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Numerical determination of thermoelectric para meters for two-stage sintering of cBN–Al system samples in a high-pressure cubic cell

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Preliminary numerical modeling of coupled electric and thermal fields in a six-punch HPA during reaction sintering of massive samples of PCBN has been performed. It was assumed that the sintering of the superhard composite is carried out from the cBN–Al charge. The FEM code was used to solve the steady problem of electro- and thermal conductivity. The conductive properties of the cell materials were adjusted to meet the conditions of its thermobaric loading. The obtained results allow us to determine the level of thermoelectric parameters of two-stage reaction sintering and evaluate the effect of changes in the conducting properties of the cell on the thermal state of PCBN samples.

Keywords: polycrystalline cubic boron nitride (PCBN), high-pressure apparatus (HPA), thermal state, modeling.

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Introduction

Polycrystalline cubic boron nitride (PCBN) is widely used for the manufacture of tools intended for blade processing of hardened and alloyed steels, cast irons and other hard-to-machine iron-based materials [1]. Today, a topical issue is the synthesis of large size samples of PCBN, which can be used as a structural material, in particular, for the manufacture of highly loaded compression elements in high-pressure apparatus (HPA) [2].

One of the methods for obtaining PCBN is based on the reactive two-stage sintering of samples of the cBN–Al system under conditions of high pressure and temperature [3]. Various types of HPA are used to generate the high pressure and temperature required for sintering. In the Bakul Institute for Superhard Materials of the National Academy of Sciences of Ukraine PCBN composites are sintered in "recessed anvil" and "toroid" types of HPA, which provide a pressure value of 5–7 GPa and a temperature of 1500–2000 °C. However, it is impossible to obtain sufficiently large samples in such apparatuses due to the limited sizes of the cell. A six-punch HPA can be used to solve the actual problem of obtaining large size samples of superhard PCBN composites. The cubic cell of such apparatus with a rib size of up to 100 mm allows to provide the necessary *p*, *T*-parameters for sintering (4–5 GPa, 1600–1800 °C).

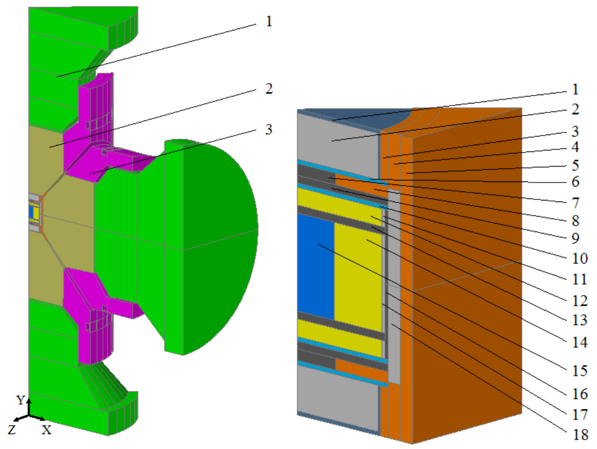
It is known [4, 5], that the temperature distribution in the reaction volume of HPA has a decisive influence on the structure and properties of PCBN samples during their reaction sintering. Computer simulation is used in advance to design HPA cells and ensure the necessary thermal conditions in them. This makes it possible to determine the thermoelectric parameters of the external action on the HPA and, accordingly, to optimize the voltage fields, current density, Joule heat sources, and the temperature in the cell elements. By making changes to the configuration and conductive properties of the elements of the electroresistive circuit of the cell, it is possible to achieve the required thermal conditions in its reaction volume. Thus, preliminary modeling allows to reduce the time of design works and obtain a more effective and high-quality project of the reaction cell. The thermal state for various types of HPA is modeled in detail, in particular, in works

[6–12].

The purpose of the work is preliminary determination using computer modeling of the thermal state of the cell and the corresponding thermoelectric parameters of action on the six-punch HPA at various stages of reaction sintering of the composite of 90 wt.% cBN–10 wt.% Al system. The academic version of the ANSYS software was used.

I. Formulation of the problem

The calculation scheme of the HPA and the cell for sintering PCBN composites (1/8 part in a compressed state) are shown in Fig. 1. The reaction volume 15 is surrounded by disks 13 and cylinder 14 (see Fig. 1, b).



a b

Fig. 1. a – general scheme of the six-punch HPA: 1 – support plates (steel), 2 – punch (hard alloy), 3 – water cooling system (steel); b – scheme of a high-pressure cell: 1 – cup (stainless steel), 2 – disc (dolomite), 3, 4, 8 – rings (pyrophyllite), 5 – cubic container (pyrophyllite), 6, 10 – discs (titanium), 7, 9 – disks (graphite), 11 – ring (dolomite), 12, 14 – disk and cylinder (aluminum oxide), 13 – disk (graphinite), 15 – sample 90 wt.% cBN–10 wt.% Al, 16 – heater (graphite), 17, 18 – bushings (dolomite).

Electroresistive heating of the cell occurs due to the release of Joule heat in its graphite elements. In this case the thermal state is modeled by solving the coupled problem of electrical and thermal conductivity. Assuming the absence of electric charges, such process is described by a system of nonlinear equations

$$\mathrm{div}[\gamma(T)\mathrm{grad}U] = 0, \qquad (1)$$

$$\mathrm{div}[\lambda(T)\mathrm{grad}T] + \gamma(T)|\mathrm{grad}U|^2 = 0, \qquad (2)$$

where γ – electrical conductivity coefficient; T – temperature; U – electric field potential; λ – thermal conductivity coefficient; $\gamma|\mathrm{grad}U|^2$ – specific power of Joule heat sources. Equations (1) and (2) are supplemented by boundary conditions

$$U_{S_U} = U(\mathbf{r}), \qquad (3)$$

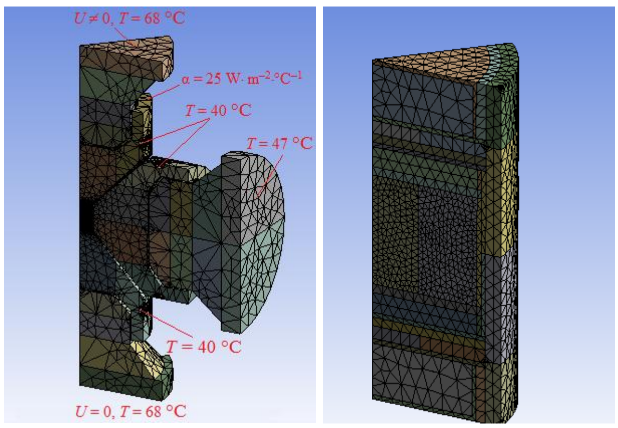
$$T_{S_T} = T(\mathbf{r}), \qquad (4)$$

$$h_{S_\alpha} = \alpha(\mathbf{r})[T - \Theta(\mathbf{r})] = -\mathrm{n}\cdot\lambda(T)\mathrm{grad}T, \qquad (5)$$

where S_U, S_T, S_α – respectively, the boundary surfaces of the HPA with radius vector **r**, on which there are set the values of voltage $U(\mathbf{r})$, temperature $T(\mathbf{r})$ and the condition of convective heat exchange with the heat transfer coefficient $\alpha(\mathbf{r})$ and the temperature of the external environment $\Theta(\mathbf{r})$; h – is the projection of the heat flow vector onto the external normal **n** to S_α.

The electrical and thermophysical properties of the materials of the components of the HPA were taken from publications [12–15], taking into account the dependence of the conductive properties on temperature.

The meshes of elements for the six-punch HPA and its cell with the corresponding boundary conditions are showed on Fig. 2. The value of the electric potential (3) on the end surfaces of the upper and lower support plates was varied based on the need to obtain the appropriate temperature in the reaction volume. The temperature (4) on the end surfaces of the support plates was determined by measurement. To simulate the forced cooling of the HPA, a temperature (4) of 40 °C was set on the inner surfaces of the cooling channels. On the surfaces in contact with air, the condition of convective heat exchange (5) was set (α = 25 W$\cdot$ m$^{-2}\cdot$°C^{-1} [9], Θ = 40 °C).



a b

Fig. 2. Finite-element meshes of 1/8 parts of HPA (a) and high-pressure cell (b).

Calculations were performed for the case of the transition of the sample under consideration from the insulating to the conductive state and vice versa, i.e. the melting of aluminum with infiltration of the charge in the process of reactive sintering and the reactive interaction of aluminum with cubic boron nitride with the formation of non-conductive phases (aluminum nitride and borides and solid solutions) were taken into account. First, the HPA heating current power was determined under the conditions when the cBN–Al sample is an insulator at a maximum temperature of ~900 °C and a pressure of 2 GPa (task 1). The next stage – the cBN–Al sample acquired conductive properties after aluminum melting, the heating power coincides with the previously determined one (task 2). These two problems relate to the first stage of reactive sintering. The second stage of sintering is carried out at a temperature of ~ 1400 °C and a pressure of 4.2 GPa, when the sample can also acquire both conductive and insulating properties. Therefore, the necessary values of the heating current power of the cell were determined and its thermal state was analyzed at the maximum sample temperature of 1400 °C in the insulating (task 3) and conductive (task 4) states.

II. Results and discussion

Task 1 – to investigate the thermal state of the composite of 90% cBN–10% Al system for the heating scheme corresponding to the insulating state of the sample ($\rho = 10^5$ Ω·m, $p = 2$ GPa, $T_{max} = 900$ °C). In Fig. 3–6 the calculated voltage, current density, Joule heat source density, and temperature fields in the cell elements were showed.

According to the proposed scheme of reaction volume heating for sintering PCBN based on a powder mixture of 90% cBN–10% Al, when the maximum temperature in the sintering zone is equal to 900 °C, the maximum temperature difference does not exceed 3 °C (see Fig. 6), which satisfactorily affects to formation of a uniform structure and properties of the sample as a whole.

Task 2 – to investigate the thermal state of the 90% cBN–10% Al composite for the heating scheme corresponding to the conductive state of the sample at $p = 2$ GPa and the heating current power $W = 6.59$ kW, calculated for the insulating state of the sample heated to $T = 900$ °C.

Fig. 7 shows the temperature fields in the sample, which are significantly different from the previous ones. In the case of aluminum melting, the sample becomes conductive, and the stabilization of the power at the level of 6.6 kW, required to heat a non-conductive sample to 900 °C under a pressure of 2 GPa, leads to an increase in the temperature drop in it from 3 to 14 °C, as well as to increase in the heating current and, accordingly, to reduction in voltage. Thus, by recording the current-voltage characteristics of the PCBN sintering process, it is possible to monitor the change in the phase state of the material in the reaction volume of the cell.

Task 3 – to investigate the thermal state of the 90% cBN–10% Al composite for the heating scheme corresponding to the insulating state of the sample ($\rho = 10^5$ Ω·m, $p = 4.2$ GPa, $T_{max} = 1400$ °C).

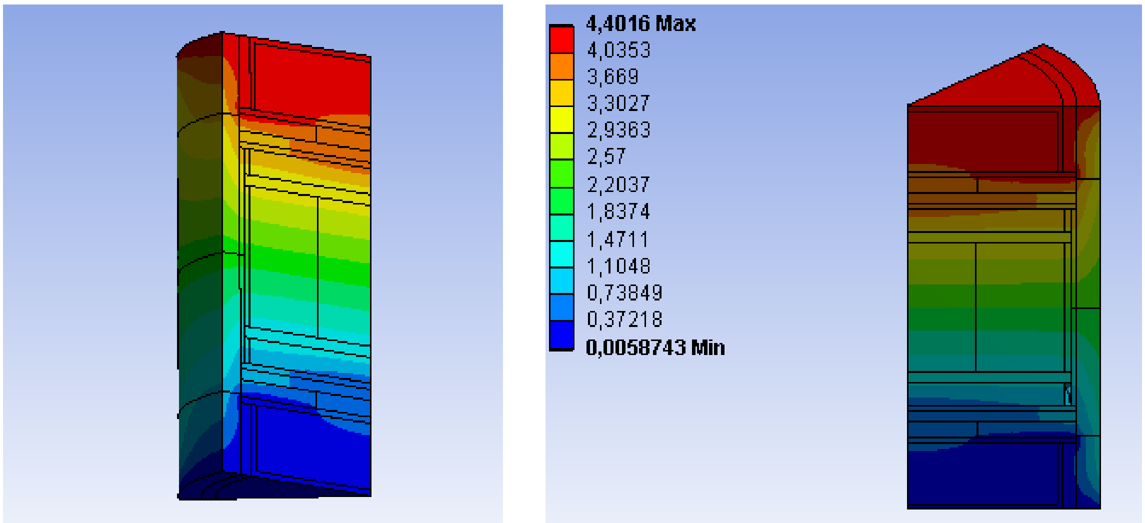


Fig. 3. Voltage fields in diagonal and frontal sections of the cell, V.

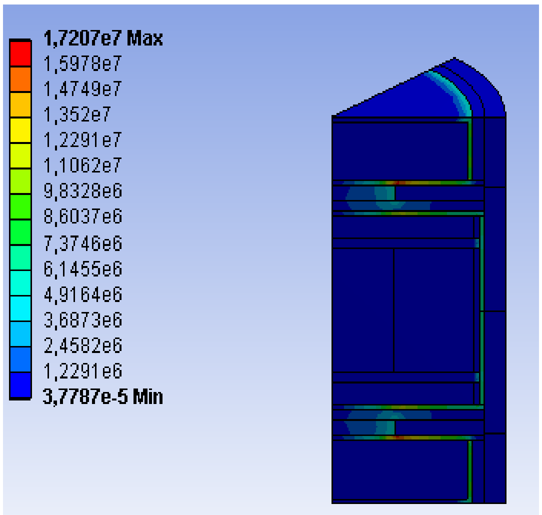


Fig. 4. Current density field, A/m^2.

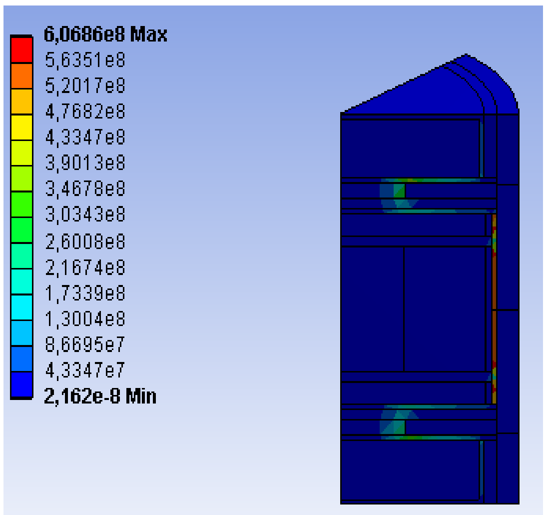


Fig. 5. Density field of heat sources, W/m^3.

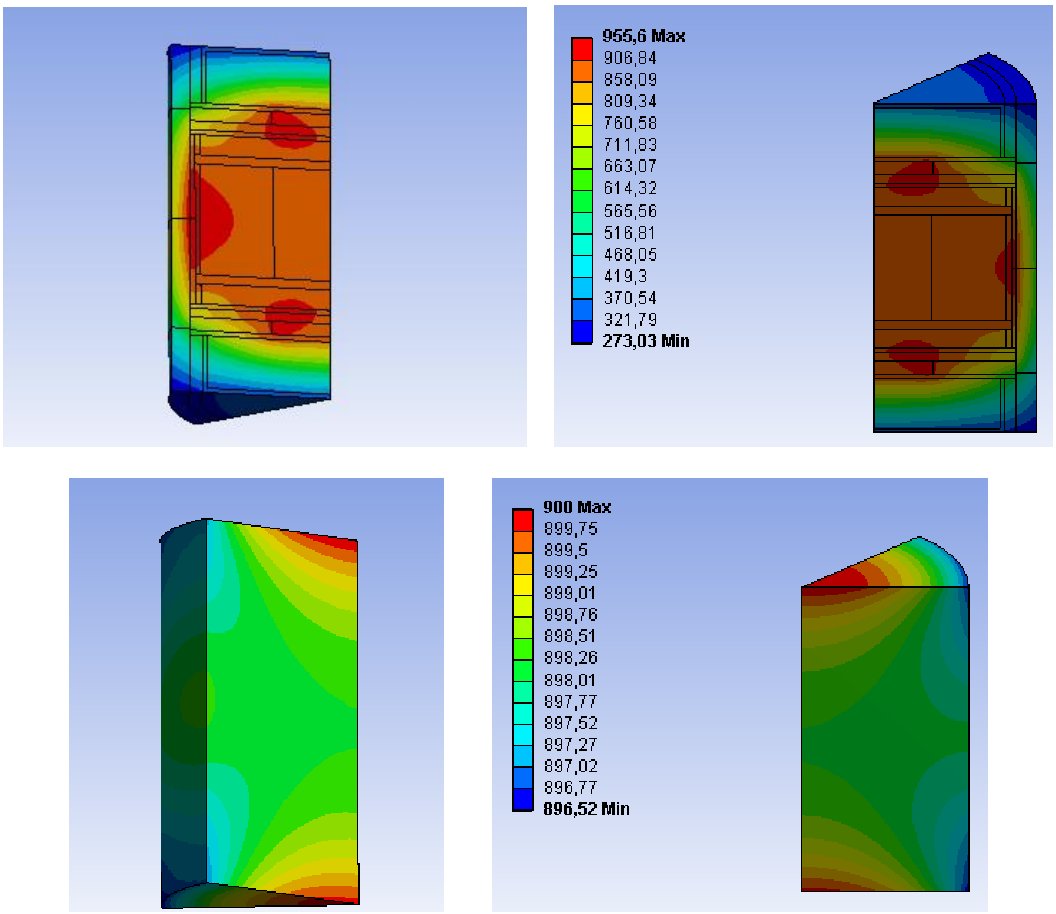


Fig. 6. Temperature fields in diagonal and frontal sections of the cell and the reaction volume, °C.

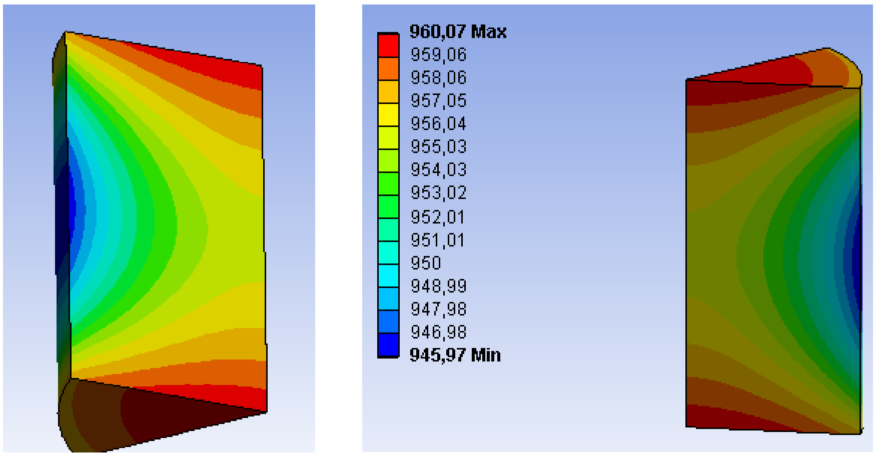


Fig. 7. Temperature fields in diagonal and frontal sections of the reaction volume, °C.

In Fig. 8 the calculated temperature fields in the PCBN sample are showed. The determined thermal state for the scheme of heating the reaction volume, when the maximum temperature in the sintering zone will be 1400 °C at a pressure of 4.2 GPa, is characterized by a maximum temperature drop of 5 °C, which confirms the effectiveness of the selected scheme of heating the HPA cell.

Task 4 – to investigate the thermal state of the 90% cBN–10% Al composite for the cell heating scheme corresponding to the conductive state of the sample at p = 4.2 GPa and the heating current power of 10.59 kW, calculated for the insulating state of the sample heated to T = 1400 °C.

Fig. 9 shows the calculated temperature fields in the sample. In the case of aluminum melting, the sample becomes conductive, and the stabilization of the power at the level of 10.59 kW, required to heat a non-conductive sample to 1400 °C under a pressure of 4.2 GPa, leads to increasing temperature drop from 5 to 23 °C in sample, to increase in the heating current and, accordingly, to a decrease in voltage. However, even with such a temperature difference, we have sufficiently uniform conditions of thermobaric action in the volume of the PCBN material.

The summarizing Table 1 gives the pressure values and calculated sintering parameters (temperature extremes, voltage, current, power) in the reaction volume

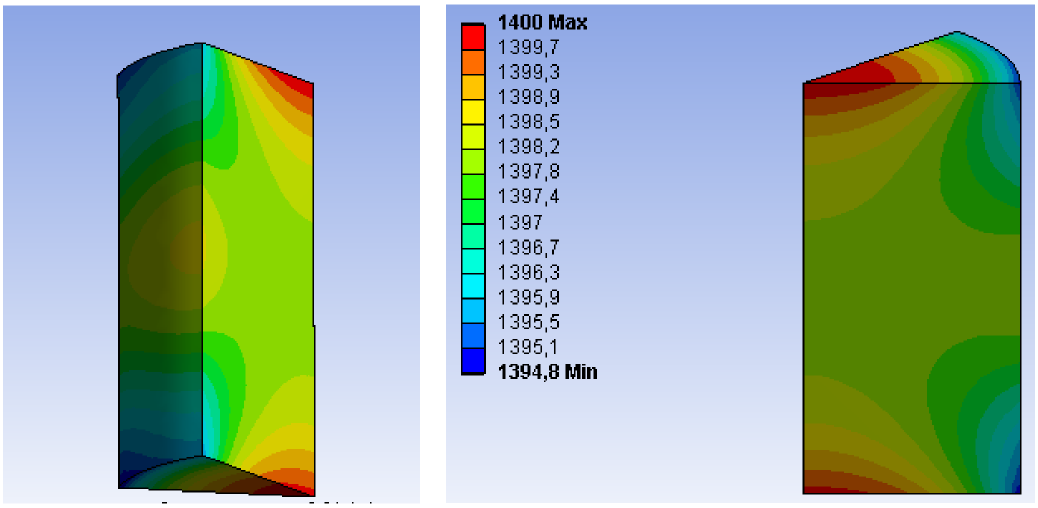


Fig. 8. Temperature fields in diagonal and frontal sections of the cell and the reaction volume, °C.

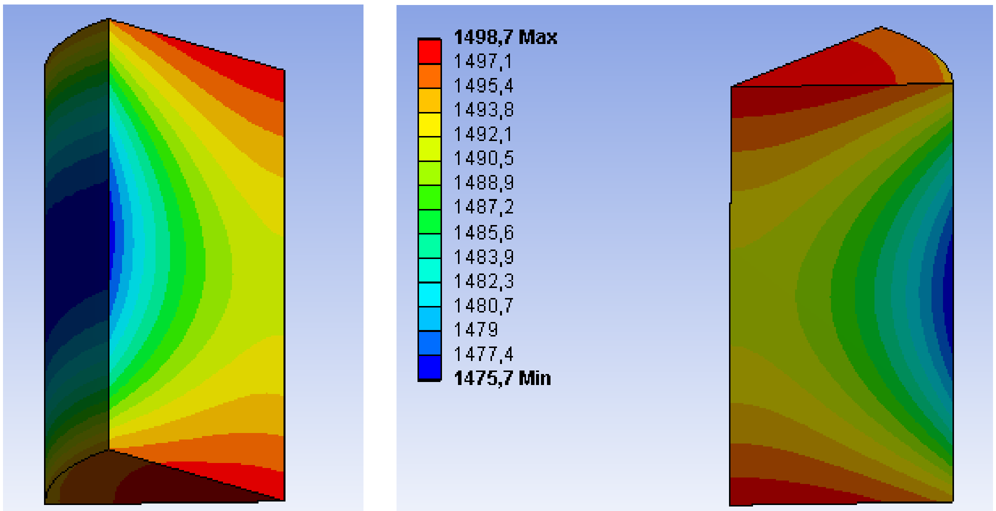


Fig. 9. Temperature fields in diagonal and frontal sections of the cell and the reaction volume, °C.

Table 1.

Parameters of the PCBN 90 % cBN–10 % Al sample sintering

Scheme №	*T*, °C	*p*, GPa	*U*, V	*I*, kA	*W*, kW
1 (sample – insulator)	T_{max} = 900 T_{min} = 897	2	4,4075	1,495	6,59
2 (sample – conductor)	T_{max} = 960 T_{min} = 946	2	4,018	1,642	6,60
3 (sample – insulator)	T_{max} = 1400 T_{min} = 1395	4,2	5,8732	1,803	10,59
4 (sample – conductor)	T_{max} = 1499 T_{min} = 1476	4,2	5,345	1,981	10,59

for four sintering schemes.

The above data were obtained for two edge values of the resistance of the cBN–Al sample (insulating and conducting). To analyze the influence of sample resistance on changes in the thermoelectric parameters of sintering, they were calculated and graphed the dependence of voltage *U*, current *I* in the HPA and temperature drop ΔT in the sample on its resistance (Fig. 10). Calculations were performed for two stages of heating: under the condition of constant heating current power W = 6.59 kW (first stage) and W = 10.59 kW (second stage). To simulate the transition of the cBN–Al sample from the insulating to the conducting state, the value of its specific resistance was set in the interval 10^{5}–10^{-5} Ω·m with a logarithmic step of -1.

As can be seen from the graphs, for a sample resistance value of ~ 10^{-3} Ω·m, there is a sharp increase in the HPA heating current and the temperature drop in the sample with a simultaneous drop in a voltage in the apparatus. The value of the calculated temperature difference in the sample of 14 °C at the first and 23°C at the second stage of sintering for its resistance of 10^{-5} Ω·m ensures a fairly uniform thermal state of the sample during its reactive sintering.

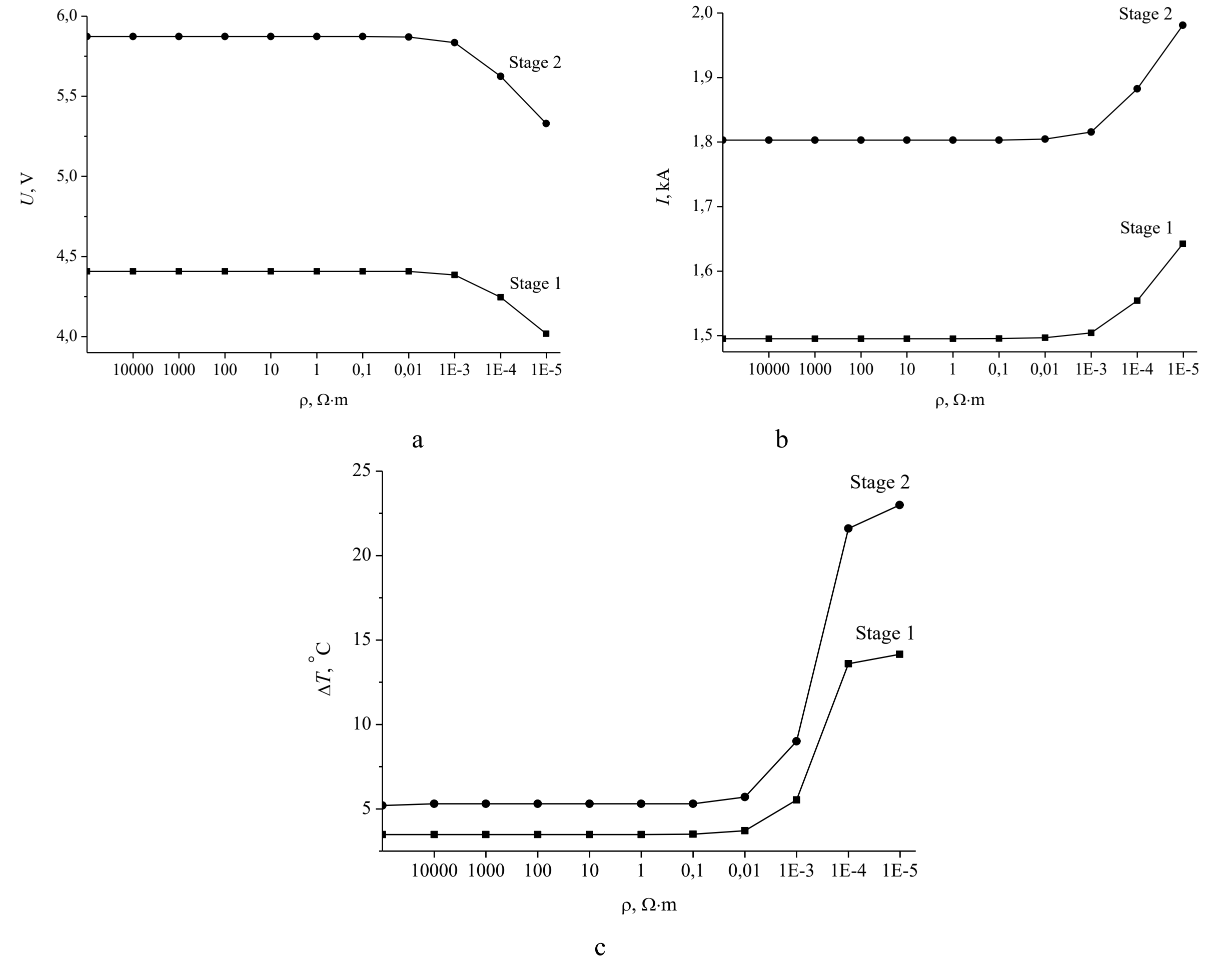


Fig. 10. Dynamics of changes in thermoelectric parameters in the HPA during the infiltration of the PCBN sample with molten Al at the first and second stages of reaction sintering, respectively, at powers of 6.59 and 10.59 kW.

Conclusions

1. The developed method of modeling electroresistive heating of a six-punch HPA in the process of two-stage reaction sintering of cBN–Al system PCBN composite allows to determine the coupled thermoelectric fields of voltage, current density, sources of Joule heat and temperature. The corresponding simulation was carried out taking into account the dependence of the conductive properties of the materials of the HPA components on temperature and pressure. The thermoelectric state of the HPA cubic cell was determined under the conditions of the insulating and conductive state of the 90 wt.% cBN–10 wt.% Al sample under consideration.

2. Calculations showed that under pressure of 2 GPa and heating current power of 6.6 kW for the insulating state of the sample the maximum temperature difference ΔT_{max} in it is 3 °C, for the conductive state – 14 °C. At a pressure of 4.2 GPa and a heating current power of 10.6 kW ΔT_{max} = 5 °C and ΔT_{max} = 23 °C, respectively, for the insulating and conductive states of the sample. Thus, the transition from the insulating to the conductive state of the sample leads to an increase in the maximum temperature difference in it. At the same time, the temperature conditions remain fairly uniform and acceptable for reaction sintering.

3. It was established that the transition of the sample from the dielectric to the conductive state caused by the melting of aluminum, the thermoelectric parameters of sintering begin to change at a sample resistance of ~ 10^{-3} Ω·m. This process is characterized by a drop in voltage with a simultaneous increase in current and temperature at the same heating current power of the HPA.

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О.П. Людвіченко, О.О. Лєщук, О.М. Анісін, М.П. Беженар

Чисельне визначення термоелектричних параметрів двостадійного спікання зразків системи cBN–Al у кубічній комірці високого тиску

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Проведено попереднє чисельне моделювання зв'язаних електричних і теплових полів в шестипуансонному АВТ за реакційного спікання масивних зразків PCBN. Передбачалося, що спікання надтвердого композита здійснюється із шихти cBN–Al. Для вирішення стаціонарної задачі електро- і теплопровідності скористались методом скінченних елементів. Провідні властивості матеріалів комірки були відкориговані таким чином, щоб відповідати умовам її термобаричного навантаження. Отримані результати дозволяють визначати рівень термоелектричних параметрів двостадійного реакційного спікання і оцінювати вплив зміни провідних властивостей комірки на тепловий стан зразків PCBN.

Ключові слова: полікристалічний кубічний нітрид бору (PCBN), апарат високого тиску (АВТ), тепловий стан, моделювання.