.7 Check the specimen temperature. If the resistance of the sensor is not constant throughout the test, this could indicate either that too much power is being supplied to the heater or a lack of thermal equilibrium in the enclosure. In such circumstances, repeat the test with less electric power and/or wait longer to allow the enclosure to come to equilibrium.

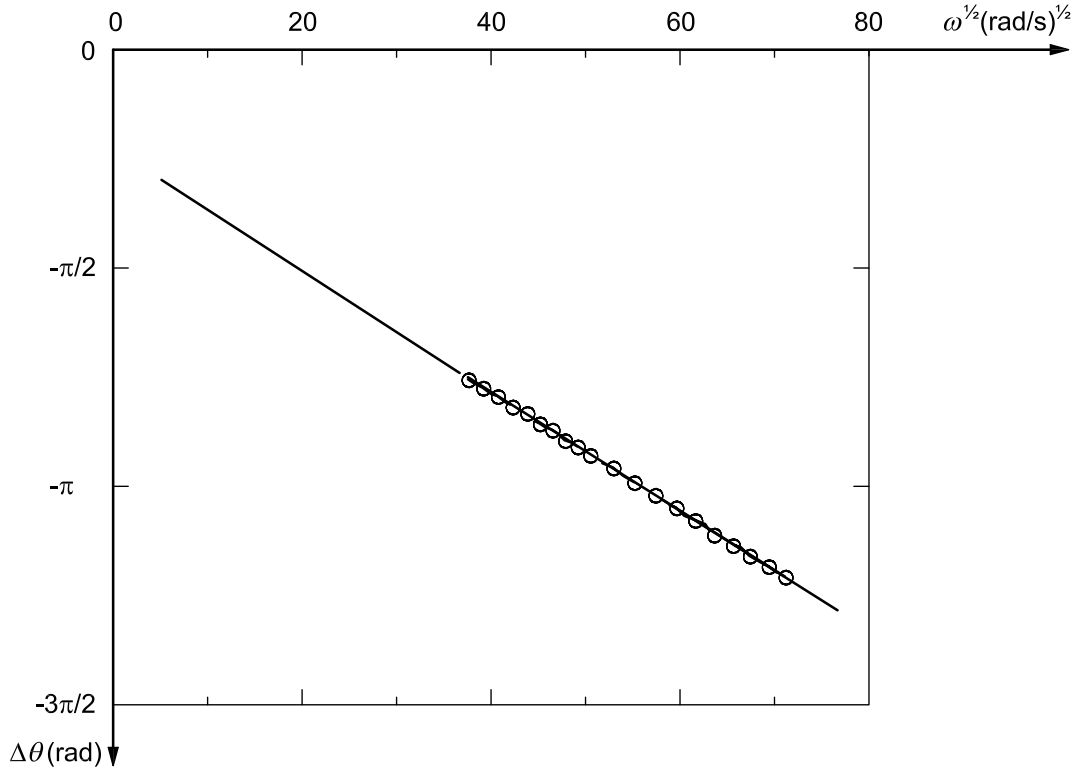


Figure 3 — Example of a plot of phase shift, $\Delta\theta$, versus square root of angular frequency, $\omega^{1/2}$

8.8 Close the measurement circuit, generate an a.c. current in the heater element and apply a bias d.c. current to the sensor. Adjust the power supplied to the heater as a function of the electrical resistance of the heater element and the wave frequency in order to limit the specimen temperature rise to less than 1 K.

8.9 Measure the phase difference of the temperature wave between the front and rear surfaces of the specimen at a fixed temperature wave angular frequency with a phase detector, such as a lock-in amplifier, using a reference signal from the function synthesizer.

8.10 Repeat the procedure described in 8.8 and 8.9 to obtain further measurements of the phase shift at other frequencies. Select at least five frequencies for which the empirical criterion $\Delta\theta < -3\pi/4$ is satisfied (see Figure 3.)

NOTE The empirical criterion $\Delta\theta < -3\pi/4$ is a necessary and sufficient condition to ensure that $kd > 1$ (i.e. $\omega > \omega_c$) is satisfied. This criterion is useful to estimate a suitable frequency range when the thermal properties of the specimen are not known (see also Annex D).

8.11 If the test is also performed at one or more higher or lower temperatures, raise or lower the temperature of the constant-temperature enclosure to the next temperature at not more than 10 K/min. Perform again the procedure specified in 8.7 to 8.10 after the temperature has stabilized, in order to obtain the measurements of the phase shifts.

9 Analysis of results

9.1 For a given specimen thickness and temperature, the thermal diffusivity is calculated from the slope, A, of a plot of phase shift, $\Delta\theta$, versus the square root of the angular frequency, ω, of the temperature wave [1].

9.2 Plotting the phase shift as a function of the square root of the angular frequency should produce a straight-line plot (see Annex A and Figure 3). If this is not the case, it is likely that the angular frequency used

does not fulfil the conditions necessary for this test. In such circumstances, repeat the test over a different frequency range.

9.3 Calculate the specimen thickness at the test temperature, taking into account the change in its thickness due to thermal expansion.

9.4 The thermal diffusivity, α, in square metres per second, of the material is given (see Annex A) by the equation

$$\alpha = \frac{d^2}{2A^2}$$

where

A is the slope of the plot of phase shift, $\Delta\theta$, versus the square root of the angular frequency, ω, of the temperature wave;

d is the thickness of the specimen.

9.5 Check that the condition $\omega > \omega_c$ for the calculation in 9.4 is fulfilled. If it is not fulfilled, repeat the tests using higher angular frequencies.

10 Calibration and verification of apparatus and method

10.1 Calibration

10.1.1 Temperature wave analysis is an absolute method which allows the user to perform measurements that are directly traceable to primary SI units without calibration using reference materials. All elements of the apparatus for temperature wave analysis shall be calibrated separately.

10.1.2 Calibrate the thickness-measurement instrument (see 7.2) to an accuracy of at least $\pm$ 0,1 µm.

10.1.3 Calibrate the device used to measure the steady-state temperature of the test specimen assembly (see 6.7) to an accuracy of at least $\pm$ 0,3 K.

10.1.4 The frequencies of the power supply and the phase-shift measurement device may be calibrated in accordance with the instrument manufacturer's guidelines for the equipment. Check the zero phase shift in the electric circuit itself.

10.2 Verification

Verification of the method and the apparatus can be carried out by making measurements on standard reference materials, preferably covering the range of thermal diffusivities of the materials to be tested and the range of temperatures to be used. It is recommended that any deviation detected during verification be less than 5 % of the reference values. If this limit is exceeded, the measurement conditions should be re-examined and/or the various items of apparatus re-calibrated until the verification is successful.

NOTE Typical values for thin polyimide film are given in Reference [4] and Annex B. Typical values for Pyrex7740[1)] and poly(methyl methacrylate) are given in Reference [5].

1) Product available commercially. This information is given for the convenience of users of this part of ISO 22007, and does not constitute an endorsement by ISO of the product named.

11 Precision and bias

11.1 Uncertainty

In routine measurements near room temperature for a specimen with a heater and sensor prepared by direct sputtering, the expanded uncertainty (confidence level of 95 %) of thermal-diffusivity determination by the temperature wave method is estimated to be better than 5 % in the range $1{,}0 \times 10^{-7}\ m^2 \cdot s^{-1} < \alpha < 1{,}0 \times 10^{-6}\ m^2 \cdot s^{-1}$ when determined in accordance with ISO/IEC Guide 98-3. If an unsputtered sensor in contact with the specimen is used, the expanded uncertainty is estimated to be in the range 5 % to 10 % (see Annex E).

11.2 Repeatability

The repeatability of successive measurements using the same test conditions and the same specimen is expected to be better than 1 %.

12 Test report

The test results shall include the following information:

a) the name and address of the test laboratory;

b) the date of the test;

c) a reference to this part of ISO 22007;

d) all details necessary for complete identification of the sample tested (manufacturer, product, type, batch number, etc.) and details of its thermal history;

e) the shape, thickness and other dimensions of the specimens and the number of specimens tested;

f) the specifications of the instrument used;

g) the type of sensor and heater used and their shapes, dimensions and electric resistances;

h) the measurement conditions, such as temperature, modulated power supplied to heater, bias current on sensor, atmosphere in the constant-temperature enclosure, angular frequency of temperature wave;

i) the test results, such as the phase shifts at the various test frequencies and temperatures;

j) a plot of phase shift versus the square root of the angular frequency (see Figure 3) for each temperature;

k) the value of the slope, A, of the plot and the thermal diffusivity, α, at the various test temperatures;

l) any other relevant items.

Annex A
(informative)

Mathematical background to temperature wave analysis

A one-dimensional heat conduction model is assumed for a specimen of thickness d, located between backing plates of semi-infinite thickness. When a sinusoidal temperature wave is generated at the front surface of a specimen ($x = 0$), it is propagated in the through-thickness direction and is detected at the rear surface ($x = d$). If the amplitude of the temperature wave decays to zero at an infinite position in the backing plates, the temperature oscillation $T(x,t)$ at $x = d$ is described as follows, by solving the one-dimensional diffusion equation [1] to [3]:

$$T(d,t) = \frac{\left[j_0 \exp(\mathrm{i}\omega t) \Big/ (1+\mathrm{i}) \right] \exp\left[-(1+\mathrm{i}) kd \right]}{\left\{ (\lambda k + \lambda_\mathrm{b} k_\mathrm{b})^2 - (\lambda k - \lambda_\mathrm{b} k_\mathrm{b})^2 \exp\left[-2(1+\mathrm{i}) kd \right] \right\} \Big/ 2\lambda k} \tag{A.1}$$

where

$T(d,t)$ is the temperature oscillation at $x = d$;

t is time;

j_0 is the periodical heat flux generated at $x = 0$, where $j = j_0 \exp(\mathrm{i}\omega t)$;

i is $(-1)^{1/2}$;

ω is the angular frequency;

λ is the thermal conductivity;

$k = (\omega/2\alpha)^{1/2}$, where α is the thermal diffusivity;

subscript "b" refers to the backing plates.

If the condition $kd > 1$ or $\lambda k \cong \lambda_\mathrm{b} k_\mathrm{b}$ is satisfied, Equation (A.1) can be simplified as follows:

$$T(d,t) = \frac{\sqrt{2} j_0 \lambda k \exp(-kd)}{(\lambda k + \lambda_\mathrm{b} k_\mathrm{b})^2} \exp\left[\mathrm{i}\left(\omega t - kd - \frac{\pi}{4} \right) \right] \tag{A.2}$$

The phase shift between the heater and the sensor is described by

$$\Delta\theta = -\sqrt{\frac{\omega}{2\alpha}} d - \frac{\pi}{4} \tag{A.3}$$

The phase shift, $\Delta\theta$, is a linear function of the square root of the angular frequency, ω, and thus the thermal diffusivity of the specimen can be determined from

$$\alpha = \frac{d^2}{2A^2} \tag{A.4}$$

where A is the slope of the linear part of the plot of phase shift versus the square root of the angular frequency for a given thickness, d.

Annex B
(informative)

Typical thermal-diffusivity data for a typical polyimide

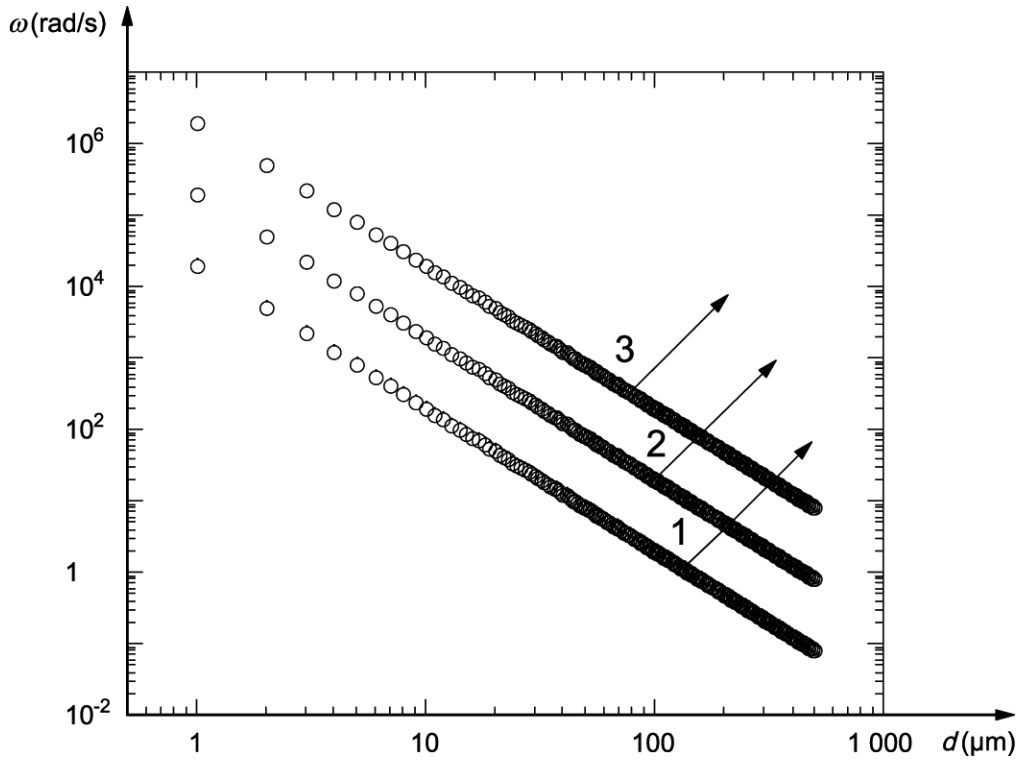
Table B.1 — Typical thermal-diffusivity data for a typical polyimide [4]

T °C	α $mm^2 \cdot s^{-1}$	T °C	α $mm^2 \cdot s^{-1}$
− 50	0,147	130	0,108
− 40	0,144	140	0,106
− 30	0,141	150	0,104
− 20	0,138	160	0,102
− 10	0,135	170	0,101
0	0,133	180	0,099 0
10	0,131	190	0,097 4
20	0,129	200	0,095 9
30	0,127	210	0,094 4
40	0,125	220	0,093 0
50	0,123	230	0,091 5
60	0,121	240	0,090 0
70	0,119	250	0,088 5
80	0,117	260	0,087 3
90	0,115	270	0,085 7
100	0,113	280	0,084 4
110	0,111	290	0,083 2
120	0,110	300	0,081 8
Sample: Kapton 100[2)]. Thickness: 25 µm.			

2) Product available commercially. This information is given for the convenience of users of this part of ISO 22007, and does not constitute an endorsement by ISO of the product named.

Annex C
(informative)

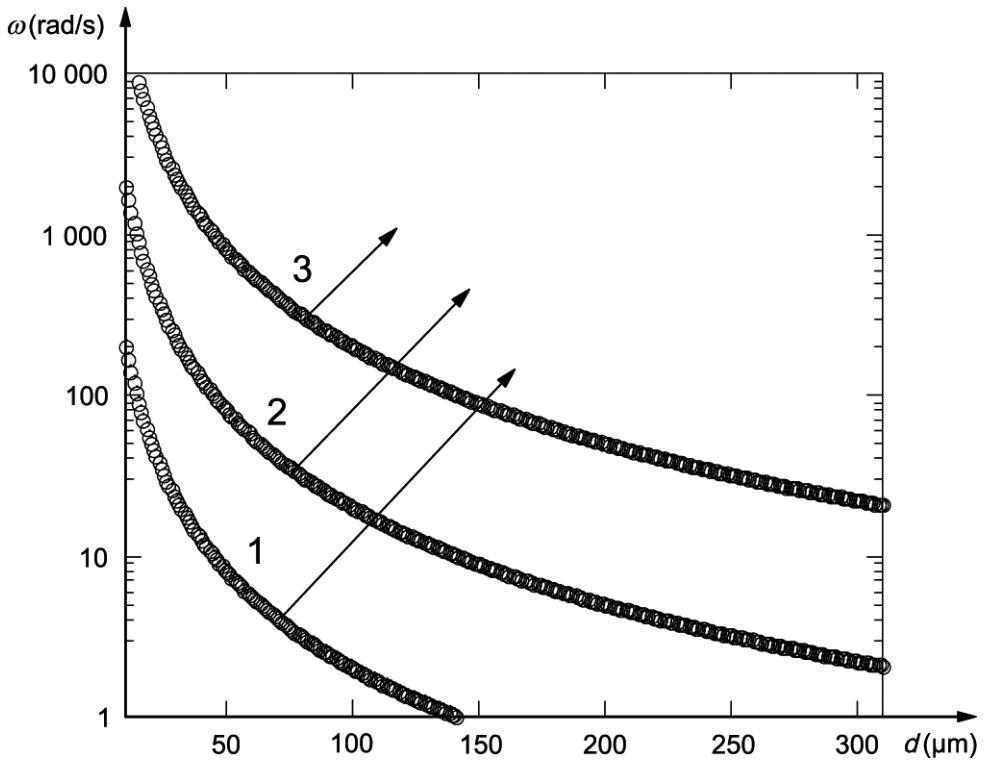
Example of frequency-thickness relationships required for acceptable measurements



Key

1 $\alpha = 1{,}0 \times 10^{-8}$ m^2·s^{-1}
2 $\alpha = 1{,}0 \times 10^{-7}$ m^2·s^{-1}
3 $\alpha = 1{,}0 \times 10^{-6}$ m^2·s^{-1}

Figure C.1 — Frequency-thickness relationships required for acceptable measurements (log-log plot)



Key

1 $\alpha = 1{,}0 \times 10^{-8}$ m^2·s^{-1}
2 $\alpha = 1{,}0 \times 10^{-7}$ m^2·s^{-1}
3 $\alpha = 1{,}0 \times 10^{-6}$ m^2·s^{-1}

Figure C.2 — Frequency-thickness relationships required for acceptable measurements (log-linear plot)

Annex D
(informative)

Numerical simulations of the phase shift, $\Delta\theta$, as a function of kd and ξ

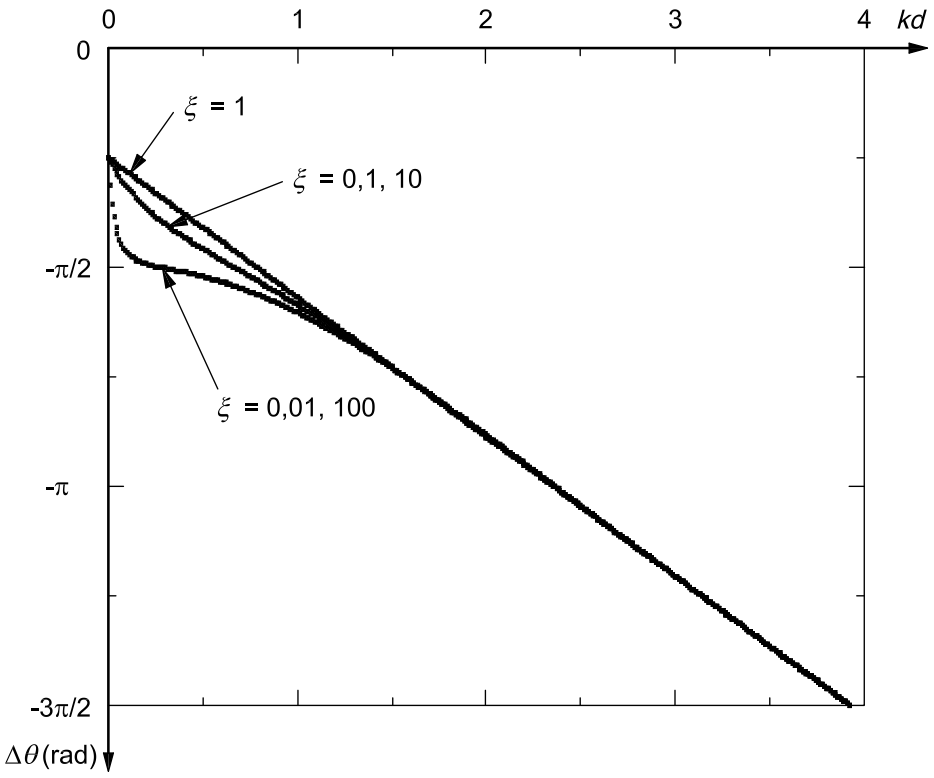


Figure D.1 — Numerical simulations of the phase shift, $\Delta\theta$, as a function of kd and ξ, where ξ is defined as $\xi = \lambda k/\lambda_s k_s$ showing sensitivity to ξ for $\Delta\theta > -3\pi/4$

Annex E
(informative)

Examples of uncertainties in thermal-diffusivity measurements

Table E.1 — Examples of uncertainties in thermal-diffusivity measurements at room temperature

Material	d µm	α $m^2 \cdot s^{-1}$	Expanded uncertainty, % ($k = 2$)
PMMA	126	$1{,}15 \times 10^{-7}$	3,6
PES	200	$1{,}24 \times 10^{-7}$	2
Zirconia	500	$1{,}08 \times 10^{-6}$	2,3

Bibliography

[1] MORIKAWA, J., TAN, J. and HASHIMOTO, T.: Study of change in thermal diffusivity of amorphous polymers during glass transition, *Polymer*, **36** (23), pp. 4439-4443 (1995)

[2] MORIKAWA, J. and HASHIMOTO, T.: Analysis of high-order harmonics of temperature wave for Fourier transform thermal analysis, *Jpn. J. Appl. Phys.*, **37**, pp. L1484-L1487 (1998)

[3] HATTA, I., SAKAKIBARA, K., SUZUKI, J. and YAO, H.: An a.c. calorimetric method for measuring thermal diffusivity of liquids, *Jpn. J. Appl. Phys.*, **27**, pp. 2156-2159 (1988)

[4] HASHIMOTO, T.: *Data Book of Thermal Diffusivity of Polymers*, Youtes, Tokyo (1994)

[5] TYE, R. and SALMON, D.: Thermal conductivity certified reference materials: Pyrex7740 and polymethylmethacrylate, *Thermal conductivity 26/Thermal expansion 14*, pp. 437-451, DEStech Publications, Inc. (2001)

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