IOP Conf. Series: Materials Science and Engineering **15** (2010) 012064 doi:10.1088/1757-899X/15/1/012064

Analysis of influence of measurement conditions on repeatable results of thermal diffusivity of ceramic moulds designed for manufacturing the superalloys castings

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Abstract. The article presents the study results concerning development of methodology for measuring thermal diffusivity with the laser-flash method with special attention being paid to their repeatability. The first stage involved the literature review of the problem determining an influence of variable measurement parameters including presence of graphite coating, error correction methods and laser power. The main part of the study includes evaluation of the sample structure influence. A real ceramic mould of specific porosity and green compacts made after its grinding were examined. Influence of sample thickness and interval between individual laser shots were also determined. The analysis was performed by using the Netzsch LFA 427 instrument designed for direct measurement of thermal diffusivity with the laser-flash method. The material analysed included different types of ceramic moulds designed for manufacturing the elements made of nickel based superalloys. The preliminary study performed showed that the main factor that has an influence on obtaining correct and repeatable results of thermal diffusivity study is proper preparation of samples i.e. using them in a form of directly cut out from the mould, selection of data acquisition time and interval between individual shots. It is especially important in the case of measurements taken at high temperature when radiation mechanism of heat transfer is of basic significance.

1. Introduction

Laser radiation is characterized by uncommon properties, what makes these properties be a very useful tool in several investigating methods of contactless character. It enables to include the researching methods, based on usage of such radiation, to non-destructive methods. Various methods of laser microscopy [1] or the laser-flash method, named otherwise a laser impulse method is an excellent example in this question. This method is relatively simple and enables to determine some fundamental material parameters, which characterize thermal properties i. e. coefficient of thermal diffusivity (i. e. coefficient of temperature equalization) and indirectly coefficient of thermal conductivity and specific heat. The laser-flash (L-F) method was presented for the first time by Parker, Butler, Jenkins and Abbott from the Military Laboratory of Radiological Defence in 1960 [2,3]. It is one of the most popular methods in measurement of temperature conductivity of solid bodies and the same, the only standardized method in measurement of temperature conductivity in transition conditions.

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It is based on an analytical solution of the Fourier's equation with adiabatic boundary conditions for an infinite flat sample, exposed to action of the Dirac's thermal impulse. The L-F method was one of the first applications of laser radiation in metrology [4].

From beginning of origin, this method was developing in consideration of theory, methods in an analysis and error correction and possible applications as well [5]. Theoretical grounds of this method were worked out by Watt [6] and later verified by Taylor [7]. A direct effect of carried out measurement was to define thermal diffusivity i. e. coefficient of temperature equalization, which is obtained on a ground of an analysis of temperature changes, while the sample is not illuminated by a beam on its back side. Registered changes of temperature of the material are effects of absorption by front surface of a short laser radiation impulse, and obtained characteristics of temperature course is named a history of temperature. An effect of absorption, which initiated measurement of a laser impulse (in assumption, it is the Dirac's infinite short impulse) of actually selected parameters, is increase of temperature on front surface of a sample. The history of temperature, registered on back surface of a sample, describes changes of temperature in a function of time, which are effects of heat conductivity.

Considering very restrictive measuring requirements, practically it is impossible to realize it in laboratory conditions (e. g. requirements concerning excitation with infinite short energetic impulse – the Dirac's impulse), because results obtained directly on grounds of this theory, are encumbered with errors.

Fundamental requirements of a model, describing the heat transport, which are necessary to get ideal measuring conditions in the laser-flash method, can be classified as following [8]:

- infinite short time of the impulse;
- absence of heat loss (convectional losses and radiational losses as well);
- very small thickness of a sample, comparing to other dimensions (comparing to a diameter of a laser radiation beam);
- heat source, infinite thin, comparing to thickness of a sample;
- homogenous (isotropic) material;
- homogenous (in time and space) heat source.

Too much deviation from these conditions makes considerable errors originate in calculated values of thermal diffusivity. The following effects, not considered by this model, should be taken into consideration [8]:

- effects of finished time of impulse duration (laser impulse), occurring mainly for thin samples of materials, which are good thermal conductors. From calculations, delivered by Heckman [9], it results that a radiation impulse shorter 20 times than $t_{0.5}$ is quite enough to be these effects neglected;
- radiational heat losses from surface (especially front surface) from the sample – this effect occurs mainly in a case of massive samples, which are in high temperature or absorbing considerable heat amount (what makes increase in temperature in a layer, which is a heat source);
- convectional heat losses, taking place in a case of thick samples of materials with relatively small coefficient of temperature equalization, i. e. when time, which is necessary the back surface to achieve its max temperature, is very long;
- effects related to spatial inhomogeneity of surface source, while this inhomogeneity can be caused by inhomogeneity of laser radiation beam, uneven (rough or corrugated) front surface, inhomogeneous near-surface layer of material (pores, microcracks etc.);
- disturbances in temperature field, made by phase transformations;
- changes in shape of temperature history, made by transport of mass inside the sample (e. g. water vapour in porous substance);

- occurrence of "spongering" thermal capacities of a heat source and temperature sensor, which appear mainly for small samples of material of relatively small specific heat and high coefficient of temperature equalization.

In the paper [8], specification concerning researching works on correction of errors, is presented. In a case of radiational heat losses, methods of corrections were worked out by Cowan [10], Heckman [9], Clark and Taylor [11] Cape and Lehman [12] and James [13]. Investigations on this question are still continued (e. g. [14]). Occurrence of the effect of finished time of impulse duration was analyzed by Cape and Lehman [12] and Clark and Taylor [15]. Beedham and Dalrymple [16], MacKay and Schriempf [17] and Taylor [18] investigated influence of inhomogeneous surface heat source on a shape of temperature history and accuracy in determination of a coefficient of temperature equalization as well, and they stated that for correctness in achieved results, only homogeneity of a heat source in its central part is essential. The first theoretical phenomena of transport of a heat impulse, generated by laser, in inhomogeneous material were carried out by Kerrisk [19, 20], and he defined a criterion of homogeneity for materials tested by means of the L-F method.

2. Materials and methodology

A main purpose of these investigations was to define fundamental questions with methodology of evaluation of thermal diffusivity by the laser-flash method by use of the A427 apparatus produced by Netzsch Company and ceramic materials, of which ceramic elements of casting systems provided for elements made of nickel superalloys. Stand instructions, which define fundamental operations related to a way of carrying out the investigations, are a ground to perform the actions.

The carried out recognizing works proved that the following parameters were main factors, which influenced to get the credible and repeatable measurements of thermal diffusivity by the laser – flash method (besides standard procedures) by use of the LFA427 apparatus produced by Netzsch Company:

- thickness and shape of sample;
- correct selection of time between individual „shots" from laser;
- getting the actual layer of graphite on surface on a tested sample.

It is an obvious problem, in which a fundamental question is to obey the searching procedures, and above mentioned factors require only being more precise from a point of view of specificity of tested materials, which were ceramic materials, provided for casting moulds.

3. Results

Thickness of a sample is the first of the discussed factors, and it has got influence during investigations, what has very essential meaning. Application of samples approx. 99 mm thick is possible to be used because of technical possibilities of apparatus, however practically it was revealed that for ceramic materials of relatively good insulating properties, application of round or square samples is recommended, which are no more than 5mm thick (max thickness, unless there are technical possibilities to produce samples of less thickness). It results from a simple fact, that together with increase in thickness, measurement time increases vehemently (in extreme case even to 150 000 ms – in comparing to magnesium alloys, this time is approx. 1000 ms). In an effect, practically it is impossible to get stable initial conditions – reference (so called a base line, which should reveal line stability of stress for duration time corresponding to 25% total acquisition time). In an effect, a boundary condition is not met in order to perform a laser shot (impulse), of which role is to generate an elastic wave (measured quantity) and define time of its propagation through a sample (half-time). At so long time in measurement of stability in a base line, it is very difficult to get conditions, which enable to carry out an actual and repeatable measurement. Of course, it is possible to create such conditions, in which measurements are possible, however scatter in obtained results is very high. Application of samples of less thickness, predominantly makes shorter measurement time. Exemplary results for two different thicknesses are presented in figure 1. Almost triple increase in measurement time is visible, what is a consequence of bigger thickness of the sample. An analogue effect is observed in a case of too long measurement time. In this case, difficulties to match a mathematical model appear and obtained results are not precise.

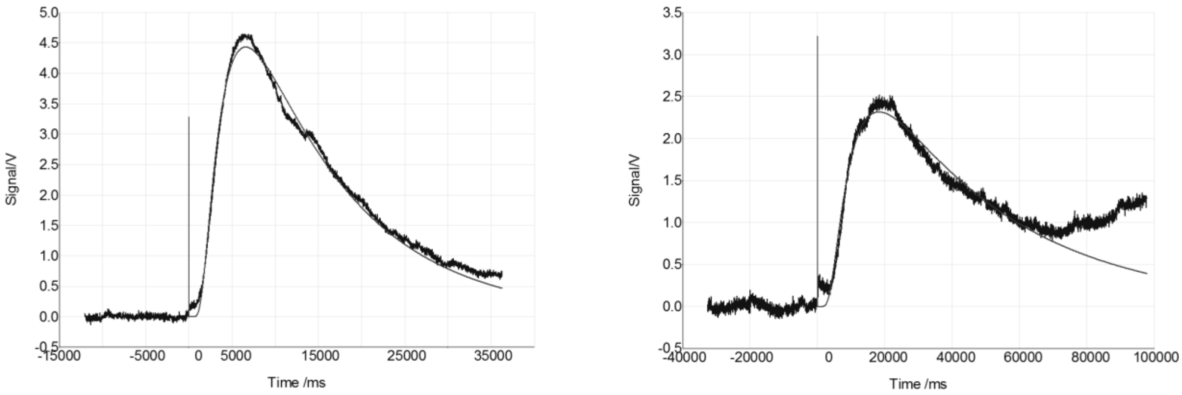

Figure 1. Influence of thickness of a ceramic sample, taken from a real element, on character of curve, which describes temperature history.

Thickness of a sample is directly related to maintenance in parallelism of bottom and top measurement surface. Non-parallel surfaces of strong roughness will make real thickness be different from a measurement value. It requires very precise and „delicate" preparation of sample surfaces, particularly in a case of porous ceramic materials, which are provided for moulds. Origin of mechanical cracks and ply separation, during mechanical treatment, fundamentally do not influence on obtained results in measurements of diffusivity and absence of precise in these results.

The next factor, related to dimensions of a sample, is a way to fix it. In a case of too big dimensions, relating to dimensions of a carrier and "rigid" fixation of a sample, during investigations in high temperature, then an effect, which is observed in figure 2, can occur. Vehement drop in thermal diffusivity is visible, because it is related to"jumping" of a sample in a carrier, what is a consequence of increase in dimensions. In an effect, instantaneous apparent drop in thermal diffusivity occurs, and it influence on non-precise matching of a model.

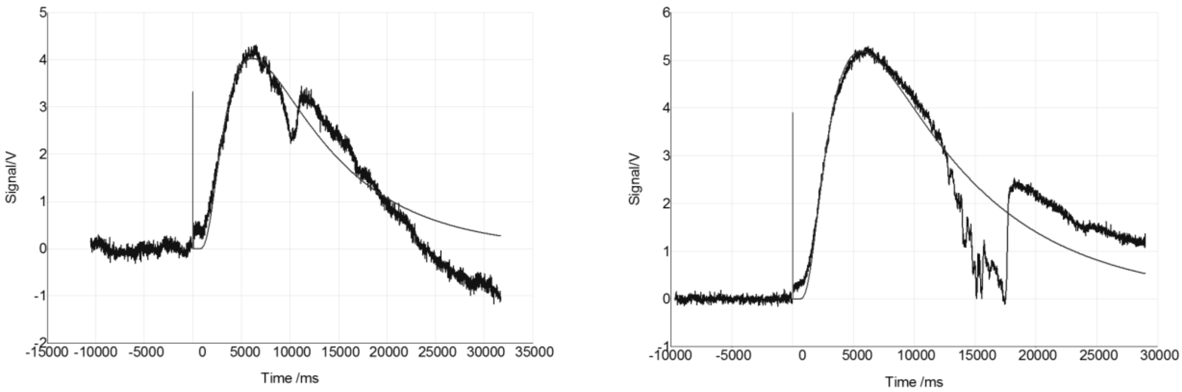

Figure 2. Temperature history of samples, which are too rigid fixed in a carrier.

A necessity to select actual acquisition time is the first problem, and selection of enough long time between individual shots in a given measurement temperature, is the second problem. Three laser shots are applied, as a standard, in each measurement temperature, what enables to draw up a mean value. Time - break between them did not exceed 1 minute. However, the performed investigations revealed, that for ceramic materials of good insulating properties, it is recommended to apply time – break from 5 to 10 minutes between individual shots, what enables to get repeatable results. An effect of too short time - break between individual shots, is presented in figure 3.These results show, that at too short time – break makes scatter of single measurements increase, particularly in high temperature. Increase in measurement temperature makes the signal be weaker and non-stabilized (figure 4). A similar scatter in results (absence of repeatability in measurements) was observed in a case of incorrect selected thickness of a sample and measurement time.

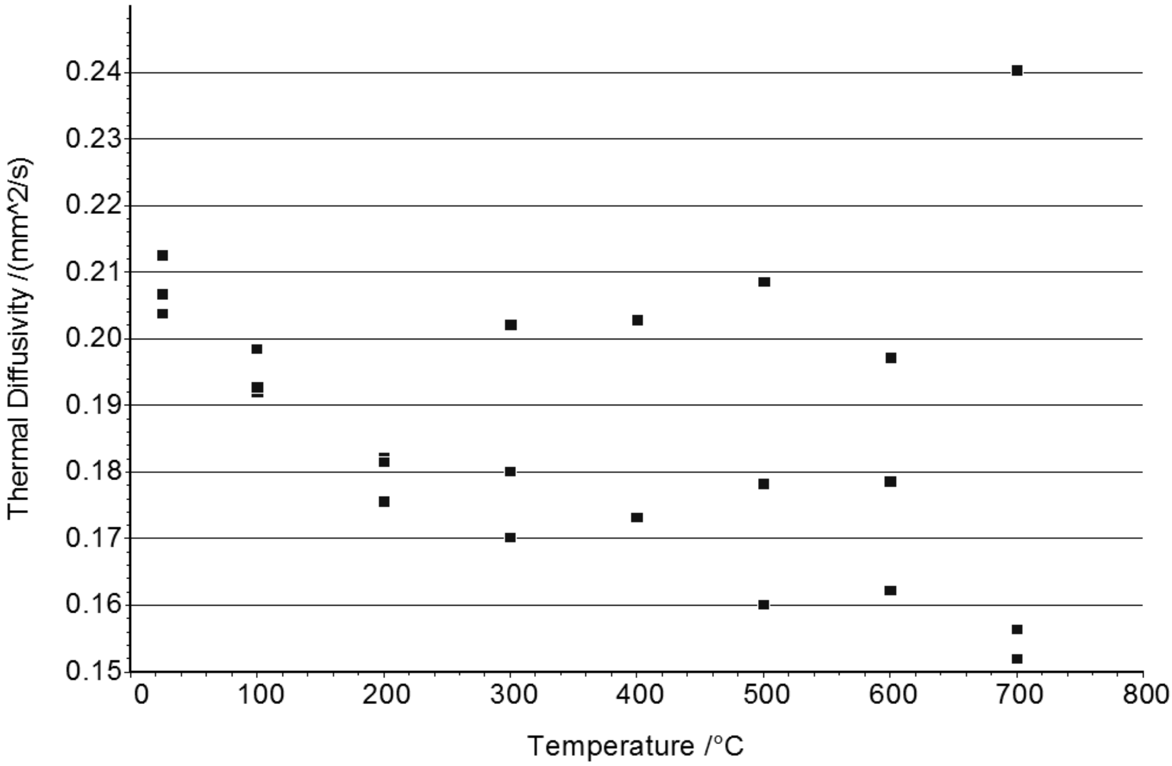

Figure 3. Influence of too short time between individual shots on repeatability in results of thermal diffusivity.

The carried out tests revealed, that improvement in quality of a graph, which describes temperature history, can be got by application of a layer to coat on both sides with colloidal graphite, what enables to improve not only transmission of a laser impulse and to reduce scattering, caused by surface roughness and emission of radiational radiation in the first milliseconds of measurement. First of all, it enables to absorb max energy of an impulse by base material and to avoid losses of the material. On the other hand, too much graphite leads to adulterate results by great overestimation of them.

The above mentioned effect of radiational emission is made by heating the front surface of a sample by a laser beam and it causes an additional impulse to originate in the graph of temperature history, while this impulse appears during the first 0-100ms in measurement. This effect has essential meaning to select a model, which describes the obtained curve. At automatic selection and criterion of the best matching, a radiational model is usually accepted. A real course of the model curve should not consider this effect, because it makes considerable overestimation in results. An example of such a curve is presented in figure 5.

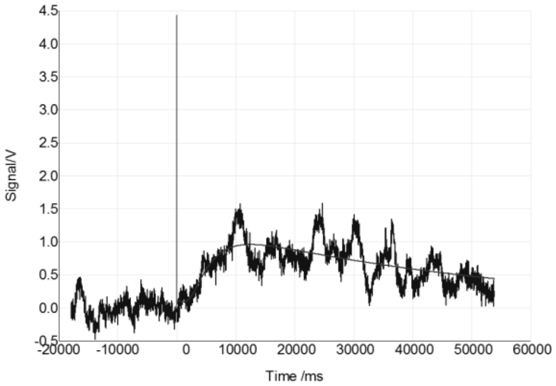

Figure 4. Attenuation effect of temperature signal in high temperature.

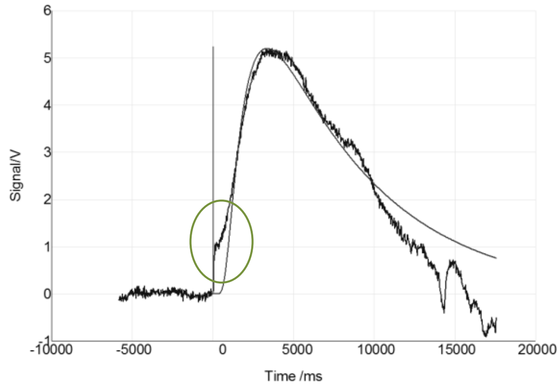

Figure 5. Temperature history with apparent thermal diffusivity in an initial period of measurement and correctly selected the Cape-Lehman model.

Considering all mentioned factors i. e. selecting individually and practically the detection time for each shot and accepting thickness 5 mm, as the most advantageous and selecting time between impulses approx. 5 – 10 minutes, and coating the samples with graphite on both sides, it was possible to get a satisfying effect, which was related to repeatability in obtained results (figure 6). However, it concerns only parameters, related to technical performance of measurement on the LFA 427 apparatus.

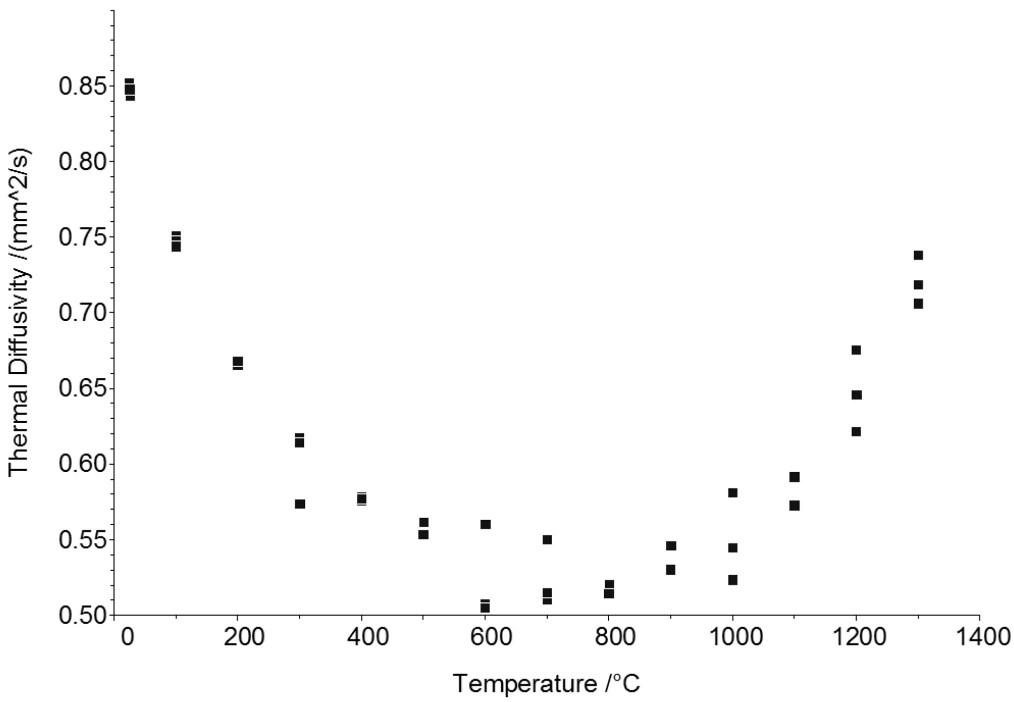

Figure 6. Results of measurements of thermal diffusivity in ceramic samples, coming from casting systems after having applied optimized parameters in the L-F test.

In the presented results, differences in preparation of samples, are not considered. Samples, cut out directly from elements of a gating system and casting moulds in a real surface state, which is practically used, were subjects of this analysis. It makes difficulties to get repeatability in results, what is a consequence of big thickness of samples and roughness of surface. To subject samples to tests, while these samples were in a form of pellets, pressed of grinded material of a mould, was one on the proposals in the investigations. However, the obtained results reveal a fundamental role of porosity in a mould, what is presented in figure 7. It is particularly essential from a point of view of data, which are indispensable to model crystallization phenomena of ready-made products.

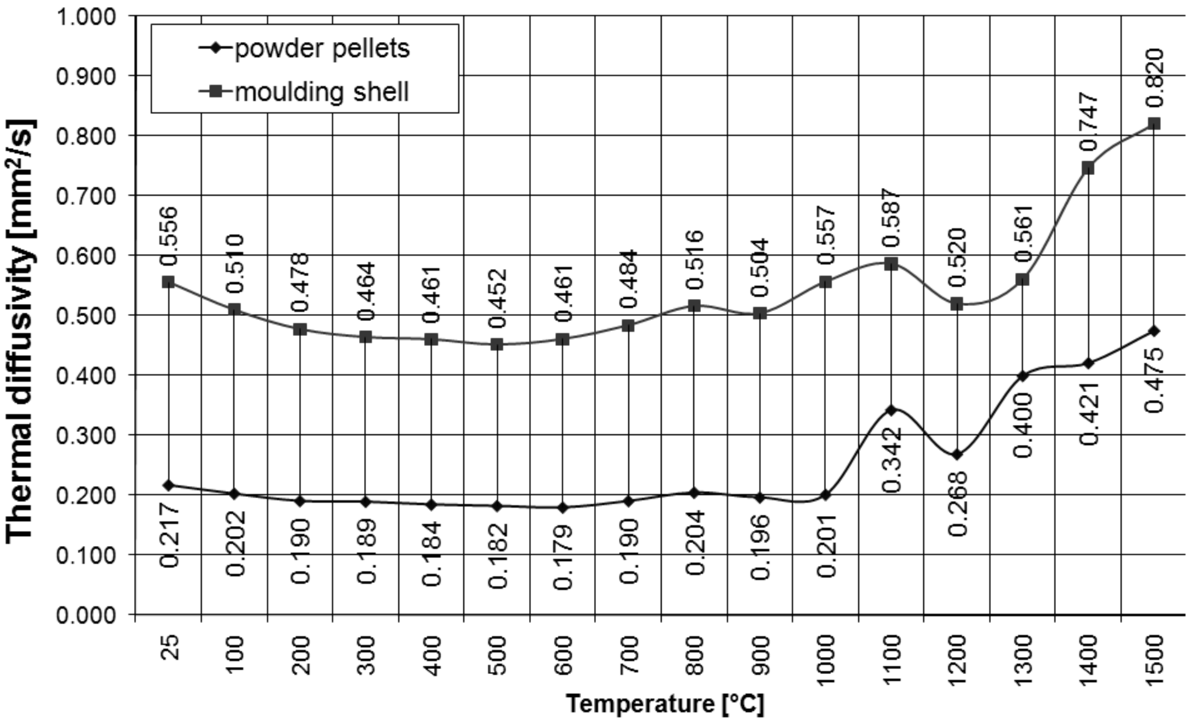

Figure 7. Influence of preparation of a sample as material taken from a real mould and in a form of pressed pellets, made of grinded powder, coming from a mould.

4. Summary

A purpose of carried out investigations was to make more precise the procedures in measurement of thermal diffusivity from a point of view to get repeatable results for ceramic materials, coming from moulds, pans and other elements of a casting system, which is used to vacuum casting process of elements in an aero-engine.

The carried out investigations revealed, that it is necessary to make thinner a sample up to dimensions approx. 5 mm, in order to shorten a measurement time. Simultaneously, acquisition time should be selected in such a way, that it makes a mathematical model, describing temperature history, be actual. Application of coating the sample with graphite on both sides is recommended as well and prolongation of time, up to 10 minutes, between individual shots in a given temperature.

Acknowledgements

Financial support of Structural Funds in the Operational Programme – Innovative Economy (IE OP) financed from the European Regional Development Fund - Project No POIG.0101.02-00-015/08 is gratefully acknowledged.

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