OPTIMISATION METHODOLOGY FOR THE PULSE TRANSIENT METHOD AND ITS APPLICATION AT THE MEASUREMENT OF THERMOPHYSICAL PROPERTIES OF MATERIALS

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Abstract

Measurement regime analysis and the analysis of experimental geometry of the specimen for the pulse transient technique are presented. Optimization of the measurement of thermal diffusivity, thermal conductivity and specific heat is analyzed for building materials like autoclaved aerated concrete (AAC), calcium silicate, glass BK7 and polymer PMMA. The disturbance effects connected with specimen geometry and the heat source are discussed.

Key words: pulse transient technique, thermal diffusivity, thermal conductivity, specific heat

1 Introduction

The thermophysical properties of materials are of high importance for their use in industry. We should know the stability of measured parameters under various experimental conditions to develop suitable materials for specific industrial use. Quasi-equilibrium or non-equilibrium thermodynamic states induced into the material structure during manufacturing could cause different properties of the same material. Thus, the thermodynamical state of produced material should be a measure of the quality and stability of material. The practice demands this kind of measurements to predict their behavior in real conditions. The detailed study of several classes of materials was done. The measured results discussed in this paper were collected during several years in basic and applied research.

The primary observations when some of the specimens come to a laboratory show some discrepancies when the geometry of the specimen was insufficient and the basic criterion of infinite large specimen was not possible to fulfill. Then the values of thermophysical properties were shifted to lower values especially in the case of small and thin specimens. Thus, the analysis of the influence of specimen thickness was done for various materials and their geometry.

2 Pulse transient technique.

2.1. Principle of the experimental technique.

Pulse transient method for the measurement of thermophysical properties of several kinds of materials was used [1]. This method is measuring the specific heat, thermal diffusivity and thermal conductivity within a single measurement.

The principle of the Pulse transient method is simple. The heat pulse is generated by the passing of the electrical current through the plane electrical resistor (heat source) made of a nickel foil of 20 microns thick. The temperature response is measured by thermocouple on the distance h from the heat source.

The ideal model supposes non-limited specimen geometry. The real specimen arrangement and heat loss model is drawn in Fig. 1.
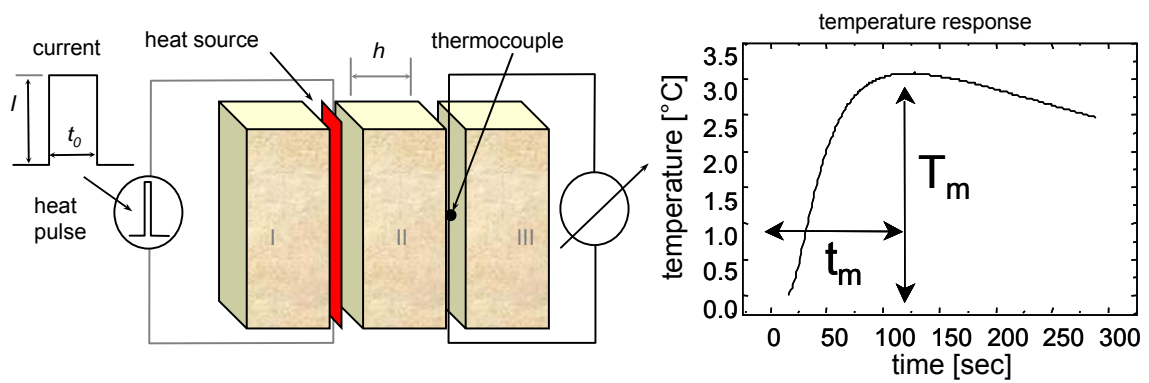

Figure 1. Arrangement of the specimen set that consists of three parts (left). The example of the temperature response for PMMA material is on the right. T_m and t_m are the maximum temperatures of the temperature response and the time when this maximum is set.

2.2 Theory

Temperature function for ideal model
The ideal case if experiment should fulfill several basic conditions. The heat source is infinitely thin and infinitely large having the same thermophysical properties like investigated material. The specimen is infinitely large also. The heat pulse is generated in the form of Dirac pulse with infinitely large instant heating power. Then, the temperature response to the heat pulse generated by planar heat source is described by temperature function [1]

$$T(x,t) = \frac{Q}{c_p \rho \sqrt{\pi a t}} \exp(-\frac{h^2}{4at})$$

(1)

The given function should be used for the fitting procedure to evaluate parameters thermal diffusivity a and specific heat at constant pressure c_p. For the simplicity of data treatment the next simple formulas for the maximum of the temperature response were derived

$$a = h^2/2t_m \tag{2}$$

$$c = \frac{Q}{\sqrt{2\pi e}\,\rho h T_m} \tag{3}$$

where $Q = RI^2 t_o$, $\textbf{\textit{R}}$ is the electrical resistance of the heat source, ρ is density, $\textbf{\textit{T}}_m$ is maximum temperature of the temperature response, $\textbf{\textit{t}}_m$ is time when temperature rich the maximum and other parameters are given in the Figure 1.

These simple formulas [1] are used for the standard evaluation of the experiment - one point evaluation.

Temperature function for model considering real pulse width
In the case when technically it is not possible to generate heat pulse of required energy that can rise up temperature response large enough for detection, the Dirac's pulse should be replaced by the pulse of lower instant power, but of longer duration (pulse width). This technique is usually used in the cases when big instant power can damage specimen, especially at the materials that could melt or burned easily like polymers, biomaterials etc.

Using model that accounting the heat pulse duration of several seconds, the thermophysical parameters are calculated from the parameters of the temperature response to the heat pulse [1].

$$T(h,t) = \frac{2 \cdot Q}{c\rho\sqrt{a}} \left[\sqrt{t} \cdot i\Phi^* \left(\frac{h}{2\sqrt{at}} \right) - \sqrt{t - t_0} \cdot i\Phi^* \left(\frac{h}{2\sqrt{a(t - t_0)}} \right) \right] \tag{5}$$

where

$$i\Phi^* = \frac{e^{-x^2}}{\sqrt{\pi}} - x \cdot erfc(x)$$

The next formulas were derived for the maximum of the temperature response.
The thermal diffusivity is given as

$$a = h^2/2t_m \cdot f_a \tag{6}$$

where

$$f_a = (t_m/t_0 - 1) \cdot \ln\left(\frac{t_m/t_0}{t_m/t_0 - 1} \right)$$

the specific heat,

$$c = \frac{Q}{\sqrt{2\pi e}\,\rho h T_m} f_c \tag{7}$$

where

$$f_c = 2 \cdot \exp(1/2)\sqrt{\pi \ f_a} \cdot t_m/t_0 \left\{ 1/\sqrt{\pi} \left[\exp(-f_a/2) - \sqrt{(t_m/t_0 - 1)/t_m} \exp(t_m/t_0 \ f_a/2(t_m/t_0 - 1)) \right] - \right.$$

$$\left. - \sqrt{f_a/2} \left[\Phi^* \left(\sqrt{f_a/2} \right) - ercf \left(\sqrt{t_m/t_0 \ f_a/2(t_m/t_0 - 1)} \right) \right] \right\}$$

and the thermal conductivity

$$\lambda = ac\rho \tag{8}$$

The symbols used are of the same meaning like in the case for ideal model. The time of the pulse duration (pulse width) is denoted as t_0, **erfc** is complementary error function.

Fitting evaluation procedure or derived one point evaluation procedure for maximum of the temperature response should be applied to a measured temperature response.

2.3 Real experiment

Differences in ideal and real experiment
In real life, the infinitely large specimen could not be produced and measured at all. In real experiment, the geometry is limited. The finite size of the specimen could not fulfill the conditions given by ideal model.
The ideal model is accounting the same thermophysical properties of the heat source as well as measured specimen and the negligible heat capacity of the heat source. In this case, the negligible effect was found when the heat source has at least ten times lower heat capacity than that of the specimen [1].
Next additional effects, like contact effects and heat losses from the specimen surface has to be included into physical model.
Contact effects and patterns of a heat source that is made in a form of metal meander produce non-uniform heat flux that penetrates into the specimen. The heat flux is homogenized after some time, e.g. at some distance from the heat source. Then the temperature response measured at short distances is influenced by this effect.
When the heat pulse generated by heat source is penetrating into the material, the heat flow is decreased by heat losses at the specimen walls. Heat losses effect takes place when the thickness of the specimen is too big comparing to the radius of the specimen. The heat flow transferred from specimen surface into surrounding atmosphere influences the maximum of the temperature response. To avoid this effect, the optimized specimen geometry and different way of data evaluation has to be used [2].
Under these circumstances, the simple model could undergo some revisions. All effects have to be involved in a more complicated model, however on the cost of more complicated evaluation procedure.
Simple model should be used providing the geometry of the specimen was manufactured according the optimized criteria. The next prerequisite condition is that during the recording of the temperature response the mentioned effects take no influence, otherwise at the data evaluation we have to skip some data at the end of experiment that was influenced by above-mentioned effect of heat losses.

Heat losses

The specimen geometry in real experiment is limited in space, so one has to set carefully the criteria that fulfill the conditions given by ideal model. No disturbance effects could influence the measurement. The geometry of the specimen have to be set in a way that during the experiment the heat losses from the specimen surface do not influence the measured temperature in the axial direction from the heat source (Fig. 2). This means that the heat pulse is penetrating into the specimen in planar form, e.g. which at the various points at the same distance from the heat source the temperature is uniform in radial direction. Towards the specimen edge, e.g. the specimen free surface, the heat loss causes the drop of the temperature. Even in this case one can get large flat region with planar isotherms along the specimen axis at not very large specimen thickness as it is drawn in figure 2. This effect is defined by the relation of the penetration depth of a measured material that is given by its thermal diffusivity and of the specimen radius and thickness.

The temperature drop caused by the heat losses from the specimen surface do not influence the measured temperature data used for the evaluation procedure in this region of specimen thickness. The thickness optimization procedure was used in several cases [2, 3, 4] that are summarized now in this paper.

The optimization was found during a series of measurements performed at different distances of the heat source from the thermocouple, e.g. the different thickness of a middle part of the specimen (Fig. 2). The optimization results specify the conditions when simple ideal model without influence of heat losses from the free specimen surface is possible to use. A more complicated model including heat losses from the specimen surface can be used on account of complicated mathematical calculations [1,2].
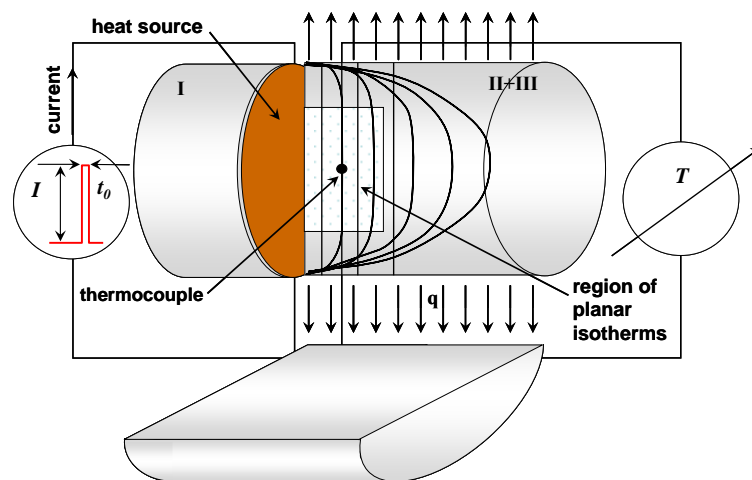

Figure 2. Model for heat losses from the specimen surface. The front of heat flux that is deformed by heat flow q=H(Ts-Ta) that is going out from the free specimen surface (Ts and Ta are surface and ambient temperatures and H is Heat loss coefficient. The region of planar isotherms is represented by white rectangular area.

The validity of the heat loss model was check on square shape of the specimens to get flat surface suitable for the infrared camera observation as there was observed an angular dependency of irradiation on cylindrical shape of the specimen. At the initial, the color given by infrared camera was black that prove the good temperature stabilization over the specimen surface. This color represents temperatures around 26.7 °C. The video camera

recorded infra pictures of the specimen surface during all the time of experiment. After some time, the heat pulse was spread out and some part of the heat has flow into the surrounding from the specimen surface. The situation after some decades of seconds is illustrated on Fig. 3. The yellow lines at Fig. 3 represent the specimen edges. The red line represents the heat source position. The recorded temperatures from marked nod points are on the left. Nod points are in the same distance from the heat source like the thermocouple. In this case the area of the nod N was heated up from 26.8 up to 28.1°C. The air out of the specimen at the surrounding of the heat source was heated up on 0.8°C.
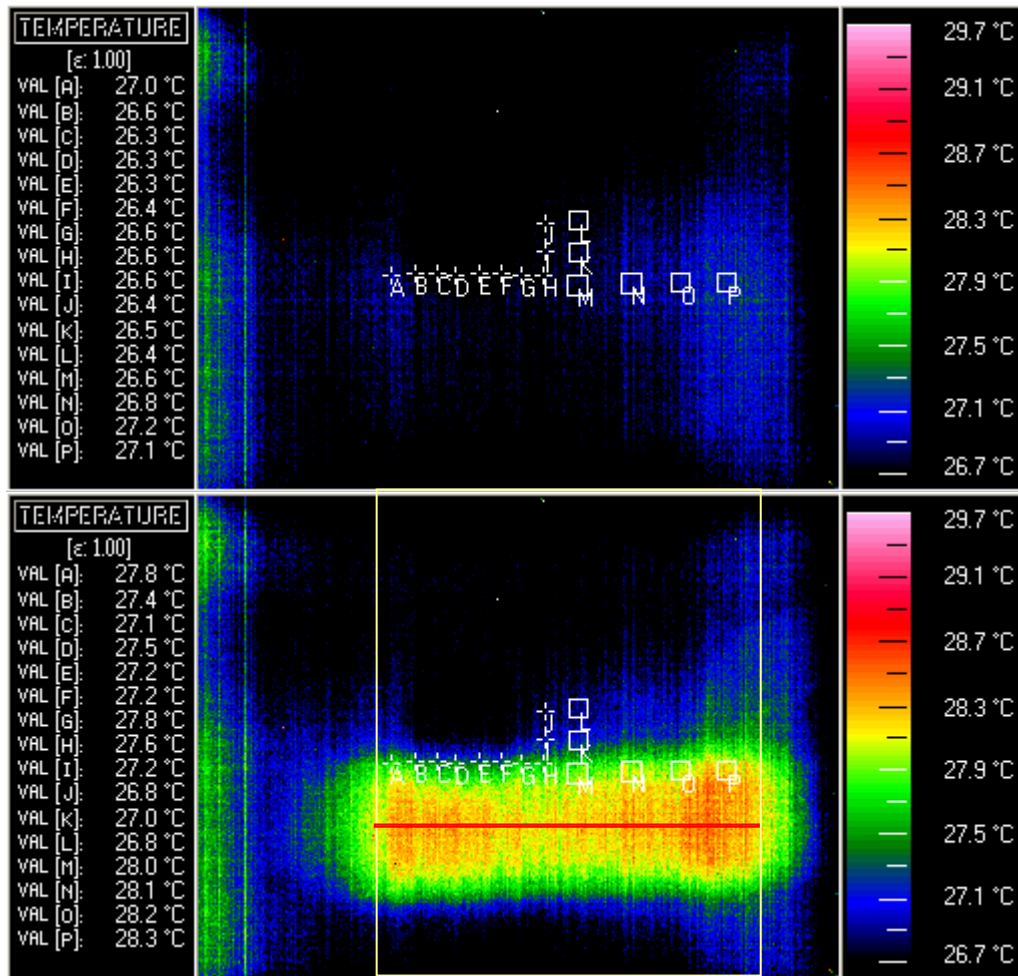

Figure 3. Picture of the heat losses from the specimen surface was taken before the heat pulse (up) and during the measuring the temperature response (down).

3 Results

The measurements of different materials having different size and geometry were done by pulse transient technique. It was found that the specimens having different thickness show differences in measured parameters.

In principle, the collected data should be divided into two main groups. As the first group, there were the first attempts to measure enough data to confirm that the described effects take place for a broad range of materials. In the second group are the data that

were collected more carefully and the effects influencing the measurements were studied in more detail. The main influences were observed for small and very large thicknesses of the measured materials.

The first measurements were performed on PMMA of different origin, aerated concrete and porofen. Later on more detailed study was dedicated to calcium silicate reinforced by cellulose fibers and finally again the PMMA, as we were involved into the intercomparison measurements where the PMMA was chosen as possible standard material. Thus, the research was concentrated on this material in more detail.

Some differences in data stability region that is already depicted on a given figures were found for different kind of heat sources. The first one is smaller with the active area dimension up to 50x50 mm and with the space between the metallic strip meanders 0.15 mm and strip width 2.357 mm produced at IP SAS. The big sources produced by MINCO Company with active area 146x146 mm have the space between the metallic strips 2.65 mm wide and metallic strips are 2.5 mm wide (Fig. 4 and 5). The effective area at the MINCO heat source is just 47.3% while the one of IP SAS is 94.3%.

There are differences in the data stability regions (free of mentioned effects) dependent on specimen thickness and diameter. It's because of difference of heat flow homogeneity produced by those two different sources. A heat source with smaller empty spaces between conductive metallic strips produces more homogeneous heat flow. Even in this case at smaller thickness of the material the heat flow produced by strips separated by empty spaces is homogenized after some time, e.g. at some distance from the heat source. This means at bigger thickness of material. It is practically the onset of data stability region – data are not influenced by heat source effect. For the big thicknesses, the heat loss effects take place. This effect is dependent on the specimen diameter. Both effects described deform the temperature response. Thus, the evaluated parameters are overestimated. The maximum temperature and time of the temperature response are increased or lowered. The illustration of used heat sources is in Fig. 4 and 5.
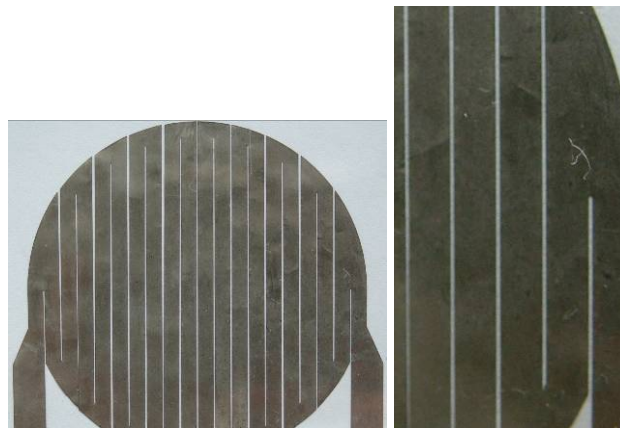

Figure 4. Heat source produced at IP SAS. The empty space between the strip is minimal.
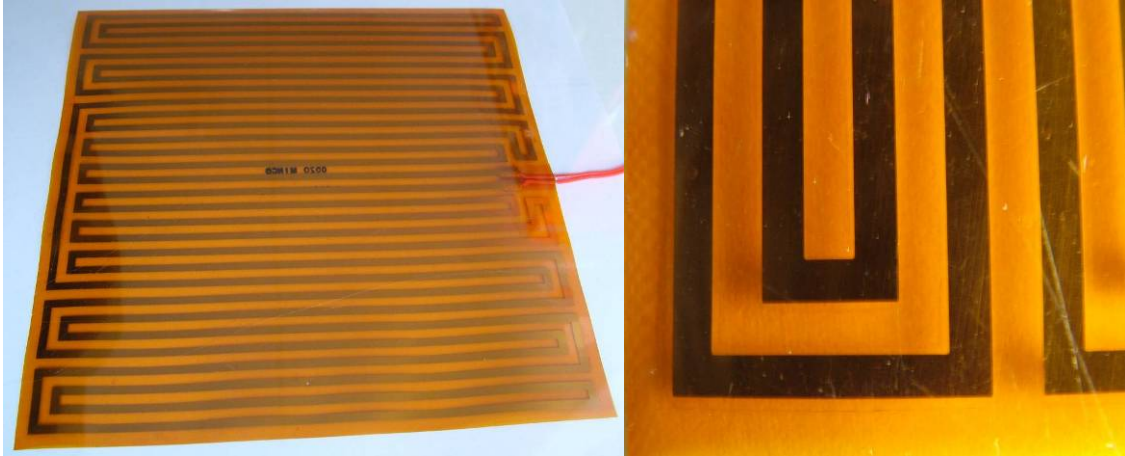
Figure 5. MINCO heat source. The detail of the ratio of the strip width and empty space.

Measured materials

Autoclaved aerated concrete – AAC (HEBEL).

For the measurement of AAC specimen blocks having cross section dimensions 150x150x40 mm were used. The material density was 586.1 kg m^{-3} and porosity 72%. For the comparison we found in literature the results of the authors M S Goual et al [5] that published the dependency of thermal conductivity of AAC on porosity (Fig. 6). Thermal conductivity calculated from the fit for 72% porosity is 0.1944 and was used for next comparison in Fig. 7. Measured materials in this reference were of different origin and thus we may expect some differences in their thermal behavior. The next parameters - specific heat and thermal diffusivity were not included in this reference.
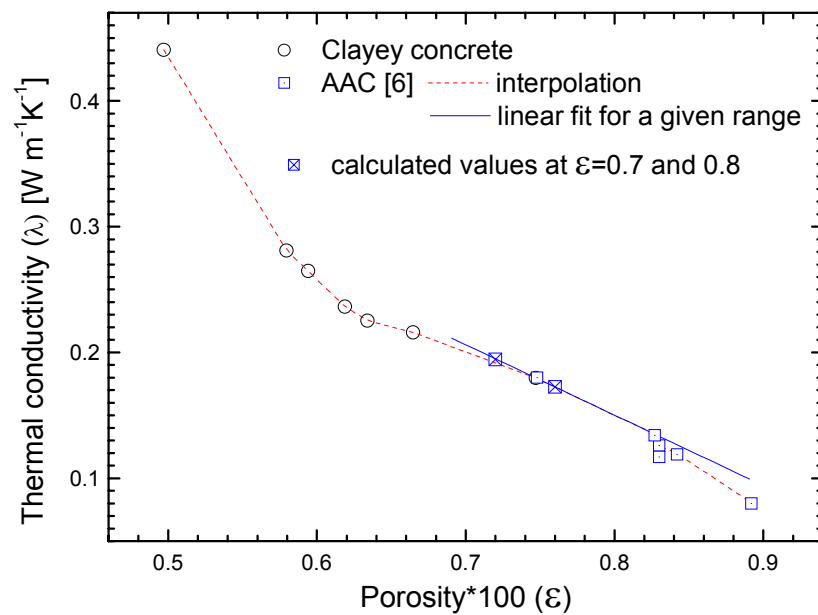

Figure 6. Dependency of thermal conductivity on porosity. M S Goual et al.[5].

For the measurements of AAC blocks by the Pulse transient technique the heat source MINCO having square dimensions 150x150 mm was used. The effect of non-homogeneous heat flow generated by this heat source is evident for small thickness (up to 10mm). The data stability region is in between 15 and 22 mm. The values of thermophysical parameters data above plateau values were measured for thickness above 25mm. The measured thermophysical parameters are on Fig. 7.
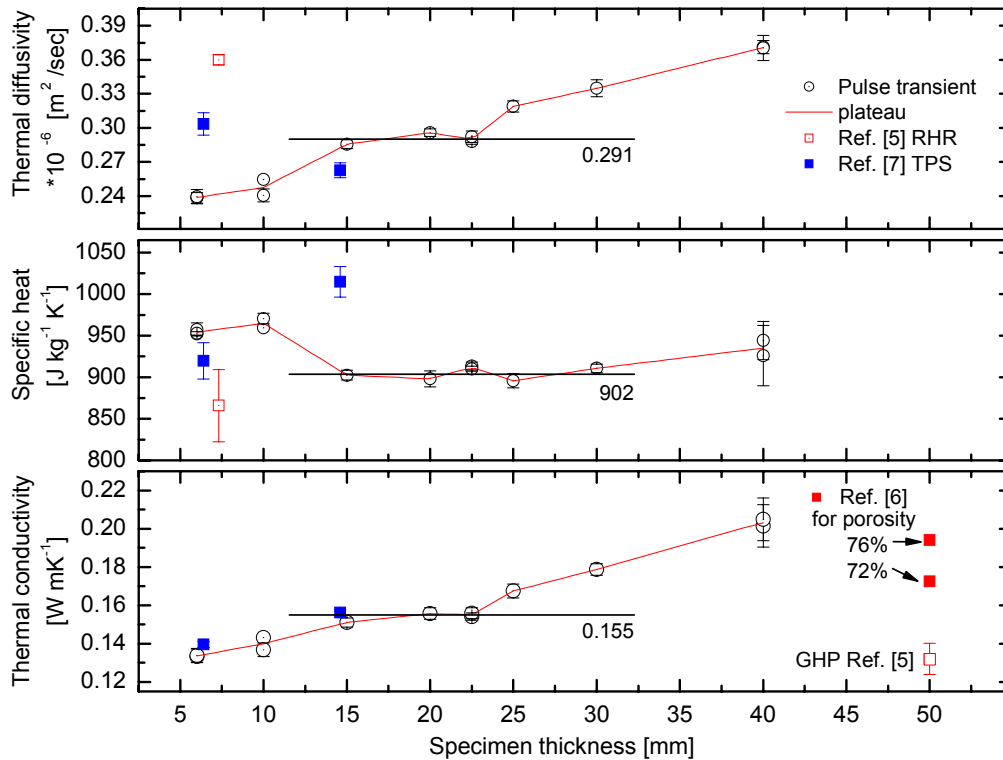

Figure 7. Thermophysical properties of autoclaved aerated concrete measured for different thicknesses of the specimens compared with data from literature.

Thermophysical data in Fig. 7 were compared with data measured by guarded hot plate method - GHP (thermal conductivity) regular heating regime method – RHR (thermal diffusivity and specific heat) [6] and transient plane source technique [7]. In the case of GHP method, the specimen dimension was 400x400x40mm and in the case of RHR method 75x75x75mm. The specimen size in ref. [5] was 100x100x100mm and in the case of ref.[6] we used the same specimen blocks as for the pulse transient method. The difference in graph should be caused by fact that the density in ref. [6] was 560 kg m^{-3} and porosity 76%. The next influence should be caused some moisture content in the case of ref. [6].

The main knowledge obtained from this first experiment was, that the measured thermophysical data were influenced by some disturbance effect that are depended on specimen thickness.

Calcium Silicate

A more detailed study was done for calcium silicate board material reinforced by cellulose fibers. Here we used squared shape heat source MINCO having dimension 150x150 mm like in the case of AAC. The material density was 280 kg m^{-3}.

The data were measured at 15 different thickness of the material (Fig. 8). The values of thermophysical parameters show clear plateau in between 15 and 25 mm of thickness scale (Fig. 8.) The data stability region is marked in yellow.

The measurements were done also in dry and slightly moisture state. The porous specimen was in moisture equilibrium up to a level given by surrounding air having stabile relative air humidity (30 and 60%). The saturation takes for several weeks.

The measured values of thermal conductivity at the material thickness 15 mm were compared here with the data measured by guarded hot plate method (GHP) at various moisture contents (Fig. 9).
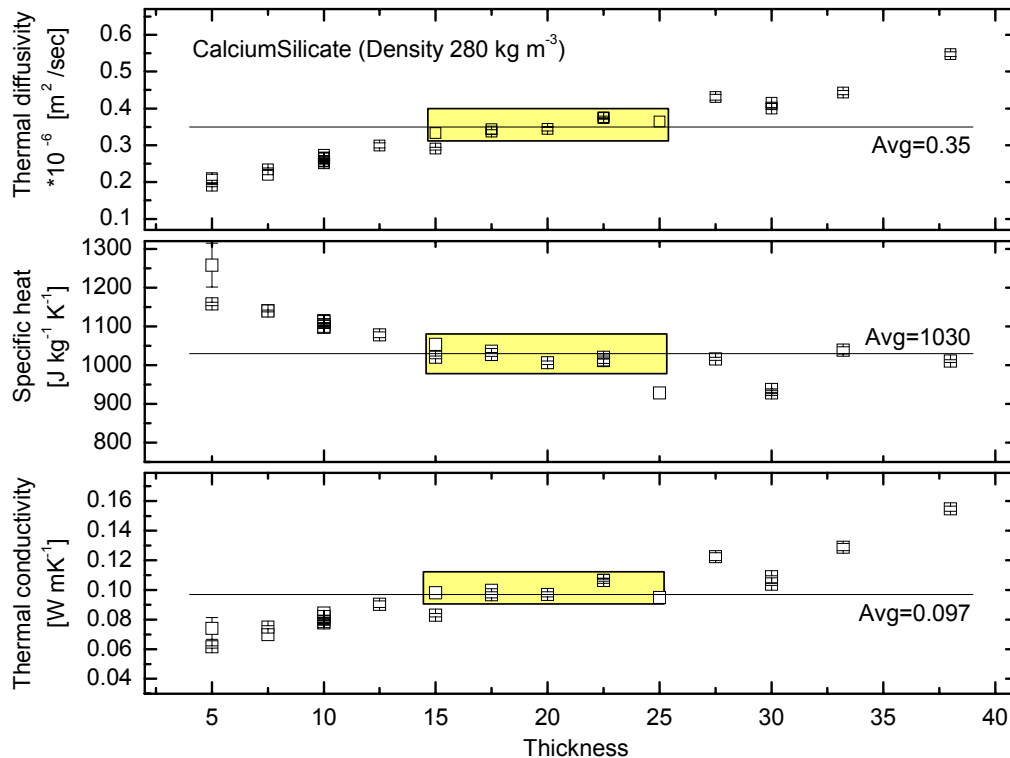

Figure 8. Thermophysical properties of calcium silicate boards reinforced by cellulose fibers (SilCal250) measured for different thicknesses of the specimen.

In the case of drying in normal atmosphere (GHP method) the values are higher than those one measured after evacuation and aeration (PTM), e.g. the vacuum drying process. The values of thermal conductivity measured by pulse transient method were compared with Guarded hot plate method. In Fig. 9 the measurements by pulse transient method started with virgin specimen having moisture content 0.6÷0.8 volume % that correspond to the saturation on air having relative humidity approximately 60 %.

Then the specimen was measured under the vacuum (lowest values). After those measurements, the specimen was aerated by air having relative humidity 30% (circle below the last square point that represents data in dry state measured by guarded hot plate method.

In the case of measurement by guarded hot plate method the drying process was not performed in vacuum, so the specimen should contain some residual moisture. Thus the data for dried material are of higher values than the ones for dried in vacuum. Later on, by storage of the material under the air condition, the value of thermal conductivity increases in time as the moisture sorption takes place (red arrow in Fig. 9). Compared

data for virgin specimen show high reliability for the thickness 15 mm within the data stability region in Fig. 8. Red arrow on Fig. 9 shows the way of increasing thermal conductivity in time when moisture penetrates back into the porous specimen.
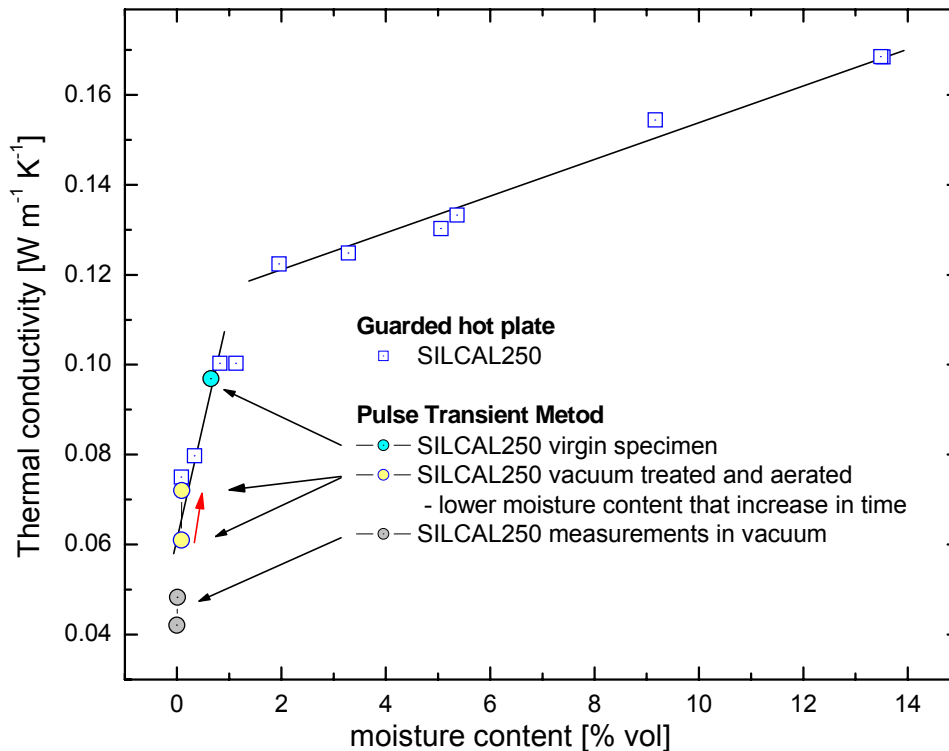

Figure 9. Thermal conductivity of calcium silicate measured by guarded hot plate method [8] and by pulse transient method for different moisture content.

Optical glass BK 7
Optical glass is homogeneous material and thus much smaller specimen is required. For the measurement the specimen having square dimension 50x50 mm was used. The density of BK 7 was 2510 kg m^{-3}. The difference regarding previous results on non-homogeneous materials is in different type of heat source (50x50 mm) made by IP SAS in cooperation with Institute of Informatics SAS. The empty spaces between the heat source strips are 20 μm that is negligible comparing those one from MINCO. Thus, the homogeneity of the heat flux is better and stabile plateau values of the measured thermophysical parameters can be reached at lower thicknesses. On the other hand, the smaller dimension in diameter influences the measurements due to heat losses from the sample surface at bigger thicknesses of the material. Thermophysical parameters were measured for different thicknesses (Fig. 10).

Polymethyl methacrylate - PMMA
PMMA is polymeric and optically transparent material. There were measured specimens of different origin. PMMA from NPL England and PMMA that was used as a construction material at IP SAS.
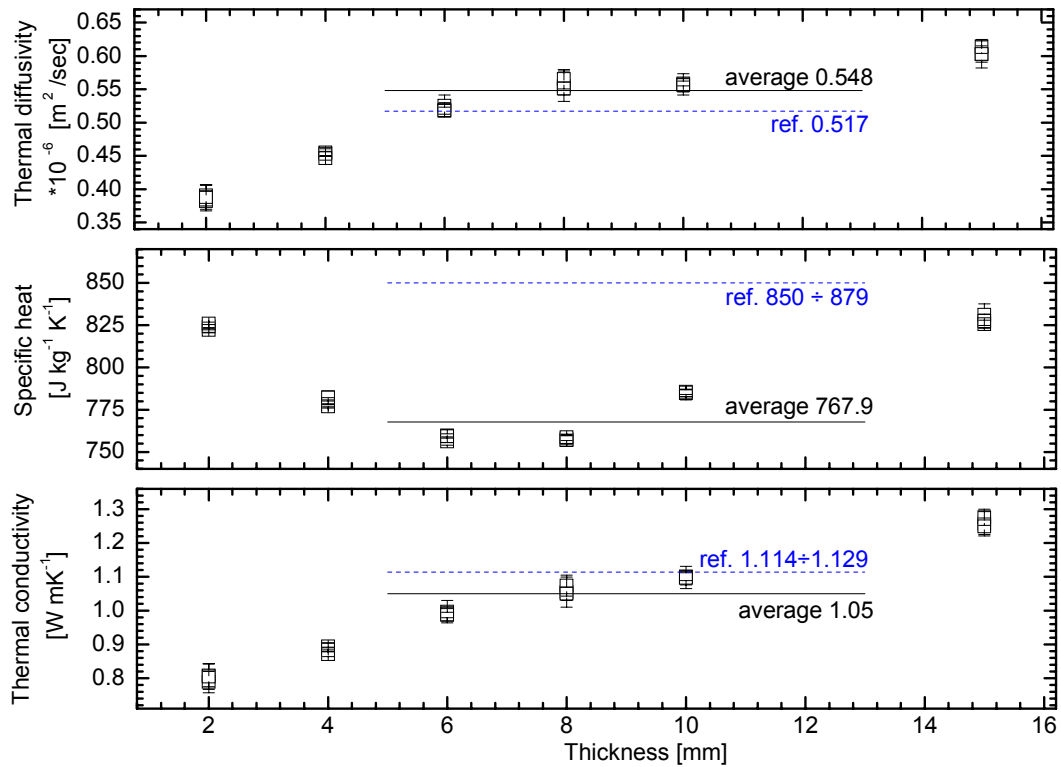
Figure 10. Thermophysical properties of optical glass BK 7 measured by heat source produced at IP SAS (square dimension 50 mm). The data stability region is in between 6 and 8 mm. The reference values for comparison were found in literature on web [9].

The sample sets had different diameters or cross sections – 20, 30 and 50 mm. The density of PMMA was 1182 kg m^{-3}. The plot of thermophysical properties vs. material thickness for the specimens having circle cross section 20 and 30 mm in diameter is in Fig. 11. It was clear that for smaller specimen diameter the plateau is not recognizable.

The values for the comparison were given by NPL. Thermal conductivity data were compared with the results of Koniorczyk [10] measured by guarded hot plate method on the specimens having square dimensions 500x500 mm. The data stability region measured by pulse transient method was found for the thickness 5 and 6 mm for the specimen diameter 30 mm. Similar values slightly above recommended values were measured by TPS method also.

A more detailed study was performed on specimens of IP SAS origin having diameters 30 and 50 mm. The plot is in Fig. 12. Measurements were performed with the heat sources produced at IP SAS. The plateau with data stability for 30mm specimen is between 6 and 8 mm and for the specimen with 50 mm diameter between 8 and 10 mm. Recommended values are depicted as a straight lines. The data represented by blue triangles were obtained by fitting evaluation method [2] that prolong data stability region. The fit was performed just for the onset of the temperature response.

In this case, it is evident that the bigger sample radius, the longer data stability region is observed.
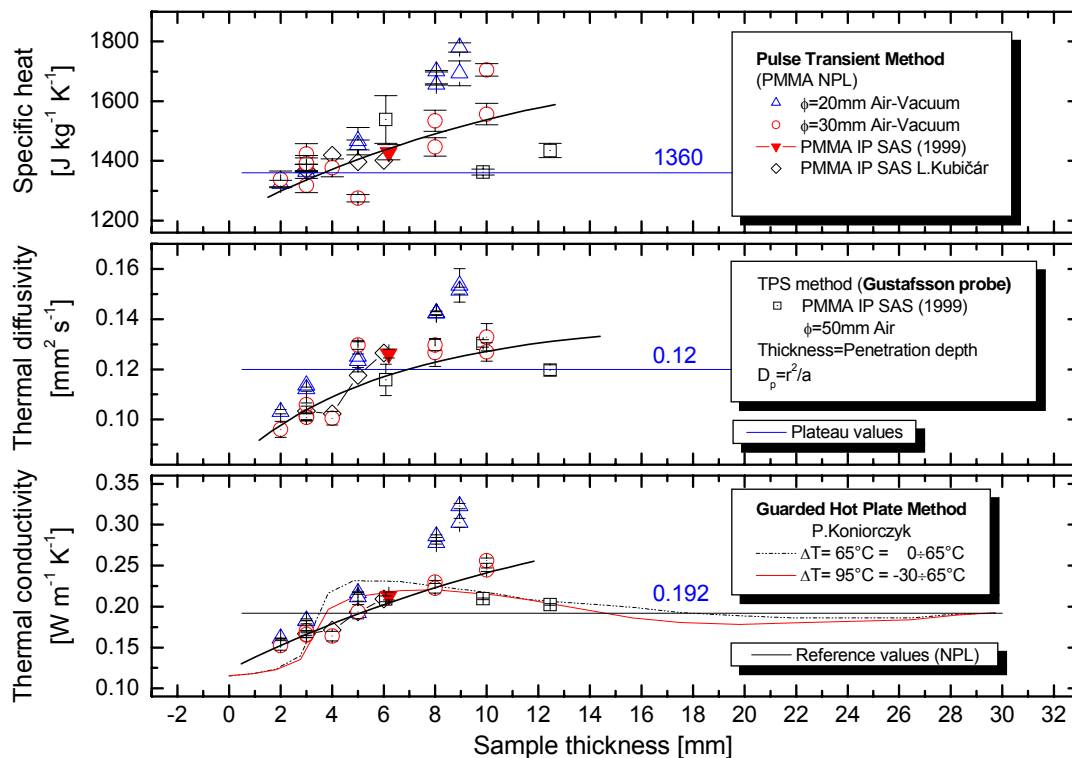
Figure 11. Thermophysical properties of PMMA vs. material thickness. The origins of the specimens are denoted in the legend.
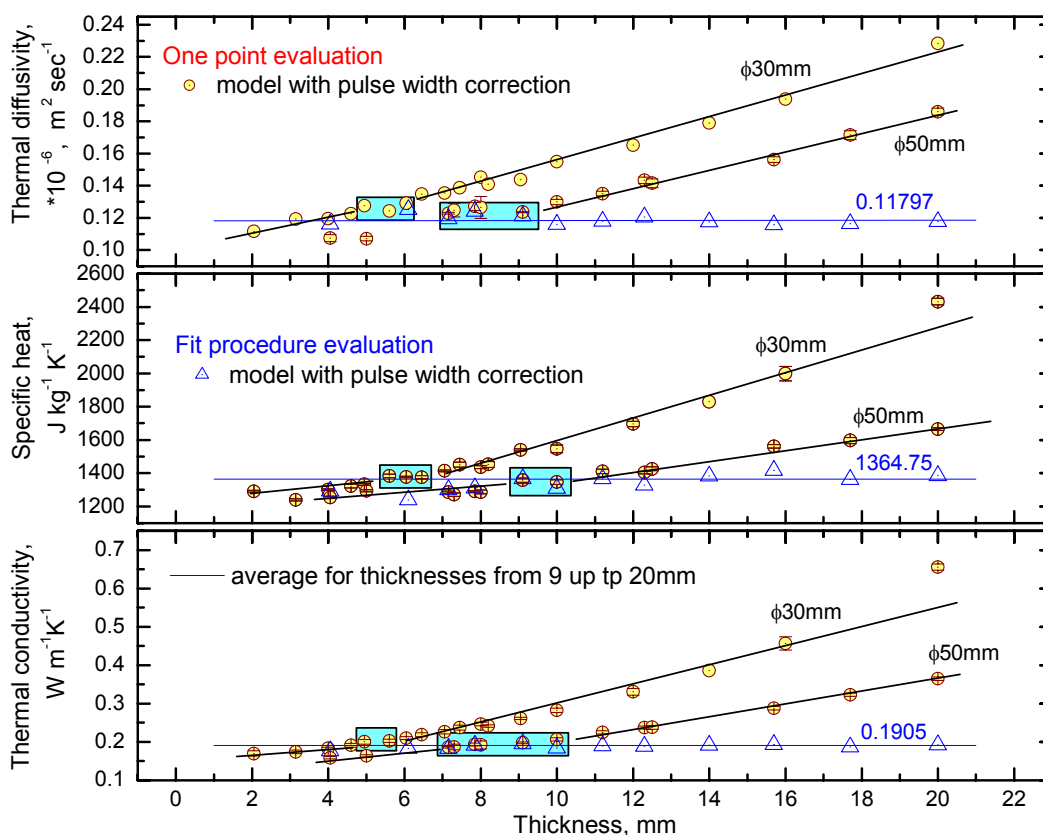

Figure 12. Thermophysical data of PMMA (IP SAS). Specimen diameter was 30 and 50 mm. The values drawn in blue triangles were calculated by fitting procedure [2].

Conclusions

The thermophysical properties of different kind of materials – homogeneous and non-homogeneous were measured.
The effects caused by the heat source design as well as the effects of the heat loss from the sample surface were discussed and shown on different types of materials. Measured thermophysical data of homogeneous as well as non-homogeneous materials shows similar behavior in the apparent dependency on the material thickness. The fact is that there are disturbance effects of the heat source as well as the heat loss effect from the sample surface that influence the measured data. Those disturbance effects are dependent on the specimen thickness and diameter.

Methodology of data evaluation in the case when disturbance effects influence the measurement is given.

The presented data show different data stability regions for the MINCO sources and the sources produced at IP SAS. The difference is in homogeneity of produced heat flow generated by metallic strips separated by various empty spaces.

Discussion of heat source effect as well as heat loss effect for small and big thickness of the material show the optimal thickness available for the measurement with minimal measurement error is given.

The presented methodology optimizes geometry of the specimen. This gives the possibility to design a specimen dimensions on the base of "guess" values of thermophysical properties of unknown specimen and one can conclude the specimen diameter (or square dimension) and its thickness to measure values in the data stability region.

For the routine measurement, the dimensions that correspond to data from data stability region have to be used. This general rule reduces the error of measured parameters.

This methodology is applicable to the ideal physical model that includes simple correction on real pulse width.

To include the effects of heat losses and evaluate data correctly for the specimen thickness above given optimized limit, a newly developed model will be used in the near future [11,12].

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