EXTENDED VERSION OF PULSE TRANSIENT METHOD

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Abstract

The aim of the paper is a modification of Pulse transient method for a specimen in sandwich-like setup, where outer parts of the specimen setup are replaced by sample reference and thus, measured sample does not need to be cut into three parts as in original method. The Pulse transient method, known as classical transient method, is used for measurement of three thermophysical parameters, namely, thermal diffusivity, thermal conductivity and specific heat. Complete analytical solutions of temperature distribution in individual parts of specimen setup are presented for a heat produced in form of the Dirac or the Step-wise function. Sensitivity coefficients of thermophysical parameters and their linear dependency were inspected. In order to verify functionality of the proposed model, an experiment on ceramic SiC was carried out. The experimental data were compared with Flash and DSC data, where satisfying results were found.

Keywords: pulse transient, sandwich-like, thermal conductivity, thermal diffusivity, specific heat

1 Introduction

In the last several years the transient methods [1] started to be widely used in measurements of thermophysical parameters of materials. When compared to classical methods (Guarded hot plate, DSC), their main advantages are quite short time of measurement, simple build up of experimental apparatus and capability of three thermophysical parameters measurement in one experiment.

The use of the Pulse transient method [2] is conditioned by need to have a sample that is cut into three parts. In some cases like complicated preparation of sample, insufficient resources for production, expensive technological process or uniqueness of a sample, we have no chance to gain the sample in quantity of three pieces. To overcome these obstacles, a model with outer parts of specimen setup replaced by sample reference is proposed as an extension to the Pulse transient method.

In this paper I present the complete one-dimensional analytical solution of temperature distribution in each part of the specimen setup due to a heat produced in the form of the Dirac or the Step-wise function. Solution of similar problem with heat produced in form of the Dirac pulse was presented by Kulakov [3]. Surprisingly, his equations of a temperature distribution are completely different from those, presented here. The sensitivity coefficients of the thermophysical parameters and their linear dependency are shown and inspected. Moreover, the sensitivity coefficients in dependence on the thermophysical properties of outer parts and middle part are depicted and discussed. Experiments were carried out on sample made of ceramic SiC (middle part) with sample references (outer parts) made of PTFE at room temperate and air atmosphere.

2 Model

The extended version of the Pulse transient method is depicted in Fig. 1, where the specimen setup consists of two outer parts (I, III) having known thermophysical properties (sample reference) and one middle part (II) of unknown properties. The heat pulse is produced due to Joule heating from electrical resistance of a planar source situated between the first and the second part. A thermocouple is placed between the second and the third part. The method can be described as follows. Temperature of the specimen is stabilized and uniform. Then a small disturbance in form of a heat pulse is applied to the specimen. The thermophysical parameters of unknown sample are calculated upon temperature response according to the model used.

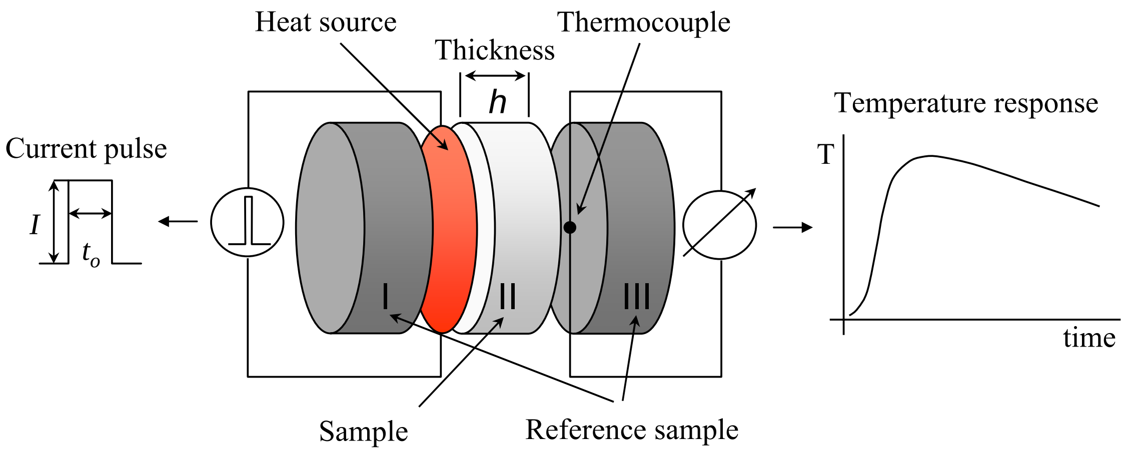

Fig 1 Pulse transient method in sandwich-like specimen setup

The model considering real experimental setup leads to complicated mathematical expressions with unknown solutions. Therefore, some simplifying assumptions have to be postulated. The following assumptions (•) and criteria (○) for their fulfillment in real experiment are considered:

- one-dimensional heat transfer, isotherms of temperature field have form of plan parallel plains perpendicular to the direction of heat propagation,
  - ○ influence of a heat loss from the specimen surface should be negligible,
- the outer parts (I, III) of specimen setup have infinite thickness,
  - ○ thermal thickness of the outer parts (I, III) should be enough thick corresponding to time length of the temperature response,
- an infinitesimal thickness of the heat source with the same thermophysical properties as the specimen,
  - ○ a heat capacity of the heat source should be negligible in comparison with heat capacity of the specimen volume involved,
- an ideal thermal contact between the heat source, the thermometer and the specimen,
  - ○ the thermal contact resistance should be negligible in comparison to thermal resistance of the specimen parts,
- negligible mass of the thermometer,

- a time constant of the thermocouple should be sufficiently small in comparison to time rate of the temperature response.

An ideal model based on the previous assumptions is schematically depicted in Fig. 2, where a_1, c_1, ρ_1 and a_2, c_2, ρ_2 represent physical properties (thermal diffusivity, specific heat and density) of the outer parts (I, III) and the measured unknown part (II) of the specimen setup, h is the sample thickness (part II) and Q_1, Q_2 are individual parts of heat Q dissipating into the specimen by the heat source.

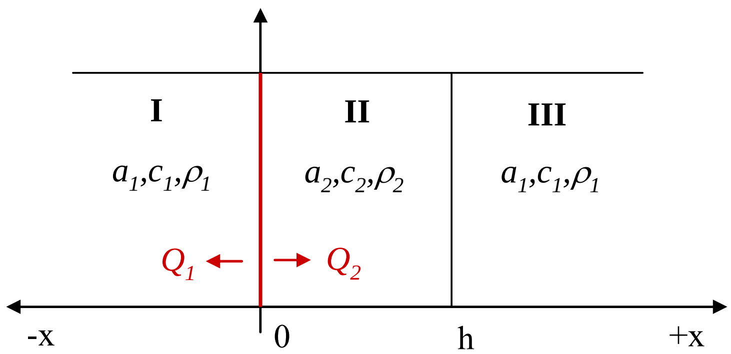

Fig 2 Schematic diagram of model of the Pulse transient method in sandwich-like specimen setup

The general one-dimensional heat conduction in the individual parts i has the following form

$$\frac{\partial T_i(x,t)}{\partial t} = a\frac{\partial^2 T_i(x,t)}{\partial x^2}, \qquad i = 1, 2, 3, \tag{1}$$

where T_i stands for the temperature and a is the thermal diffusivity a_1 for $i = 1, 3$ or a_2 for $i = 2$.

Solution of equations (1) has to fulfill following initial and boundary conditions in accord with model:

$$T_i(x, t \to 0) = 0, \qquad i = 1, 2, 3, \tag{2}$$

$$-\lambda_i \int_0^{t_0} \frac{\partial T_i(x=0,t)}{\partial x} ds = Q_i, \qquad i = 1, 2, \tag{3}$$

$$Q = Q_1 + Q_2, \tag{4}$$

$$T_1(x \to -\infty, t) = T_3(x \to \infty, t) = 0, \tag{5}$$

$$T_1(x=0,t) = T_2(x=0,t), \tag{6}$$

$$T_2(x=h,t) = T_3(x=h,t), \tag{7}$$

$$\lambda_2 \frac{\partial T_2(x=h,t)}{\partial x} = \lambda_1 \frac{\partial T_3(x=h,t)}{\partial x}, \tag{8}$$

where Q is a heat supplied by the heat source, t_0 is duration of heat pulse and λ is the thermal conductivity defined by well-known relation

$\lambda = ac\rho$. (9)

Using Laplace transform the temperature functions $T_{di}(x, t)$ for $i = 1, 2, 3,$ conforming the equation of heat conduction (1) and fulfilling conditions (eq. 2 – 8) are found for the heat generated in form of the Dirac pulse ($t_0 \to 0$)

$$T_{d1}(x,t) = \frac{Q}{k_2\sqrt{\pi t}}\left[\exp\left(-\frac{x^2}{4a_1 t}\right) + \left(\frac{k_1^2}{k_2^2} - \frac{k_1}{k_2}\right)\sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n}\exp\left(-\frac{\left(-x+2(n+1)h\sqrt{a_1/a_2}\right)^2}{4a_1 t}\right)\right]$$

for $x \in (-\infty, 0\rangle$, (10)

$$T_{d2}(x,t) = \frac{Q}{k_2\sqrt{\pi t}}\left[\exp\left(-\frac{x^2}{4a_2 t}\right) + \frac{k_1}{k_2}\left[\frac{k_1}{k_2}\sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n}\exp\left(-\frac{(x+2(n+1)h)^2}{4a_2 t}\right) - \sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n}\right.\right.$$

$$\left.\left.\exp\left(-\frac{(x-2(n+1)h)^2}{4a_2 t}\right)\right]\right] \qquad \text{for } x \in \langle 0, h\rangle, \quad (11)$$

$$T_{d3}(x,t) = \frac{Q}{k_2\sqrt{\pi t}}\left[\exp\left(-\frac{\left(x-h+h\sqrt{a_1/a_2}\right)^2}{4a_1 t}\right) + \frac{k_1}{k_2}\left[\frac{k_1}{k_2}\sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n}\right.\right. \quad (12)$$

$$\left.\left.\exp\left(-\frac{\left(x-h+(2n+3)h\sqrt{a_1/a_2}\right)^2}{4a_1 t}\right) - \sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n}\exp\left(-\frac{\left(x-h+(2n+1)h\sqrt{a_1/a_2}\right)^2}{4a_1 t}\right)\right]\right]$$

for $x \in \langle h, \infty)$,

where the constants k_1 and k_2 are defined by

$k_1 = c_1\rho_1\sqrt{a_1} - c_2\rho_2\sqrt{a_2}$, (13)

$k_2 = c_1\rho_1\sqrt{a_1} + c_2\rho_2\sqrt{a_2}$. (14)

Integration of the temperature functions $T_{di}(x, t)$ eq. (10 – 12), valid for the Dirac pulse, over time domain [4] leads to the solution of temperature functions $T_{si}(x, t)$ for $i = 1, 2, 3,$ for a heat generated in form of the Step-wise function, where instead heat Q the constant heat flux q is used.

$$T_{s1}(x,t) = \frac{2q\sqrt{t}}{k_2}\left[i\Phi_c\left(\frac{x}{2\sqrt{a_1 t}}\right) + \left(\frac{k_1^2}{k_2^2} - \frac{k_1}{k_2}\right)\sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n} i\Phi_c\left(\frac{-x+2(n+1)h\sqrt{a_1/a_2}}{2\sqrt{a_1 t}}\right)\right]$$

for $x \in (-\infty, 0\rangle$, (15)

$$T_{s2}(x,t) = \frac{2q\sqrt{t}}{k_2}\left[i\Phi_c\left(\frac{x}{2\sqrt{a_2 t}}\right) + \frac{k_1}{k_2}\left[\frac{k_1}{k_2}\sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n} i\Phi_c\left(\frac{x+2(n+1)h}{2\sqrt{a_2 t}}\right) - \sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n}\right.\right.$$

$$i\Phi_c\left(\frac{2(n+1)h-x}{2\sqrt{a_2 t}}\right)\Bigg]\Bigg] \qquad \text{for } x \in \langle 0, h\rangle, \tag{16}$$

$$T_{s3}(x,t)=\frac{2q\sqrt{t}}{k_2}\left[i\Phi_c\left(\frac{x-h+h\sqrt{a_1/a_2}}{2\sqrt{a_1 t}}\right)+\frac{k_1}{k_2}\left[\frac{k_1}{k_2}\sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n} i\Phi_c\left(\frac{x-h+(2n+3)h\sqrt{a_1/a_2}}{2\sqrt{a_1 t}}\right)\cdot\right.\right.$$

$$\left.\left.-\sum_{n=0}^{\infty}\left(\frac{k_1}{k_2}\right)^{2n} i\Phi_c\left(\frac{x-h+(2n+1)h\sqrt{a_1/a_2}}{2\sqrt{a_1 t}}\right)\right]\right] \tag{17}$$

for $x \in \langle h, \infty)$.

The function $i\Phi_c(x)$ is defined by

$$i\Phi_c(x)=\frac{1}{\sqrt{\pi}}e^{-x^2}-x\Phi_c(x), \tag{18}$$

where $\Phi_c(x)$ is a complementary error function [4].

Finally, the solution $T_{pi}(x, t)$ involving the heat pulse width t_0 one can obtain by subtraction of the functions $T_{si}(x, t)$ eq. (15 – 17) for time t and t-t_0 resulting in

$$T_{pi}(x,t)=T_{si}(x,t)-T_{si}(x,t-t_0) \qquad i = 1, 2, 3. \tag{19}$$

All infinite series stated above converge very well just for several elements.

3 Analysis of temperature function

An ideal temperature response (shown in Fig. 3) is calculated from the function T_{p2} ($x = h$, t) for thermometer position between the second and the third part of the specimen setup. In order to inspect a time interval in which the temperate function is sufficiently sensitive to change of the thermophysical parameters a, c, the sensitivity coefficients β_a, β_c and its linear dependency γ are employed. The reduced sensitivity coefficient β_p is given by [5]

$$\beta_p(t)=p\frac{\partial T(t)}{\partial p}, \tag{20}$$

where p is a parameter for which the temperature response $T(t)$ is analyzed. The linear dependency of the sensitivity coefficients β_a, β_c is then simply defined as

$$\gamma(t)=\beta_a/\beta_c\,. \tag{21}$$

The reduced sensitivity coefficients and its linear dependency are plotted in Fig. 3 as a function of time. A time interval suitable for evaluation of the thermophysical parameters a, c is determined by position of both maxima of absolute values of the sensitivity coefficients $t_m^{\beta a}$, $t_m^{\beta c}$ and by increasing their linear dependency γ, which is characterized by its co-linearity with time axis. It's evident that the time interval appropriate for the thermophysical parameters evaluation should be at least up to the maximum value of β_c. For the case depicted in Fig. 3 the time interval is then from 0 to 6 s.

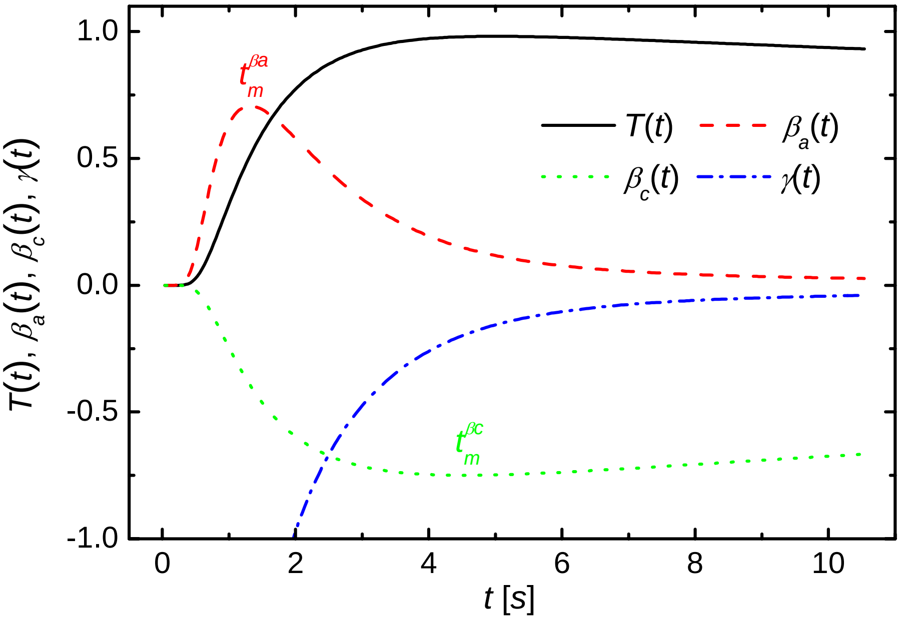

Fig 3 Temperature response $T(t)$, sensitivity coefficients of thermal diffusivity and specific heat $\beta_a(t)$, $\beta_c(t)$ and their linear dependency $\gamma(t)$ as functions of time t

Another way to analyze the temperature functions is from the point of view of its sensitivity on the thermophysical properties of the outer parts of specimen setup. In the next, two marginal cases of specimen setup are analyzed: insulator – conductor – insulator and conductor – insulator – conductor. The temperature distributions in each part of the specimen setup are shown in Fig. 4 and 5 for two different times of evolution.

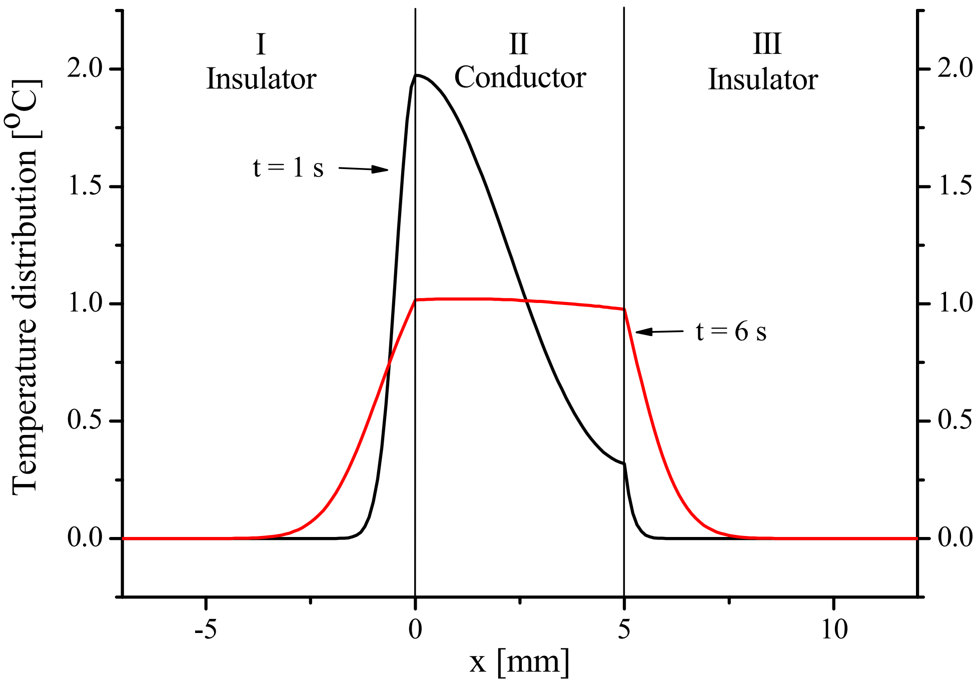

Fig 4 Temperature distributions in specimen setup insulator – conductor – insulator

In the case of specimen setup insulator – conductor – insulator shown in Fig. 4 a heat penetrates particularly into the middle part (conductor). After some time the temperate field inside the conductor is uniform and this second part starts to serve as a heater for neighboring outer parts. From this point, the temperature response is useless to characterize the middle part of specimen and further temperature recording is not needed.

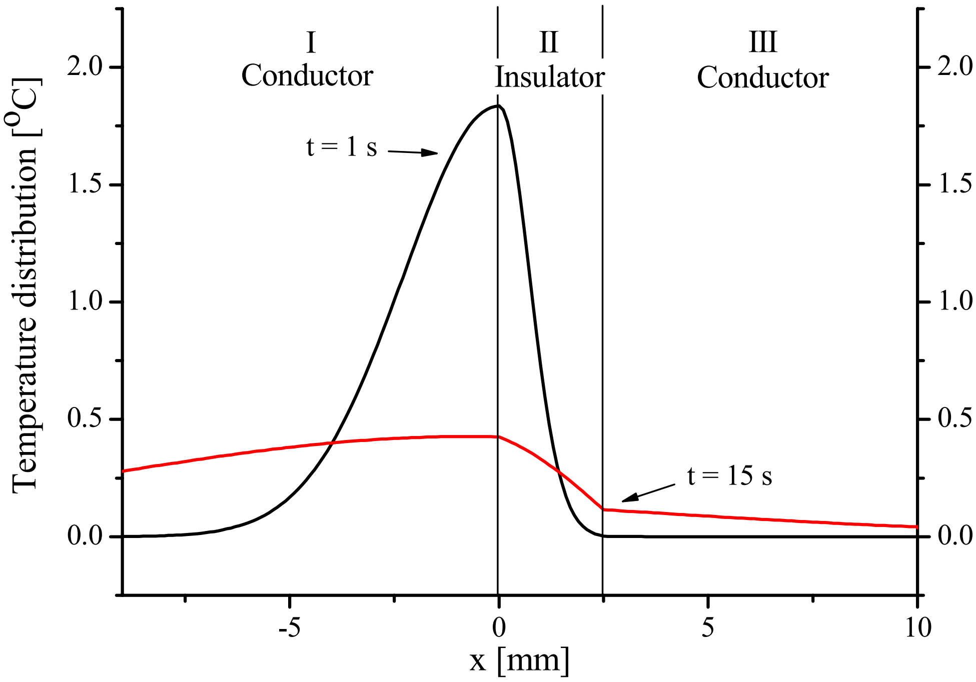

Fig 5 Temperature distribution in specimen setup conductor – insulator – conductor

In the case of specimen setup conductor – insulator – conductor shown in Fig. 5 a heat penetrates particularly into the first part (also conductor). The measured middle part of the specimen setup affects then as a heat barrier between both conductors. For longer times of a temperature response the spreading temperate field inside the first part of the specimen setup can reach its boundary at x = -10 mm. One has to choose enough thermally thick first part as well as enough thermally thin second part to avoid this break of the model assumptions including infinitive thickness of the outer parts as seen on the left side in Fig. 5 for t = 15 s.

The normalized sensitivity coefficients were used in order to analyze the sensitivity of temperature function on the thermophysical parameters change in dependence on the properties of the outer parts of the specimen setup. 3D graphs in Fig. 6 and 7 represent an influence of thermal properties of the outer parts on these normalized sensitivity coefficients of thermal diffusivity β_{na} and specific heat β_{nc}, where a heat capacity is a product of the specific heat times the density. The normalized sensitivity coefficient is defined as a product of the maximal value of sensitivity coefficient (eq. (20)) divided by a value of the temperature response in time of that maximum $t_m^{\beta p}$ (look in Fig. 3). The resulting relation has a form

$$\beta_{np} = \frac{p}{T(t_m^{\beta p})} \frac{\partial T(t_m^{\beta p})}{\partial p}, \tag{22}$$

where p is a parameter for which the temperature response $T(t)$ is analyzed. This sensitivity coefficient is truly dimensionless and thus it does not depend on size of

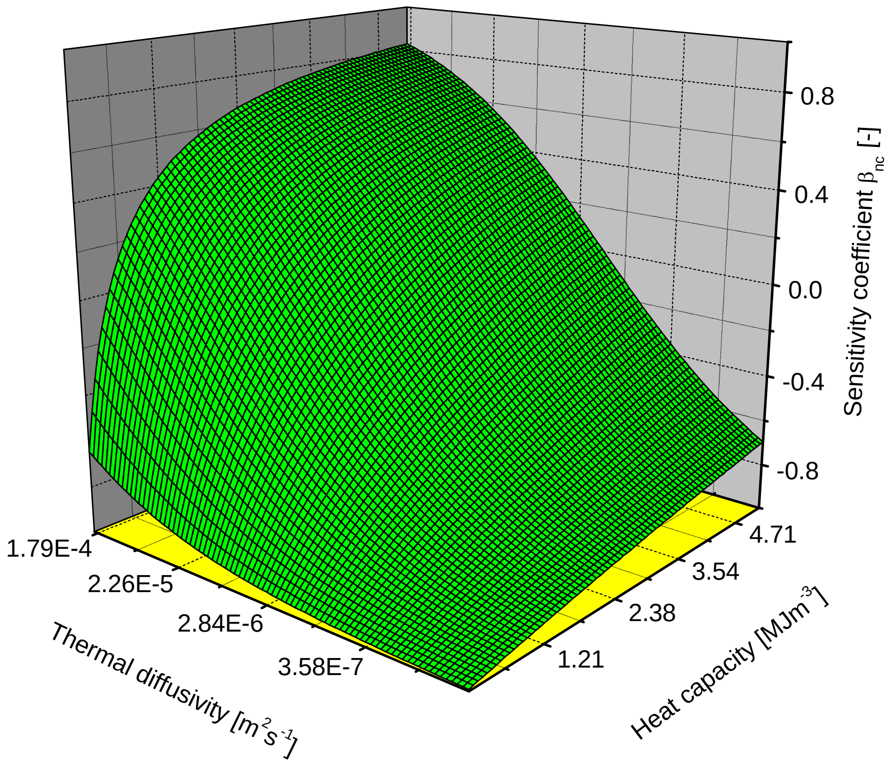

Fig 6 Dependence of normalized sensitivity coefficient of specific heat β_{nc} on the thermophysical properties of the outer parts of the specimen setup

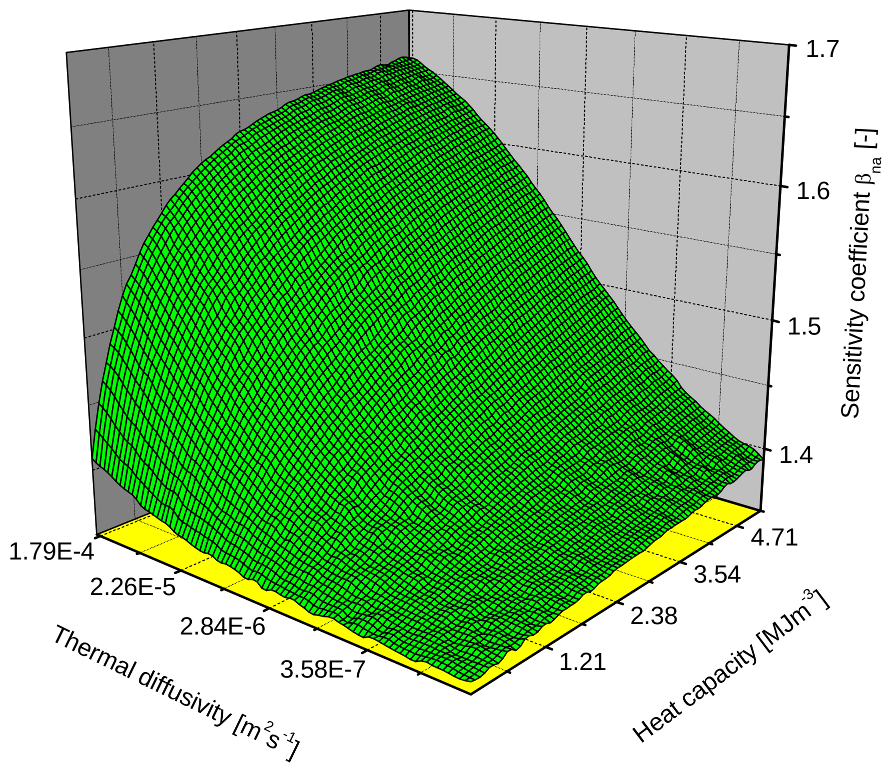

Fig 7 Dependence of normalized sensitivity coefficient of thermal diffusivity β_{na} on the thermophysical properties of the outer parts of the specimen setup

temperature response or analyzed parameter. In this meaning the normalized sensitivity coefficient is a relative change of analyzed function on a relative change of its parameter.

The 3D graph in Fig. 6 shows the sensitivity coefficient of the specific heat may reach zero values, what means that the specific heat of the measured sample can not be determined from the temperature response for whole range of the thermophysical properties of the outer parts of the specimen setup. These zero sensitivities connected with certain combinations of thermal diffusivity and heat capacity of the outer parts are depicted in Fig. 8 (solid black line) as a two-dimensional representation of 3D graph from Fig. 6 for $\beta_{nc} = 0$. To avoid zero sensitivity of the specific heat, the thermophysical properties of the outer parts have to be chosen as much as possible away from the black solid line. Here, a red cross denotes the thermophysical parameters of outer parts are the same to those of the sample (middle part). Since it lies on the zero sensitivity line (solid black) it is clear, that the outer parts of specimen setup made of the same material as the measured sample are unsuitable for the specific heat measurement. A dashed red line in Fig. 8 represents such combinations of thermal diffusivity and specific heat of the outer parts, in which their thermal conductivity (product of eq. 9) is the same to the thermal conductivity of measure sample. The line of equal thermal conductivities of the outer parts and the measured middle part nearly fits the line of zero sensitivity of specific heat.

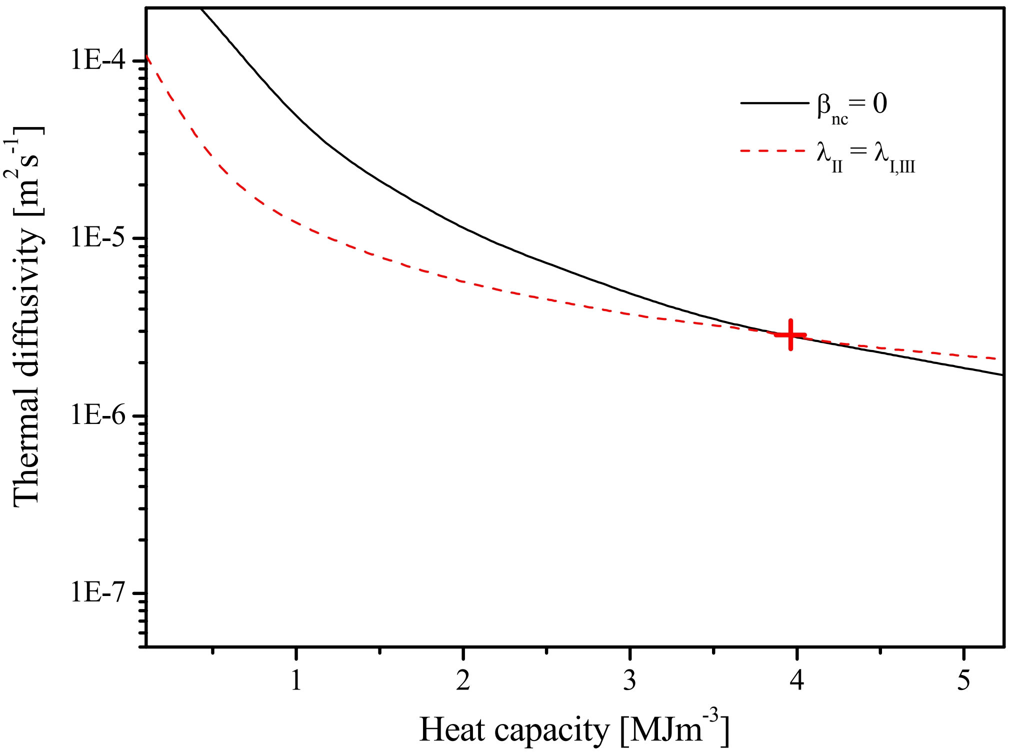

Fig 8 Dependency of thermal diffusivity on specific heat of the outer parts of the specimen setup for zero specific heat sensitivity $\beta_{nc} = 0$ (black solid line) and for equal thermal conductivity with middle part (red dashed line)

This attribute of the presented method can be elucidated as follows. A decrease of heat capacity of the middle part decreases a heat flow through it and thus increases a temperature. A decrease of the heat flow in the third (outer) part decreases then the temperature (gradient). The opposite processes should be balanced for certain values of the thermophysical parameters (Fig. 6 and 8) in the interface of both parts (position of thermometer) and thus no changes in temperature response are noticeable. A fitting algorithm can not converge in such situation.

All theoretical analyses presented in this section were calculated for sample made of stainless steel (middle part) with thickness h = 5 mm, pulse width t_0 = 0.2 s and heat flux q = 125 kJ/m^2 and for sample reference made of PTFE (outer parts) if not mentioned else. As expected, the calculation for another thickness h = 7 mm fitted the results shown in Fig. 6 – 8 very accurately.

4 Experiment

4.1 Experimental set-up and conditions

The experiment was carried out on ceramic silicon carbide SiC with density 3242 kg/m^3. The sample was prepared in form of a cylinder with diameter of 15 mm and thickness h = 2.84mm.

The outer parts of the specimen setup were made of PTFE (known as Teflon) with density 2158 kg/m^3, diameter 15 mm and thickness 15 mm. Their thermophysical properties, the thermal diffusivity 1.095x10^{-7} m^{2}s^{-1} and the specific heat 1209 J/kgK, were measured by Pulse transient method.

The instrument RT 1.02 (Institute of Physics SAS) is used for measuring the thermophysical properties. Basic scheme of the instrument is shown in Fig. 9. Thermostat in connection with the plate heat exchangers establishes the specimen temperature. An isothermal measuring regime with an isotherm within the limit of 0.02 K was used. A programmable current source KEPCO was used for generation of a heat pulse using the plane electrical resistance of 2 Ω . The planar heat source was made of a cooper foil of 20 μm etched in a form of a meander. A chromel-alumel thermocouple with thickness 40 μm was used as a thermometer. The temperature response was scanned by Keithly multimeter. A PC computer synchronizes all units. The typical parameters of the temperature response were T_m ~ 1 K and t_m ~ 1 s. The measurements were performed at room temperature and air atmosphere.

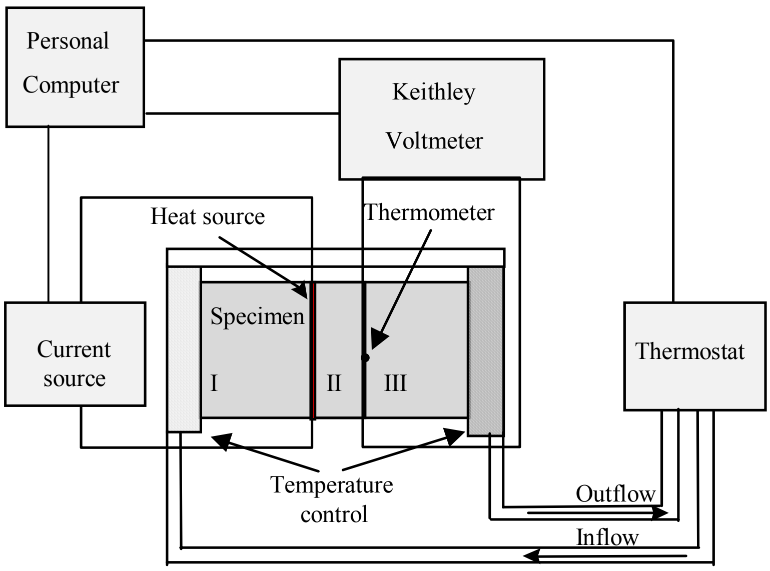

Fig 9 Basic scheme of experimental apparatus

4 Results

The thermophysical parameters of ceramic SiC measured by pulse transient method in sandwich-like specimen setup are stated in Table 1., where the values measured by standard methods (Flash method for thermal diffusivity and DSC for specific heat) are in the last column. A comparison between the values of Pulse transient method in sandwich-like specimen setup and standard methods shows an excellent agreement for all parameters. The reference value of thermal conductivity was simply calculated from Flash and DSC values using eq. 9.

Table 1. The thermophysical parameters of ceramic SiC and comparison

Parameter	Pulse transient in sandwich-like setup	Comparison
Thermal diffusivity $x10^{-6}$ [m^2s^{-1}]	21.2	21.8 Flash
Specific heat [$Jkg^{-1}K^{-1}$]	672	670 DSC [6]
Thermal conductivity [$Wm^{-1}K^{-1}$]	46.2	47.4

5 Conclusions

The complete one-dimensional analytical solutions of a temperature distribution in each part of the sandwich-like specimen setup were carried out according to a heat produced in form of the Dirac or the Step-wise function. An analysis of the solution found was performed considering the time evolution and the thermophysical properties of the outer parts of the specimen setup (Fig. 3 – 8). A limitation of these temperature functions in view of their use in thermal diffusivity and specific heat determination was found and discussed. It could be summarized in general as: the bigger difference between the thermal conductivities of the middle and the outer pars of the specimen setup, the accurate specification of the specific heat.

The measurement and comparison of the thermophysical parameters of ceramic SiC were performed in order to roughly verify the presented model of the sandwich-like specimen setup. A satisfying result was found for all measured thermophysical parameters (Table 1).

Nevertheless, additional experimental investigation and mathematical analysis of thermal contact's influences between each parts of specimen setup should be carried out.

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