Calculation of thermal fields in a graphite mold during spark-plasma sintering of non-conductive materials

N A Rubinkovskiy, A G Zholnin and E G Grigoryev
National Research Nuclear University MEPhI, Kashirskoe shosse, 31, Moscow, 115409, Russia
E-mail: fissium@yandex.ru

Abstract. The research provides modeling of spark-plasma sintering process of aluminum oxide, specifically the distribution of temperature fields over the sample volume and mold at various stages of heating. Calculation was based on the experimental data on the measurement of temperature on the surface of the mold, inside the mold cavity and in its various internal parts including punches (diameter of 15 mm), in the absence of insulation felt. Experiments established that the main source of heat emission at temperatures less than 1300°C to 1400 °C is contact resistance at the borders of the punches and the matrix molds. Then heat emission zone moves towards the punches. Based on the received data, the temperature and electric parameters were selected for the materials used for the press mold, providing a good agreement between the observed and calculated distribution patterns at different temperatures. Obtained parameters were used in calculations of temperature fields in the press molds with insulation felt and for the press molds with diameter of 15 mm and 10 mm for the production of tablets as well as glazing beads.

1. Introduction
Spark-plasma sintering (SPS) is one of the promising methods for the consolidation of ceramic materials. SPS is based on the combined effect of pulsed direct current and mechanical pressure on the particulate material. The material in the target area is heated to very high temperatures. SPS of the powder material is conducted in a current-conductive graphite press mold, therefore the technology has no restrictions on the type of sintered powder materials. During sintering of conductive powder the heating of the powders and compacts made of them is carried out by passing a direct current through them, and during sintering non-conductive materials - by heating the graphite press mold elements.

For all advantages of the SPS method there is a serious flaw: in the units when the sample temperature is monitored by measuring the surface temperature of the mold or in an opening made in the mold, it is not possible to reliably establish the temperature of the sample itself. This may lead to an underheating, overheating or uneven heating of the sintered material, which in turn will affect the final properties of the compact.

Foreign companies are actively working on the modeling of temperature fields in the mold during spark-plasma sintering, however, most of these research use metallic systems as a sintered material, and are limited to using ceramic materials at low temperatures only [1-6]. No Russian research was found on this issue.

The main objective of this research was the modeling of spark-plasma sintering of aluminum oxide, in particular the distribution of temperature fields over the volumes of the sample and the mold at
different stages of heating, in order to take into account the possible errors in the production of final products, i.e. temperature measurement for various materials with different geometry of compacts.

2. Equipment and methods of research
Spark-plasma sintering (SPS) was conducted in a LABOX machine model 625 (Sinter Land, Japan). The sintering was performed on the standard procedure in the graphite matrix with an additional layer of graphite paper between the matrix and the powder filling, and between the graphite punches and the powder to prevent sintering of the powder to the equipment material. Temperature control was performed with an optical pyrometer with a measuring range 573°C to 3000 °C. directed to the matrix through a hole cut in the graphite felt, which was used as thermal insulation to prevent heat loss by radiation. A previously sintered tablet of 15 mm diameter was used in the measurements. For observation of the temperature distribution inside the mold a quarter of the matrix was removed. For temperature measurements at different surfaces of the system the heating was carried out without using thermal insulation felt. Pictures of the mold were made with a digital camera. Modeling was carried out with a COMSOL Multiphysics software package.

3. The reference experiments
Figure 1 shows the scheme of structures used in the experimental temperature measurements.

Figure 1. Scheme of structures for temperature measurement: (a) an opening diameter of 5 mm and depth of 8 mm, (b) a quarter of matrix is removed.
Figure 2 shows a photograph of the structure (Figure 1 (a)) at surface temperature equal 1000 °C and graph of dependence of the difference between the temperature of the opening and the surface temperature on the surface temperature of the press mold.

![Figure 2](image_url)

**Figure 2.** Photograph of the press mold (a); graph of dependence of the difference between the temperature of the opening and the temperature of the surface on the temperature of the surface (b).

The figure 2 shows that as the temperature of the surface increases the temperature difference between the opening and the surface, as expected.

The next step was to determine the difference of temperatures between the surface and the sample. To do that, the experiments were conducted on the structure in Figure 1 (b). Figure 3 shows photographs of the press mold at high and low temperature, and Figure 4 presents a graph of the dependence of the difference between the sample temperature and the surface temperature on the surface temperature.

![Figure 3](image_url)

**Figure 3.** Photographs of the press mold at high and low temperature: (a) surface temperature amounts 1100 °C, (b) surface temperature amounts to 1500 °C.
It was found that at the temperatures less than about 1000 °C the sample is colder than the surface. In addition, there is a specific feature: the primary source of heat emission at temperatures about 1300°C - 1400 °C is the contact resistance at contact resistance at the punch-matrix borders. Then heat emission zone moves towards the punches. This can be explained by the fact that during the heating process the thermal expansion of punches occurs, and as a result the value of the contact resistance reduces.

4. Modeling
In addition to the parameters of materials used in the research, the modeling required determining the parameters of the thermal and electrical contact resistance at the punch-matrix border, as showed in Figure 5.

![Figure 4](image.png)

**Figure 4.** Graph of the dependence of the difference between the sample temperature and the surface temperature on the surface temperature.

![Figure 5](image.png)

**Figure 5.** Contact resistance area.
The parameters of graphite, which the punches, press mold and spacers are composed of, as well as the parameters of aluminum oxide are taken from the research [1].

The reference experiments allowed choosing the temperature correspondence between the thermal and electrical resistance of the contact area of punch-matrix. The results of the simulation are presented in Figure 6.

![Figure 6. Results of modeling: (a) 1100 °C, (b) 1500 °C.](image)

It is evident that the calculated data provide a good agreement with the experiment and reflect the changing of heat zone mechanism.

Since the actual mold is surrounded by a thermal insulation felt, it was necessary to adapt the results of modeling to this case, as shown in Figure 7.

![Figure 7. Temperature distribution in the diameter of the mold (the line passes through the sample) with thermal insulation.](image)
The results show that with a good thermal insulation and with temperature measurement in an opening of sufficient depth (8 mm) the sample temperature can be controlled with a high degree of accuracy.

The model has also been generalized to the case of obtaining glazing beads, i.e. compacts with the height of 10 mm to 15 mm, the results shown in Figure 8.

![Figure 8. The temperature distribution in the diameter of the press mold for the case of producing glazing beads.](image)

It appears that with a good thermal insulation it is possible to achieve a nearly uniform temperature distribution in the height of the sample. The difference of temperatures does not exceed 50°C to 70 °C. In this case, the Japanese machine model has an advantage over the German machine model, where the temperature is measured through an opening in the punch. Such measurement of temperature cause changes in the temperature field at the edge of the sample, which that does not allow achieving a uniform temperature distribution and therefore the uniformity of properties of the compact after sintering.

5. Discussion of results
The main feature of the experimental part of the research was a discovery that the main source of heat emission at temperatures less than 1300°C to 1400 °C is contact resistance at the punch-matrix borders. Then heat emission zone moves towards the punches. This can be explained by the fact that thermal expansion of the punches occurs during the heating process, and as a result, the value of the contact resistance reduces. Moreover, the resistance of the contact itself reduces with decrease of temperature.

Up to a certain point (1000 °C) the temperature of the sample is lower than the surface temperature. It is possible to assume that this relates to more intensive heat removal due to the emission and thermal conductivity (towards cooled electrodes) compared to its absorption in the sample. The calculation model showed a good agreement with the experiment, and its generalization to the real systems with thermal insulation will allow controlling the temperature of the sample by controlling the temperature of the surface or of the opening.

6. Conclusions
The experimental results allowed establishing the mechanism of change of the heat emission source, which moves from the contacts to the punches. Literature sources lack this information.

The temperature dependencies of electric and contact resistance on the punch-matrix border were selected. The calculation model based on these dependencies shows a good match to the experiment model.

The model was extended to the systems with thermal insulation and demonstrated the possibility of the sample temperature control by the means of controlling the temperature of the surface or the opening. In addition, the research allowed substantiating the possibility of producing glazing beads with a uniform temperature distribution in the sample height, and consequently the uniform distribution of properties. The results may be generalized to any ceramic materials and geometric features of the assembly structure.

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