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Modeling of the distribution of thermal fields during spark plasma sintering of alumina ceramics

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Abstract. The article shows the use of an experimental-calculation method for the refinement of thermoelectric constants of graphite materials and alumina ceramics by experimentally studying the heating of graphite equipment and refinement thermoelectric constants of materials using the finite element method (ANSYS WORKBENCH). To simulate the thermal fields of graphite equipment based of the data on electric current, voltage and heat exchange conditions, a coupled boundary electric and non-stationary temperature problem was realized. The application of the developed approach made it possible to clarify the thermoelectric constant of materials, which helped to reduce the difference between the experimental and simulated temperatures from 20 to 5%.

1. Introduction

The method of spark plasma sintering (SPS) belongs to a class of methods based on passing low-voltage high-density direct current pulses through a heated volume [1, 2]. Heating of the powder material is provided by the release of Joule heat by passing an electric current through the graphite die with simultaneous application of mechanical pressure (see figure 1a).

The method of electro pulse plasma sintering of powder materials has several advantages compared to traditional sintering methods such as free pressing, hot pressing, isostatic hot pressing [3, 4]. SPS allows to increase the heating rate of the sintered sample tenfold from 5–25°C/min (traditional methods) to 2500°C/min (SPS). High heating rates are especially important when nano- and ultrafine-grained materials are sintered, which allow to limit the rate of grain boundary migration and to obtain high-density metal and/or ceramic materials with high strength, hardness, radiation resistance [5], etc.

One of the features of the SPS method is the control of temperature by a pyrometer focused on an external surface of the graphite die, which makes impossible to estimate the true temperature of the sample during sintering. The presence of radial temperature fields arising under conditions of high-speed heating leads to significant differences in temperature values between the surface of the graphite die and the center of the heated sample [8, 9]. For this reason, a number of researchers mistakenly correlate the controlled sintering temperature with the truth one. To estimate the true difference between the sample temperature and the controlled one, it is necessary to solve the temperature and electrical problems using experimental current and voltage curves for heating the graphite die and the sample. Modeling by the finite element method (FEM) of the SPS process is one of the most effective methods for calculating temperature fields in the graphite die and the sintered sample [1, 6, 7].



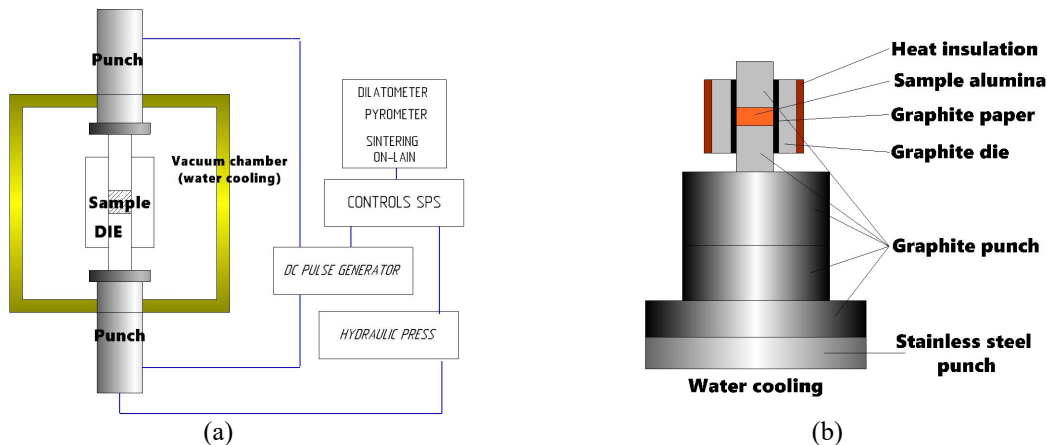


Figure 1. (a) Schematic of the SPS setup and (b) chamber design.

The results of numerical modeling of temperature fields in [8, 9] show that the temperature difference in the sample area is 1–8% of the maximum temperature. Depending on the thermoelectric properties of the materials being modeled, this difference can either increase or decrease. Often, thermoelectric properties are set the same for all grades of graphite, graphite paper and aluminum oxide, which can lead to the specified errors. Thus, the task of studying the thermoelectric properties of materials is of great interest because it helps to get modeling temperature fields inside the sample and temperature distribution in the graphite die under the heating.

This study focuses on the experimental-calculation method for the refinement of thermoelectric properties of materials (the graphite die and the sample of alumina ceramic) while heating in the SPS.

This work presents an experimentally calculated method for studying the thermoelectric properties of materials heated in SPS. The refinement of the thermoelectric properties of materials is performed by sequential experimental research and modeling of the temperature heating of the graphite die with the sample of aluminum oxide. The choice of aluminum oxide as an object of study is due to the widespread use of this ceramics in the industry, well-known constants of this material, which greatly simplifies the procedure of modeling and verification of the calculation results. Moreover, there is a large number of production data of alumina ceramics using the SPS method and hot pressing [11, 12].

2. Experimental technique

The objects of investigation of thermoelectric properties were graphite MPG-7, graphite paper “Graphlex” and sintered aluminum oxide samples α -Al₂O₃ from powder produced by Alfa Aesar. The properties of graphite materials for modeling are given in table 1. Cylindrical alumina samples had a diameter of 12, 20 and 30 mm and a thickness of 6, 10, and 8 mm, respectively. The density of the samples was 3.99 g/cm³, the average grain size of ceramics was 1 μ m. Samples were obtained using the SPS method of heating α -Al₂O₃ powders ($V_h = 100^\circ\text{C}/\text{min}$, $T_{\text{sint}} = 1150^\circ\text{C}$, $t_{\text{sint}} = 2$ min, $P = 70$ MPa). The use of samples previously SPS sintered at higher temperatures allowed us to avoid the uncertainties associated with the effect of porosity on the material properties used in modeling.

The samples are heated in a graphite die according to the standard heating mode: ($V_h = 100^\circ\text{C}/\text{min}$, $T_{\text{sint}} = 800^\circ\text{C}$, $t_{\text{sint}} = 0$ min, $P = 70$ MPa). The samples of aluminum oxide were sintered using a DR unit SINTER model SPS-625 Spark Plasma Sintering System (SPS SYNTEX INC. Ltd., Japan). The schematic diagram of the SPS installation is presented in figure 1a.

The calculation method consists of an experimental study of the heating of the graphite die at points of temperature control and the specification of thermoelectric constants of materials using the finite element method.

Thermoelectric constants were refined sequentially for all materials. At the first stage, the properties of graphite on a solid graphite billet were refined exactly in line with the geometric dimensions of the die with a diameter of 12 mm. At the second stage, the properties of graphite paper

and sintered aluminum oxide in the die with a sample of 12 mm were refined. At the third stage, the independent experiment verifies the properties of graphite paper and the aluminum oxide sample with a diameter of 20 and 30 mm. Temperature measurement of the graphite die was carried out using a chromel-copel (Cr/Ni–Cu/Ni) thermocouple (TC), connected to the device "METAKON-513". Thermocouple measurement error is $\pm 1^\circ\text{C}$. The thermocouple was placed in a blind hole 5 mm deep in graphite die. Temperature measurement was carried out with an interval of 15–60 s. The location of the points at which the temperature was measured is shown in figure 2.

Table 1. Tabular properties of materials (with T in Kelvin).

Materials	Inconel	Graphite MPG-7	Paper "Graphlex"	$\alpha\text{-Al}_2\text{O}_3$
Properties				
Thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$	$10.09 + 1.57 \cdot 10^{-2} \cdot T$ [8, 9]	$82.85 - 0.06 \cdot T + 2.58 \cdot 10^{-5} \cdot T^2$ [8, 9]	3-5 [7]	$39.5 \cdot T^{-1.26}$ [8, 9]
Electrical resistivity, $\Omega \text{ m}$	$9.82 \cdot 10^{-7} + 1.6 \cdot 10^{-10} \cdot T$ [8, 9]	$2.14 \cdot 10^{-5} + 1.34 \cdot 10^{-8} \cdot T + 4.42 \cdot 10^{-12} \cdot T^2$ [8, 9]	1-1.25 [7]	$8.7 \cdot 10^{19} \cdot T^{-4.82}$ [8, 9]
Heat capacity, $\text{J kg}^{-1} \text{K}^{-1}$	$344 + 0.25 \cdot T$ [8, 9]	$34.27 + 2.72 \cdot T - 9.6 \cdot 10^{-5} \cdot T^2$ [8, 9]	-	850 [8, 9]

For the numerical implementation of the thermoelectric problem in a non-stationary formulation, a finite element package ANSYS 18.2 was used. The constants of tables 1 and 2 were chosen as the initial thermoelectric properties of materials. The boundary conditions of this thermoelectric problem are presented in figure 3. On the line B (figure 3a), which bounds the simulated system from above, the current was set as a linear function of time. Along the lower line A (figure 3a), which bounds the simulated system from the bottom, the voltage value was set. The entire volume of the two-dimensional body A (figure 3b) was solved as a part of the electrical problem (the temperatures obtained from the calculation of the electrical problem were solved in the unsteady heat conduction problem). Across the outer contour of the simulated system B (figure 3b), besides the line C, which bounds a part of the graphite die, with the help of the value of specific thermal conductivity the convection conditions were introduced. Along line C (figure 3b), which bounds a part of a graphite die, thermal insulation conditions were imposed, prohibiting heat removal. In processing of modeling, elements of graphite die were considered as isotropic, which made it possible to set an assumption of the uniformity of all physical properties in all directions. The physical properties of the materials used in the model and the alumina constants are listed in table 1.

Figure 2 presents the results of temperature measurement at control points for a single graphite (Figures 2a, 2b) and for die with the alumina sample with diameters of 12, 20, and 30 mm (see figures 2c–2e, respectively).

3. Experimental results and discussion

The results of the research represent the constructed numerical model for a single graphite die. Based on the model, we chose graphite paper constants using the method of passive search in space to minimize the discrepancy of the function of temperature values at the control points in the experiment and numerical calculation. The next part of the calculation was the determination of the constants of the ceramic material - sintered aluminum oxide (sample diameter of 12 mm). Further, the constants obtained earlier were verified on a complete system with ceramic samples of aluminum oxide with diameters of 20 and 30 mm. The refined thermoelectric constants of the materials are given in table 2.

At the control points at the finite period of time corresponding to the experimental end of the sintering process a comparison of experimental data with data obtained as a result of numerical simulation was made. It was established the magnitude of the discrepancy does not exceed 5%, which corresponds to a good numerical result obtained

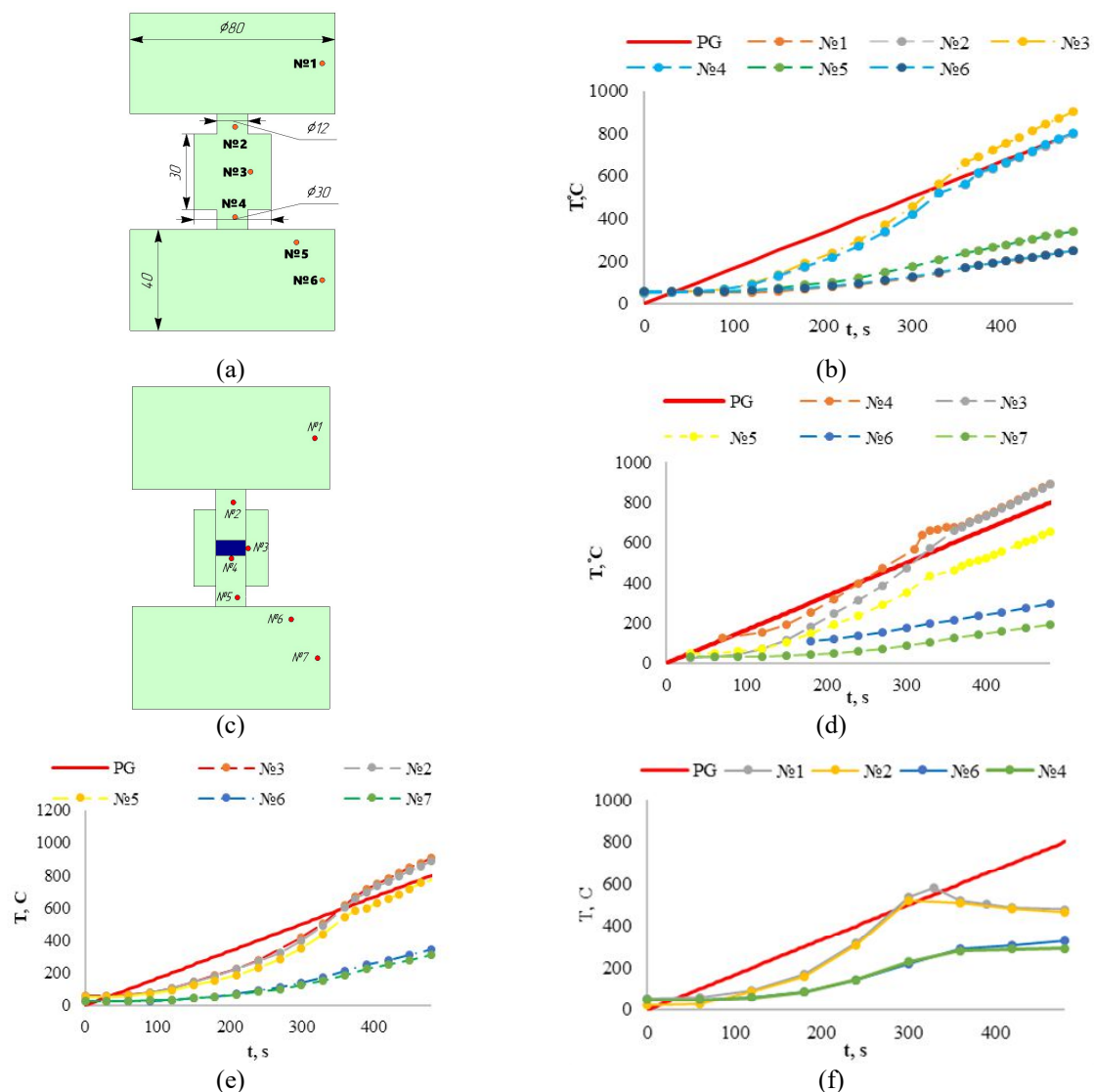


Figure 2. Experimental data: (a) temperature control points for single graphite die, (b) heat curves of single graphite die, (c) temperature control points, (d) heat curves of graphite die and sample 12 mm, (e) heat curves of graphite die and sample 20 mm, (f) heat curves of graphite die and sample 30 mm.

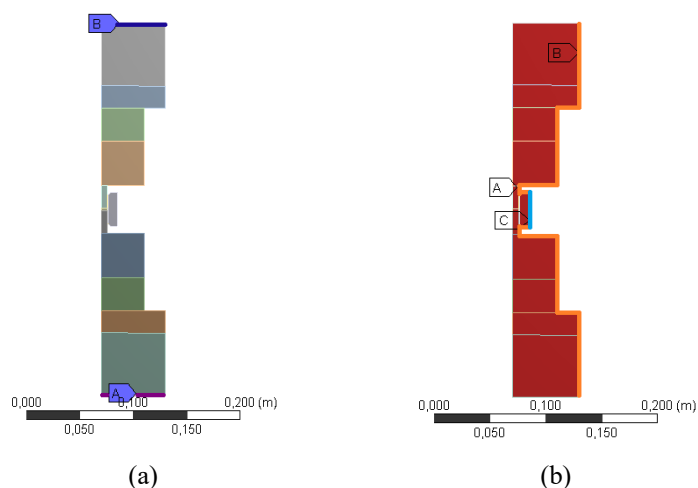


Figure 3. Boundary conditions of model FEM: (a) electrical, (b) temperature.

Table 2. Specify properties of materials (with T in Kelvin).

Properties \ Materials	Inconel	Graphite MPG-7	Paper “Graphlex”	α -Al ₂ O ₃
Thermal conductivity, W m ⁻¹ K ⁻¹	11.4+1.4·10 ⁻² ·T	103.6-0.08·T+3.0·10 ⁻⁵ ·T ²	103	27.6-2.0·10 ⁻² ·T
Electrical resistivity, Ω m	1.18·10 ⁻⁷	2.1·10 ⁻⁵	3.5·10 ⁻⁷	1.0·10 ⁸ -1.5·10 ⁻⁵ ·T ^{-4.82}
Heat capacity, J kg ⁻¹ K ⁻¹	446.5+ 0.16·T	4.2·10 ² +1.4·T	709	931+0.34·T

4. Conclusions

The developed experimental-calculation method for the study of thermoelectric constants of materials and the simulation of temperature fields for graphite die made it possible to specify the thermoelectric constants of materials, which helped to reduce the difference between experimental and simulated temperatures from 20 to 5%. The obtained results open up new opportunities for optimizing the SPS regimes to increase the heating uniformity of the sintered alumina samples.

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