Archives
of Materials Science
and Engineering

Volume 47
Issue 2
February 2011
Pages 95-102

International Scientific Journal
published monthly by the
World Academy of Materials
and Manufacturing Engineering

Thermal conductivity measuring station for metallic glasses

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Received 08.11.2010; published in revised form 01.02.2011

ABSTRACT

Purpose: In the present paper an equipment applied in thermal conductivity measurements of metallic glasses was described.

Design/methodology/approach: The paper describes the design solution of a measuring station, components, and idea of measurements of thermal conductivity. In order to correct measurement the calibration of presented equipment was realized. It was realized by determination of power losses and resistance of contacts. Methods of thermal conductivity measurements were also described in theoretical description.

Findings: The suggested method of thermal conductivity measurement allows to avoid a procedure of solving complicated equations. The developed measuring station enables measurements of thermal conductivity of bulk metallic glasses in form of rod with diameter 3 mm.

Research limitations/implications: The relationship between the thermal conductivity and the diameter of metallic glass samples is an interesting issue. In the future the authors are going to test rods with another diameters (not only 3 mm).

Practical implications: The thermal conductivity of metallic glasses is necessary to calculate cooling rates during the fabrication of bulk metallic glasses. That are very important properties. These properties are indispensable for example in a computer simulation of a solidification process.

Originality/value: Up to now there is very poor knowledge about thermal conductivity measurements of metallic glasses. There is not many references about this matter. There is no information about the thermal conductivity dependence on samples dimensions of metallic glasses.

Keywords: Thermal conductivity; Bulk metallic glasses; Computer simulation; Measuring station

Reference to this paper should be given in the following way:

A. Pusz, A. Januszka, S. Lesz, R. Nowosielski, Thermal conductivity measuring station for metallic glasses, Archives of Materials Science and Engineering 47/2 (2011) 95-102.

PROPERTIES

1. Introduction

Thermal conductivity of metals is one of the most significant parameters characterizing its metallurgical properties. This parameter is needed to calculate cooling rates during the fabrication of this engineering materials.

Bulk metallic glasses are newcomers materials which exhibit excellent physical and functional properties. In a fabrication process of this materials, very important problem is knowledge about glass forming ability of alloy [1-6]. Glass forming ability depends on various factors. Characteristic temperature such as liquid temperature (Tl), glass transition temperature (Tg) and crystallization temperature (Tx) are included in mathematical description of this factors. Among many of bulk metallic glasses systems, Fe-based are one of the most popular and exhibit good glass formability. The (Fe, Co, Ni)-B-Si-Nb system alloys exhibits high glass-forming ability, super-high fracture strength and high plastic strain. Moreover, these alloys exhibit good soft-magnetic properties [7-13].

In order to better understanding of solidification process during bulk metallic glasses fabrication, computer simulation of casting process and temperature distribution can be realized. Computer programs such as FLUENT, ABAQUS etc. allow to realize an analyze of temperature distribution or calculate cooling rates. As an input data for computer simulation of casting process few important properties are necessary. The analyze of heat flow in mould casting requires knowledge about thermal properties of casting alloys (e.g specific heat, liquids temperature, thermal conductivity).

Table 1 shows relationship between general thermal properties and others physical properties of metals, ceramics and polymers.

Table 1.
Characteristic main types of engineering materials [14]

Properties	Metals	Ceramics	Polymers
chemical resistance	low to medium	excellent	good
creep resistance	poor to medium	excellent	poor
density	high	medium	low
electrical conductivity	high	very low	very low
hardness	medium	high	low
machinability	good	poor	good
malleability	high	-	high
melting point	low to high	high	low
stiffness	high	high	low
strength	high	very high	low
thermal conductivity	medium to high	medium but often decreasing rapidly with temperature	very low
thermal expansion	medium to high	low to medium	very high
thermal shock resistance	good	generally poor	good within limited temperature ranges

Among thermal properties the most important that we can determinate are [14]:

- *Coefficient of linear thermal expansion* (coefficient of linear expansion, or α, coefficient of thermal expansion (CTE), linear expansion coefficient, linear thermal expansion coefficient, thermal coefficient of expansion, thermal expansion coefficient);
- *Emmitance* (emissivity, thermal emissivity);
- *Liquidus temperature*;
- *Melting range and melting point*;
- *Solidus temperature*;
- *Specific heat capacity* (*C*, *C*p, *C*v, heat capacity per unit mass, specific energy capacity, specific entropy, specific heat);
- *Thermal conductivity* (*k*, λ).

Figure 1 presents thermal conductivity of several materials and liquids.

The measurement of thermal conductivity involves a set of parameters that are common to different techniques and methodologies. Aside from variations due to the nature and type of samples, all methodologies require determination of the actual amount of heat transferred through the sample along and perpendicular to the heat flow path in a given thermal environment. The calculated value is expressed in the same unit as that provided for a standard of the same material. Conductivity, as opposed to conductance, provides dimensional attributes to the calculated value. Thus, thermal conductivity is related to a material property that denotes a rate process of heat transfer. Conductivity is a function of diffusivity, density and heat capacity. Whereas through-thickness thermal conductivity for fixed-dimension solids is primarily measured under steady-state conditions, accompanying transient diffusivity in the radial direction is taken into account by using the ratio of sample thickness to the total sample area as the heat flow path. The relationship is expressed as (Equation 1)[15]:

$$\lambda = \frac{q \cdot L}{A(t_1 - t_2)} \qquad (1)$$

where:

λ - thermal conductivity [W/mK],
q - time rate of heat flow [W],
L - thickness of sample in the heat flow direction [m],
A - area of sample [m^2],
t_1 - temperature of hot surface [K],
t_2 - temperature of cold surface [K].

In order to measurement thermal conductivity of bulk metallic glasses a measuring station, which is adapted for samples with diameter 3 mm, was proposed in this paper.

Methods of thermal conductivity measurements can be classified as stationary and nonstationary. An equation of thermal conduction must be solved for indicate of thermal conductivity. For stationary methods there is condition (2) which must be performed [17-19] :

$$\frac{\partial T}{\partial t} = 0 \ \# \qquad (2)$$

where:

T - temperature [ºC],
t - time [s].

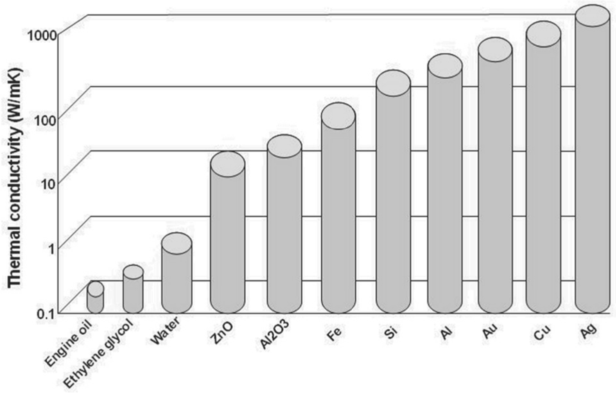

Fig. 1. Comparison of thermal conductivity of several materials [16]

In stationary methods, fields of temperature in the tested sample are unchangeable. Thanks to this methods coefficient of thermal conductivity (λ) may be direct assigned [17].

Stationary methods of thermal conductivity measurements consist of comparative stationary methods and absolute methods. In the first category for correct measurements reference standard with known thermal conductivity is demanded (for example electrolytic iron, graphite or copper).

Methods of thermal conductivity measurements are sectional for the sake of heating way of sample too. We can distinguish methods with external heating of the sample and internal heating of sample where passage of current causes heating of the sample [17-18].

Thermal conductivity calculation by specify methods depends on determine of heat flux and measure of distance between temperature sensors which are located in tested sample and adequate temperatures measurement.

Nonstationary thermal conductivity methods are called indirect. Using the Equation 3 we can define thermal diffusivity which determinates thermal conductivity ability in transient conditions [17]

$$\frac{\mathrm{dT}}{\mathrm{dt}} = a \cdot \nabla^2 T \tag{3}$$

Next thermal conductivity may be defined with Equation 4 [17-18].

$$\lambda = a \cdot \rho \cdot c_p \tag{4}$$

where:
a -thermal diffusivity [m^2/s],
c_p - specific heat [J/kg K],
ρ - density [kg/m^3].

Fig. 2 shows classification of thermal conductivity measurement methods.

Determination of thermal conductivity coefficient by stationary method requires steady distribution of temperature on cross section of the sample. It impose necessity of measurements up to temperature difference stabilization on both isothermal surfaces.

Depending on measuring system and sample thermal inertia time of measurements may last even few hours.

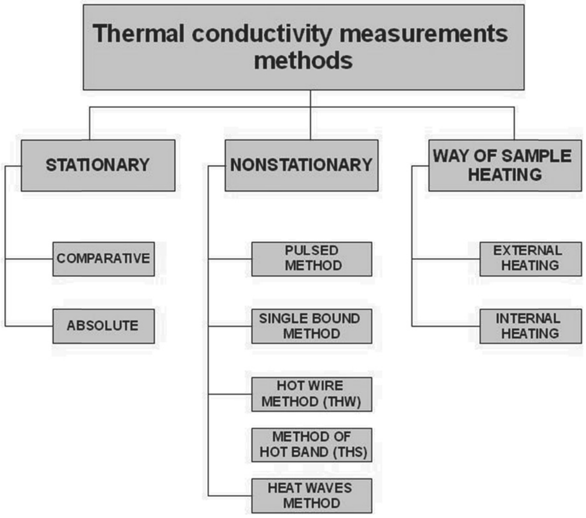

Fig. 2. Classification of thermal conductivity measurements methods [own work]

2. Technical assumptions

For the sake of the sample shape and measurements conditions a physical method of thermal conductivity measurements with maintain of stationary condition of temperature flow was used.

It demands special construction of measuring station and specific registration of results.

The measuring station was constructed based on some findings of PN-EN 12667:2002. Fig. 3 show scheme of constructed system and Figure 4 presents real picture.

The measuring station consists of the following elements:

- a measurement module,
- a DC power supply,
- temperature gauges with thermocouples,
- a PC with A/D card,
- a vacuum pump.

A heater which is mounted in module is supplied by DC power with critical parameters 30V/2 A. The heater was performed of resistance wire with determinate resistance. Temperature on the sample surface was measured using two thermocouples which are connected with temperature gauges.

A system of thermocouples and temperature gauges is connected with A/D card.

In order to elimination of convection losses, the sample with whole measurement module is located under a glass casing. Thanks to this protection measurements could be realized in vacuum.

In thermal conductivity measurement method, which is used in measurement of bulk metallic glasses with diameter 3 mm, the tested sample is placed between the heater and a cooling element. For the sake of contact resistance, a conductive paste was putted on ends of the sample.

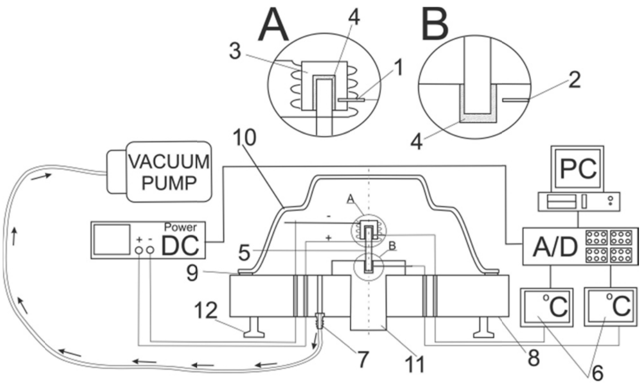

Fig. 3 Scheme of the thermal conductivity measuring system: A - upper mounted of sample; B - lower mounted of sample; 1 - an upper thermocouple; 2 - a lower thermocouple; 3 - a heater; 4 - a conductive paste; 5 - a sample; 6 - a temperature gauges; 7 - vacuum pump connection; 8 - a base plate; 9 - a gasket; 10 - a glass casing; 11 - a cooling element; 12 - a supporting leg

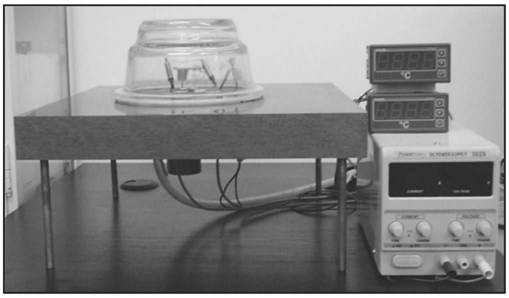

Fig. 4. Real photo of the measuring station

A source of heat has a form of a heater with resistance 10.12 Ω. The resistance wire was wounded on a copper roll and secured by a silicon layer. The cooling element has also a form of a copper roll with larger diameter.

Data registration and its visualization (voltage of the heater supply, voltage of thermocouples) are realized with the aid of special software (in Delphi). Temperature of upper and lower thermocouples and voltage in time function are recorded. Time step, thickness of the sample and name of the data file are determinate in software. On the basis of the registration data temperature difference, thermal conductivity and power of the heater are calculated.

2.1. Calibration

In order to correct operating of the measurement station the calibration of a measurement module was necessary. The calibration enclosed determination of power losses and contact resistance.

- Power losses calibration

Power losses in the measuring system appeared as heat losses to environment by radiation.

In order to determine power losses between the heater and the cooling element a heat insulator was placed. The insulator dimension was as small as it can be (Fig. 5).

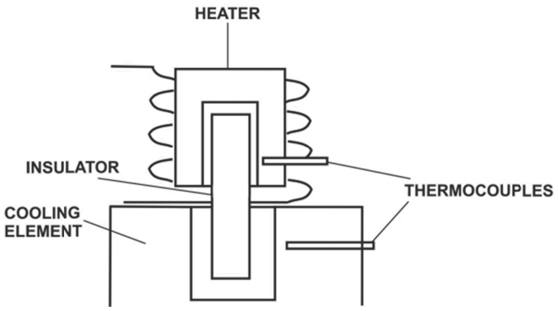

Fig. 5. Scheme of the measurement module with the insulator instead of the sample (during power losses calibration)

Heat losses measurements were realized for the heater power 0.5; 1; 1.1; 1.2; 1.5 [W].

In order to determination of the heater power, tension which is determined as $U = \sqrt{P \cdot R}$ was calculated:

where:

U - tension [V],

P - power of heater [W],

R - known resistance of heater [Ω].

During measurements, a graph of upper thermocouple temperature for adequate heater power was observed. After temperature stabilization power of the heater was increased. Figure 6 shows dependence of upper thermocouple temperature on time for adequate power of heater.

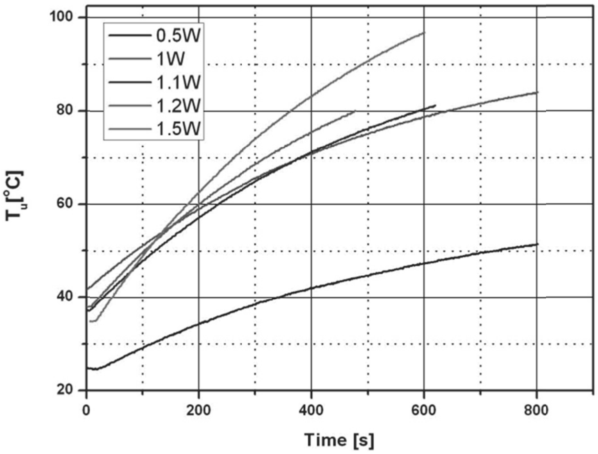

Fig. 6. Relationship between temperature of upper thermocouple (T_u), time and power of heater

Each diagram which was obtained from measurements was approximated by first-order approximation. Values of stabilized T_u were calculated.

Power of losses (P_l) is expressed as a function of stabilized temperature stabilized temperature of upper thermocouple. $P_l = f(T_u)$

To determine P_l, relationship between each power of heater and stabilized temperature of T_u was placed on a graph (Fig. 7) Next approximation by linear function was realized.

$$P_l = A + B \cdot T_u$$

where:

P_l - power of losses [W],

T_u - temperature of upper thermocouple [ºC].

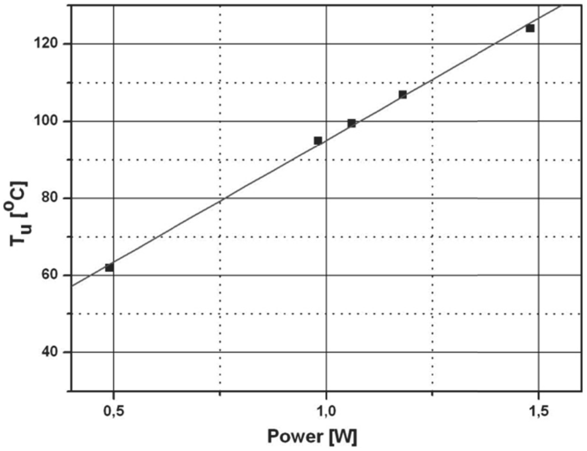

Fig. 7. Relationship between temperature of upper thermocouple (T_u) and power of heater

- Contact resistance calibration

The contact heater and the cooling element with the sample are not ideal. For that reason between thermocouples exists additional decrease of temperatures.

Resistance of contact is expressed as a difference of thermocouples upper and lower temperature (ΔT). As a layer which replaced a sample, conductive paste was used.

Calibration for contact resistance was realized for three power values: 0.5; 1 and 1.5 W. A copper standard sample was used for test. The standard sample was prepared from copper rod. Purity of used copper equal 99,9% with some alloy addition as a P, Bi, Pb [20]. In order to better contacts of the sample with heater and cooling element, the conductive paste was applied on the ends of the standard sample (Fig. 8).

Calibration test relied on measurements at one power of heater until temperature stabilization. Then, the power of heater was increased to next value and measurement was continued until temperature stabilization again.

Time of measurement for every power of heater was about 200 s. Five independent measurements for every power were realized. Next for ΔT precision an approximation was carried out. Approximation was realized by function $Y = y_0 + A_1 * \exp(-x/t_1)$.

Figure 9 presents approximation of ΔT for each of power. Table 2 shows values of the first order approximation coefficients.

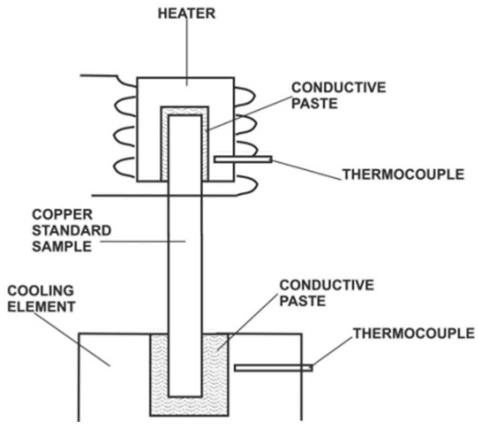

Fig. 8. Scheme of measurement module with copper standard sample (during contact resistance calibration)

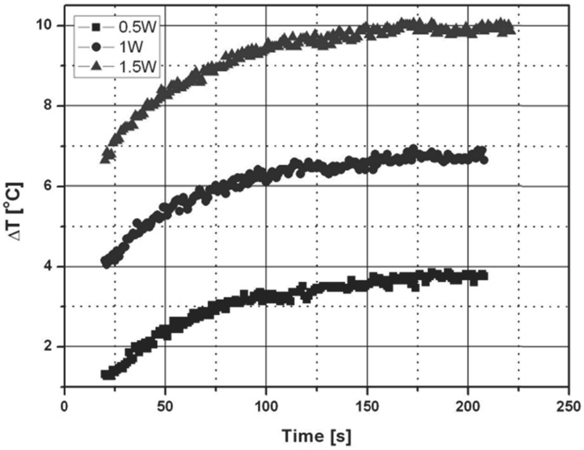

Fig. 9. Relationship between time and ΔT for adequate power of heater (0.5; 1 and 1.5 W)

Table 2
Values of the first order approximation coefficients

	$Y = y_0 + A_1 \cdot \exp(-x/t_1)$		
Power [W]	y_0	A_1	t_1
0.5	3.81	-3.86	51.34
1	6.79	-223.77	48.72
1.5	10.00	-17400.75	47.06

On the basis of the results of approximation, a standard deviation was defined and enclose on the diagram.

Results of the standard deviation calculations are presented in Table 3.

After approximation of every measurement there were possibility a draw curve which illustrate relationship between power of heater and temperature drop on sample contact (Fig. 10).

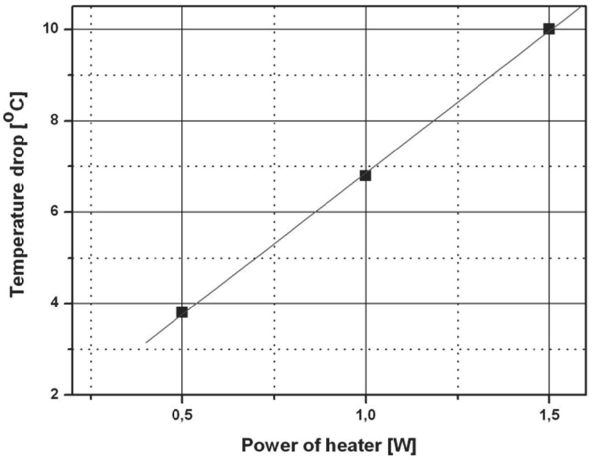

Fig. 10. Relationship between temperature drop on sample contact and power of heater

On the basis of the above diagram (Fig. 10) it was found that the lowest temperature drop occurred in 0.5 W power of heater.

In order to stabilized measurements conditions for test stand, the minimal number of measurements were defined. The thermal conductivity of copper standard sample were carried out. Ten measurements were also realized.

Mean for 5 and 10 measurements (Table 4) shows which of number of measurements is closer to real thermal conductivity of copper (398 W/mK) [21]. For five measurements, average thermal conductivity of copper standard sample was comparable to thermal conductivity from References [21].

On the basis of above calculations it was found that number of measurements should Equal 5.

Table 3.
Results of standard deviation

Power of heater [W]	Standard deviation (sd(yEr$^{+}_{-}$))
0.5	0.50
1	0.56
1.5	0.85

3. Test thermal conductivity measurement

After measurement system calibration, first thermal conductivity measurement of bulk metallic glasses was realized.

An amorphous rod with diameter 3 mm was used as a sample for analyse. After measurement parameters set up, thermal conductivity was measured. Five measurements for power of heater equal 0.5W were done. Each of them was approximated by function $Y=y_0+A_1*\exp(-x/t_1)+A_2*\exp(-x/t_2)$. The last step was mean definition. For tested sample the mean thermal conductivity equal 26.49 W/mK.

Fig. 11 shows results for five thermal conductivity measurements. Additionally Table 5 presents values of the second order approximation parameters.

Table 4.
Mean calculations for ten and for five thermal conductivity measurements

No	Thermal conductivity [W/mK]	Mean for 5	Mean for 10
1	367.19	346.88	340.84
2	332.81		
3	349.38		
4	347.82		
5	337.17		
6	313.61		
7	332.43		
8	332.73		
9	345.70		
10	349.52		

Table 5
Values of the second order approximation coefficients

	$Y=y_0 + A_1 \cdot \exp(-x/t_1) + A_2 \cdot \exp(-x/t_2)$				
No	y_0	A_1	t_1	A_2	t_2
1	31.37	35854.25	16.52	553.16	93.57
2	28.64	6481.19	15.22	437.45	89.82
3	25.45	4160.32	14.20	362.07	94.69
4	27.53	2865.11	11.19	393.01	80.94
5	19.51	230.91	123.24	1133.52	21.93

4. Conclusions

The measuring station, which was described and performed in this work, fulfilled all assumption aims. It allowed to made thermal conductivity measurements for samples with diameter 3mm.

On the basis of the measure system calibration it was found that the lowest power loses occurred in 0.5 W power of heater and it equal 62ºC. The highest power loses occur in 1.5 W.

For contact resistance calibration the lowest average of temperature drop existed in 0.5 W power of heater and equaled 3.76ºC . The highest temperature drop occur in 1.5 W power of heater and equal 9.3ºC.

Approximations which were realized during calibration of the measuring system allow to determine basic parameters for correctly functioning of whole measuring station.

A constructed measuring system is a very useful engineering tool which efficiently allows to measure temperature, visualize main parameters, calculate thermal conductivity and save all data.

In near future measuring system will be developed in order to measure samples with other diameters.

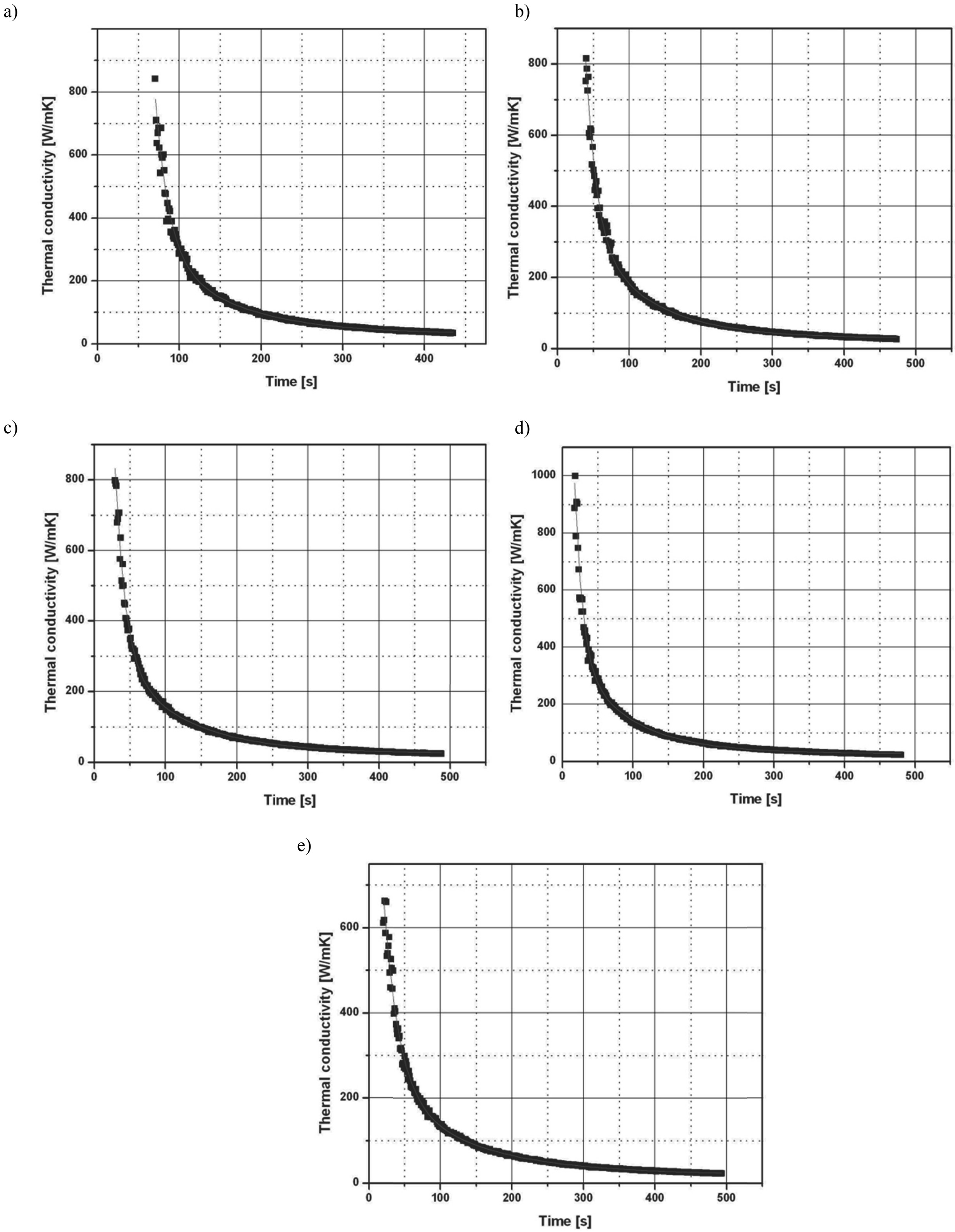

Fig. 11. Results of five thermal conductivity measurements (a-e)

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