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Metallurgy

Calculation of the thermoplastic beryllium oxide slurry molding with ultrasonic activation

[1] Zhapbasbayev U.K., [1*]Ramazanova G.I., [2] Terekhov V.I., [3] Sattinova Z.K.

[1]Satbayev University, Almaty, Kazakhstan
[2] Kutateladze Institute of Thermophysics SB RAS, Novosibirsk, Russia
[2]L.N. Gumilev Eurasian National University, Astana, Kazakhstan

[*]Corresponding author email: g.ramazanova@satbayev.university

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ABSTRACT

The article presents the results of assessing thermal shrinkage during the formation of beryllium oxide ceramics using the hot casting method. The thermoplastic slurry is a composite system with a dispersion medium (binder) that has a very low thermal conductivity compared to the dispersed phase (beryllium oxide). Ultrasonic treatment reduces the viscosity of the slurry and improves its casting properties. The formation of beryllium oxide slurry is carried out without disrupting the integrity of the system and depends on the casting speed and temperature factors. The combined influence of these factors determines the casting properties of the slurry. Cooling - solidification of the slurry in the casting mold occurs in stages in the liquid, amorphous states with a phase transition, and in the viscoplastic state of the casting. The cooling rate of the casting at all stages depends on the cavity design, the rheological properties of the slurry, and the casting process parameters. It is important to maintain the integrity of the casting due to temperature shrinkage.

Keywords: thermoplastic slurry, formation, shrinkage, casting properties, beryllium oxide, ultrasonic treatment.

Information about authors:

Zhapbasbayev Uzak Kairbeekovic	*Doctor of Technical Sciences, Professor, Head of the Research and Production Laboratory "Modeling in Energy", Satbayev University, 22 Satpaev Street, 050013 Almaty, Kazakhstan. Email: uzak.zh@mail.ru*
Ramazanova Gaukhar Izbasarovna	*Candidate of physical and mathematical sciences, Leading Researcher. Research and Production Laboratory "Modeling in Energy", Satbayev University, 22 Satbayev str., 050013 Almaty, Kazakhstan. E-mail: g.ramazanova@satbayev.university*
Terekhov Victor Ivanovich	*Doctor of Technical Sciences, Professor, Chief Researcher of the Kutateladze Institute of Thermophysics, Siberian Branch of the Russian Academy of Sciences. 1 Academician Lavrentiev av., 630090 Novosibirsk, Russia. Email: terekhov@itp.nsc.ru*
Sattinova Zamira Kanaevna	*Candidate of physical and mathematical sciences, Associated Professor, L.N. Gumilev Eurasian National University, 2 Satpayev str., 010008 Astana, Kazakhstan. E-mail: sattinova.kz@gmail.com*

Introduction

Currently, products made from beryllium oxide (BeO) ceramics, obtained through the slurry casting technology, are widely used in the following applications [[1], [2], [3], [4], [5], [6]]:

1) Refractory material for crucibles used in melting metallic beryllium, uranium, and precious metals.

2) Insulators and heat sinks, substrates for transistors, and microchips in the electronics, radio, and electrical industries.

3) Windows and insulators for microwave (MW) technology.

4) Dielectric discharge tubes, resonators, and hollow dielectric waveguides in gas lasers covering a wide spectral range.

5) Heat pipes in cryogenic engineering, where thermal conductivity at temperatures ranging from 45 to 50 K can reach tens of thousands of W/(m·K).

6) Material for the heat-generating matrix element in nuclear reactors.

7) Neutron reflectors as part of neutron filters, with additives (such as boron) for protection against neutrons of various energies.

Beryllium oxide ceramics possess high chemical, thermal, and radiation resistance [[1], [2], [6]]. Unlike other types of oxide ceramics, it can absorb and reflect neutrons of various energies [[1], [2], [6]]. It has exceptional thermal conductivity, up to 320 W/(m·K) [[1], [2], [3], [4], [5], [6]], and a sound velocity of over 12,000 m/s [2]. Without the addition of impurities, BeO ceramics exhibit high electrical resistance and mechanical strength [[1], [2], [6]]. They are effective at transmitting various forms of electromagnetic radiation [[1], [2], [6]]. With the addition of TiO2, they can be used in powerful microwave energy absorbers [[2], [6]]. Beryllium oxide ceramics have a melting temperature of around 2843 K, while ceramics based on Al_2O_3 melt at only 2323 K [[2], [6]]. When cooled to 50 K, the thermal conductivity of BeO monocrystals sharply increases to 13,500 W/(m·K), which is also characteristic of BeO ceramics [[2], [3]].

The process of forming ceramics through hot casting involves several stages [[2], [6], [7], [8], [9], [10], [11]]: movement and heat exchange in the liquid state; movement and heat exchange in the amorphous state; movement and heat exchange in the solid-plastic state.

The hydrodynamics of the slurry in the casting mold fall into the category of physical processes related to flow and deformation. The slurry retains its configuration after exiting the feeder. Experimental findings indicate that within the range of possible casting speeds, the slurry's movement in the casting mold is laminar [[6], [7], [8], [9]]. The slurry enters the casting mold at temperatures of 75-80°C and cools down to 35-40°C, allowing the casting to be removed from the mold without warping [[6], [7], [9]]. During the stage of filling the mold cavity, it is crucial to ensure maximum disruption of the structure to obtain a homogeneous suspension [[2], [6], [9]]. This is achieved through ultrasonic treatment [[6], [11], [12], [13]].

During the cooling - solidification process, there is a change in the volume of the slurry, which results in shrinkage occurring in three stages: in the liquid, amorphous, and solid-plastic states [[2], [6], [8]]. The main technological problem solved at this stage is to achieve compensation of internal shrinkage [[6], [8], [13]]. The importance of this operation lies in the fact that the absence or incomplete compensation of internal shrinkage can lead to internal defects (cracks, porosity) in castings and products [[6], [8], [13]]. During solidification, it is necessary to minimize friction against the mold walls and maximize plasticity without disrupting the newly forming structure of the casting [[6], [8], [13]].

In the previous works of the authors [[7], [9], [15]], the results of experiments and calculations of the motion and heat exchange of beryllium oxide slurry with ultrasonic activation were presented. The experimental results established the process of forming a liquid slurry with a transition to a viscoplastic state. It was shown that the calculated and experimental data are in agreement and confirm the physical validity of the proposed mathematical model of the forming process [[7], [9], [15]].

This study presents calculations of the impact of casting speed and temperature factors on shrinkage during the formation of beryllium oxide slurry with ultrasonic activation.

Forming Process

Experimental Setup

The schematic of the ultrasonic casting setup is shown in Figure 1.

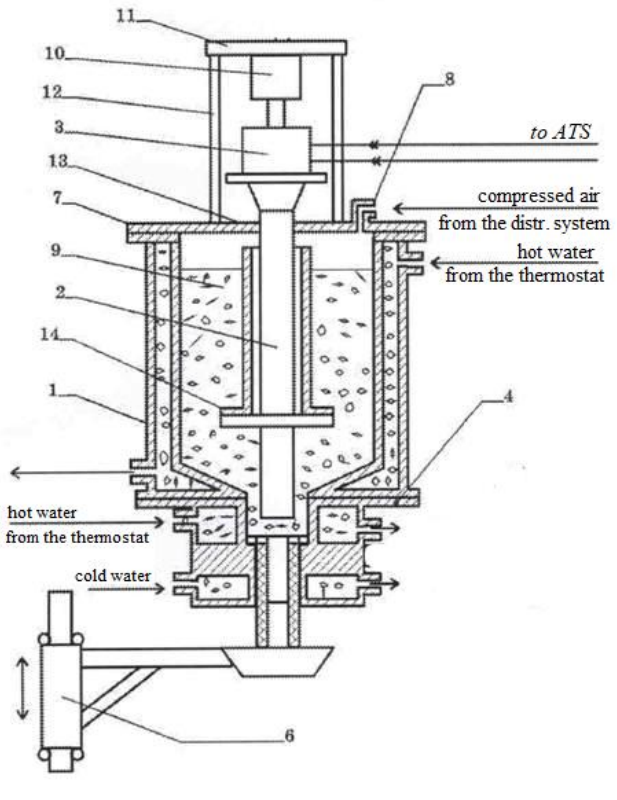


Figure 1 - Schematic of the experimental ultrasonic setup

The setup comprises a heated slurry tank 1 with an attached acoustic-technological system (ATS), consisting of a longitudinal waveguide 2 with a magnetostrictive transducer 3 of the PMS type, a three-loop filer 4, and a mechanism for controlling the movement of the billet 6.

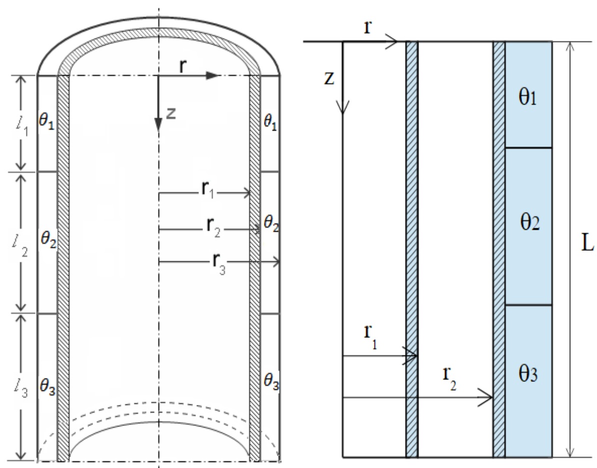
The ultrasonic generator USG2-4 was used as the power source for the acoustic systems. The main parameters of the setup are provided in Table 1.

Table 1 - Technical Characteristics of the Ultrasonic Casting Setup

Parameter Name	Value
General Information	
1. Power consumed by the equipment, kW	6.0
2. Operating voltage, V	220/380
3. Working tank volume, l	8
4. Casting speed (productivity), mm/min	10-120
Ultrasonic Equipment Data	
5. 5. Ultrasonic generator parameters: USG power, kW	4
6. Operating frequency, kHz	18±1.35
7. Operating voltage, V	220±44
8. Bias current, A	18±3
9. AFC accuracy, Hz	50
10. Radio interference level	According to GOST 23450-79
Utility product data	
11. Pressure, MPa	0.2
12. Water flow rate, m³/h	1.0
13. Water temperature, °C for the hot circuit of the die and slurry tank - for the warm circuit of the die - for the cold circuit of the die	 80 56 40
14. Compressed gas pressure, MPa	Up to 1.0

The die of the casting setup

The formation of thermoplastic beryllium oxide slurry takes place within the die of the casting setup [[6], [8]]. The die has a round-shaped cavity (Fig. 2a) or a ring-shaped cavity (Fig. 2b) with a cooling circuit.



a) b)

Figure 2 - Schematic diagram of the die:

a) round-shaped cavity; b) ring-shaped cavity

In the calculations, the round cavity has a radius of r1 = 0.006 m and a length of L = 0.089 m, while the ring-shaped cavity has radii of r1 = 0.006 m, r2 = 0.00675 m, and a length of L = 0.089 m. Liquid slurry enters the die cavity with an initial temperature of t0. As it progresses, the slurry mass cools down and solidifies. The slurry takes on a structural form, emerging from the round cavity as a rod (Fig. 2a) and as a tube from the ring-shaped cavity (Fig. 2b). The density of the slurry is variable and increases as it solidifies.

Mass and Heat Transfer Model

Rheology of Thermoplastic Slurry

In experiments [[6], [14]], BeO slurry exhibits the rheology of a non-Newtonian fluid with a yield stress and high viscosity. The effective (apparent) molecular viscosity of a viscoplastic fluid with a yield stress under shear can be expressed as [[15], [16], [17], [18]]:

$$\mu_{eff} = \begin{cases} \mu_p + \tau_0 |\dot{\gamma}|^{-1}, & \text{if } |\tau| \quad \tau_0 \\ \infty, & \text{if } |\tau| \le \tau_0 \end{cases} \tag{1}$$

Here, τ_0 is the yield stress, $|\tau| = \sqrt{\tau_{ij}\tau_{ij}}$ is the second invariant of the deviatoric stress tensor, μ_p is the plastic viscosity, $\dot{\gamma} = \sqrt{2S_{ij}S_{ij}} = S^2$ is the shear rate, $S_{ij} = 0.5\left(\frac{\partial U_i}{x_j} + \frac{\partial U_j}{\partial x_i}\right)$ is the strain rate tensor. The expression (1) corresponds to the Schwedoff-Bingham fluid model [18].

In regions where the shear stress is less than $\tau \le \tau_0$, the effective viscosity exhibits singular behavior. The effective viscosity can be represented as a smooth function using the regularization approach [19]:

$$\mu_{eff} \quad \mu_p + \tau_0 \frac{\lfloor 1 - \exp\left(-10^3 |\mathbf{S}|\right) \rfloor}{|\mathbf{S}|} \tag{2}$$

where m = 1000 is a regularization parameter [19]. Expression (2) helps overcome the difficulties in the presence of the yield limit for the Schwedoff-Bingham fluids.

Basic Equations

The forming process occurs in a steady-state mode. The length of the cavities is greater than their radial dimensions (Figure 1). Therefore, the motion equation can be written in the approximation of a narrow channel [7], taking into account the expression for effective viscosity (1):

$$\rho u\frac{\partial u}{\partial z}+\rho v\frac{\partial u}{\partial r}=-\frac{dp}{dz}+\frac{1}{r}\frac{\partial}{\partial r}\left(r\mu_{eff}\frac{\partial u}{\partial r}\right)+\rho g \quad (3)$$

$$\frac{\partial \rho u}{\partial z}+\frac{1}{r}\frac{\partial r\rho v}{\partial r}=0 \quad (4)$$

Heat exchange occurs along the length and radius of the cavity due to the cooling of the slurry. As experiments show, the change in the slurry's phase state occurs within a temperature range from 59 to 54°C [7]. In this case, the heat of phase transition is taken into account using a model of apparent heat capacity [[20], [21], [22], [23], [24], [25]]. The heat transfer equation, with the adopted assumptions, can be written as [[7], [26]]:

$$\rho u c_p\frac{\partial t}{\partial z}+\rho v c_p\frac{\partial t}{\partial r}=\frac{\partial}{\partial z}\left(\lambda\frac{\partial t}{\partial z}\right)+\frac{1}{r}\frac{\partial}{\partial r}\left(r\lambda\frac{\partial t}{\partial r}\right)+\mu_p\left(\frac{\partial u}{\partial r}\right)^2 \quad (5)$$

Here, z and r are the axial and radial coordinates, u and v are velocity components, p, ρ, t, c_p, μ_{eff}, μ_p, λ are pressure, density, temperature, apparent heat capacity coefficient, effective and plastic viscosity, and thermal conductivity of the slurry, respectively.

The pressure gradient is determined from the conservation of mass flow rate [27]:

$$\int_S \rho u dS=\rho_o u_o S \quad (6)$$

where S is the cross-sectional area of the cavity.

The boundary conditions for this problem are standard: for the slurry velocity on the cavity walls in the liquid state, no-slip conditions are applied; in the transition and solidifying zones, slip conditions are used. For the slurry temperature, Dirichlet conditions are applied to the outer cavity wall, while Neumann conditions are applied to the axis or inner cavity wall.

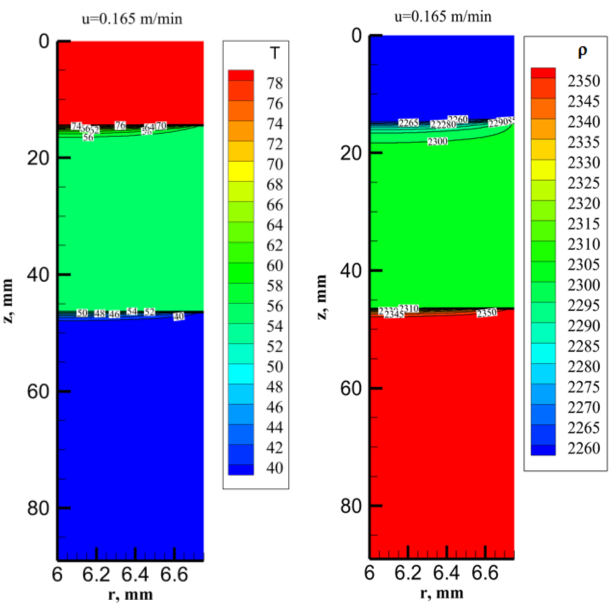
The dependencies of density, physicochemical properties of the slurry on temperature with US activation, and the consideration of phase transition heat using the apparent heat capacity method are provided in [7]. The system of equations (1)-(6) is solved by a numerical method [27]. The pressure gradient is determined by the method of splitting from the conservation of mass flow rate (6).

Verification of Calculation Data

Verification of the calculations was carried out using experimental data [[6], [15]] obtained for beryllium oxide slurry in a ring-shaped cavity. The calculations were performed under the same process parameters as in the experiments (Table 2). The cooling contour of the die is divided into three parts. In the first part, the cooling water temperature is θ_1 = 80°C, in the second part θ_2 = 56°C, and in the third part θ_3 = 40°C.

In the first cooling circuit, the wall temperature is θ_1 = 80°C. The temperature field shows a decrease in temperature from 80 to 78°C (Figure 3a), and the density field increases from 2260 to 2275 kg/m³ (Figure 3b).

In the second cooling circuit, the wall temperature is θ_2 = 56°C. The dynamic viscosity μ_p(t), density ρ (t), and yield shear stress τ_0(t) increase as the temperature decreases. The slurry temperature decreases from 78°C to 54°C (Figure 3a), and the density increases from 2275 kg/m³ to 2335 kg/m³ (Figure 3b).



a) b)

Figure 3 – Calculated data for temperature and density at a binder mass fraction ω = 0.117

In the third cooling circuit, the wall temperature is θ_3 = 40°C. The slurry temperature decreases from 54°C to 40°C (Figure 3a), and the density increases from 2335 kg/m³ to 2360 kg/m³ (Figure 3b).

The isotherm at t=59 °C represents the upper limit of the transition zone, while the isotherm at t=54 °C represents the lower limit. Within this zone, the slurry is in an amorphous state.

Table 2 - Slurry Parameters as a Function of Casting Speed in the Ring-Shaped Cavity [6]

Content of Binder in Slurry, Mass Fraction	Slurry Viscosity at T_0 = 80°C, Pa·s	Casting Ability of Slurry, mm	Casting Speed, mm/min	Mechanical Strength of Casting (Bending), MPa
0.117	2.80	89	165	8.17

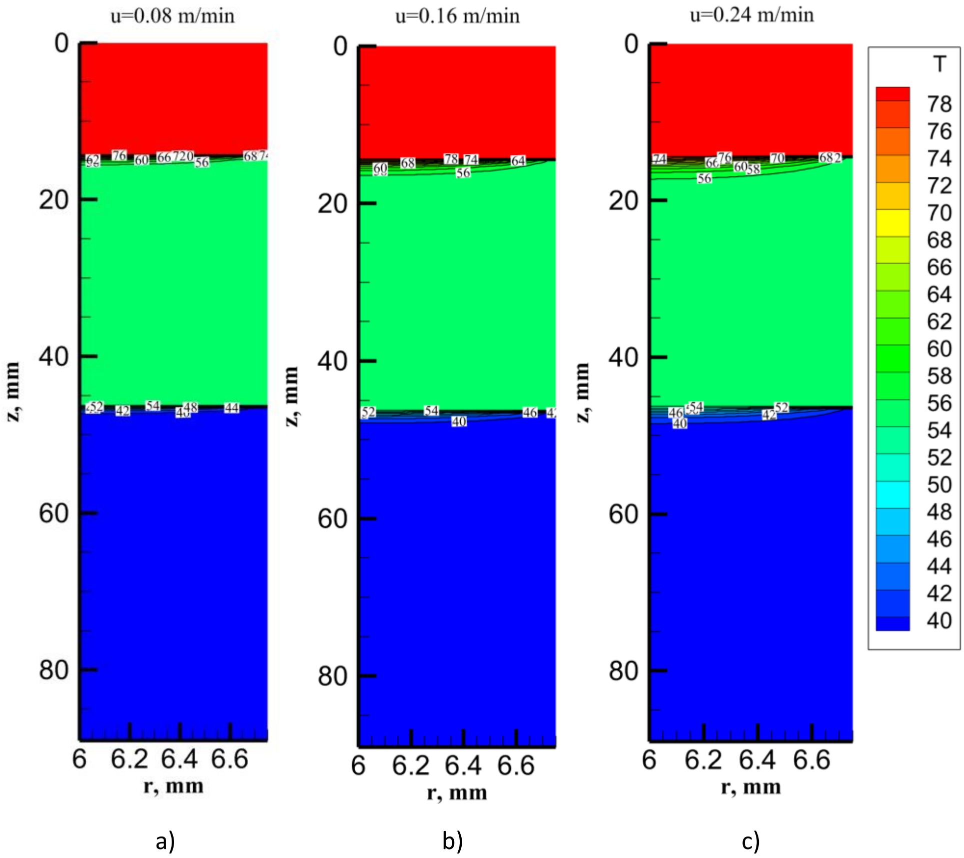


Figure 4 – Temperature field of BeO slurry in the ring-shaped cavity at different casting speeds and a binder mass fraction of ω = 0.117

The discussion of the calculation data

Figure 4 illustrates the temperature distribution in the ring-shaped cavity for three cooling circuits of the die with a cavity thickness of r_2–r_1=0.75 mm and a length of L = 89 mm. The casting speeds were u = 0.08 m/min, 0.16 m/min, and 0.24 m/min. The water temperatures in the cooling circuits are θ_1 = 80°C, θ_2 = 56°C, and θ_3 = 40°C.

The slurry temperature at the entrance to the ring-shaped cavity is constant across the section and equal to t_0 = 80 °C. In the first cooling circuit, the wall temperature is θ_1 = 80 °C, and in this region, the slurry temperature decreases slightly from 80°C to 78°C (Figure 4).

It is known that heat is transferred due to thermal conductivity against the flow of the slurry [26]. This explains the temperature reduction in the first part of the cavity. The temperature field determines the rheological and thermophysical properties of the slurry mass in its liquid state.

In the second cooling circuit, the wall temperature is θ_2 = 56°C. The dynamic viscosity $\mu_p(t)$, density $\rho(t)$, and yield shear stress $\tau_0(t)$ increase as the temperature decreases. The slurry slides along the cavity walls along the length of the second circuit. This causes the profile of the longitudinal velocity component downstream to become constant across the cavity cross-section

The increase in heat removal from the wall in the second cooling circuit results in a reduction in the temperature field (Figure 4). At the beginning of the second circuit, there is a region where the temperature field is variable and indicates the transition of the slurry from a liquid to an amorphous state. In this region, the temperature decreases from 78 to 54°C, marking the onset of a change in the slurry's aggregate state [[6], [7], [8]].

In the third cooling circuit, where the wall temperature is θ_3 = 40°C, the temperature decreases from 54 to 40°C. In the third part of the cavity, the slurry transitions from an amorphous to a viscoplastic state. The slurry's sliding motion on the wall ensures the continuity of the slurry and the preservation of the structural form of the casting without warping.

The increase in casting speed leads to an expansion of the transition zone from liquid to amorphous state and from amorphous to viscoplastic state (see Figure 4).

The variation in density reflects the transition of the slurry within the cavity from one structural state to another. Figure 5 illustrates the density field in the ring-shaped cavity at casting speeds of u = 0.08 m/min, 0.16 m/min, and 0.24 m/min. As shown in Figure 5, the slurry's density increases as the temperature decreases and the slurry cools along the length of the ring-shaped cavity.

In the first cooling circuit, there is a slight increase in slurry density, and the slurry remains in a liquid state. In the second cooling circuit, a significant increase in slurry density occurs, indicating the transition of the slurry from a liquid to an amorphous state. In this region, there is a change in the slurry's aggregate state with the crystallization of the binder material. Further increases in slurry density occur in the third circuit due to the solidification of the casting.

During the solidification stage, it is crucial to ensure compensation for the internal shrinkage of the casting. Without adequate compensation for internal shrinkage, internal defects such as shrinkage cavities and porosities can occur in castings and products.

In the ring-shaped cavity, the computational data consistently show a continuous change in density at all stages of forming, taking into account compensation for internal shrinkage through the supply of liquid slurry (Figure 5).

Therefore, the density field visually illustrates the entire forming stage with the shrinkage of beryllium oxide slurry.

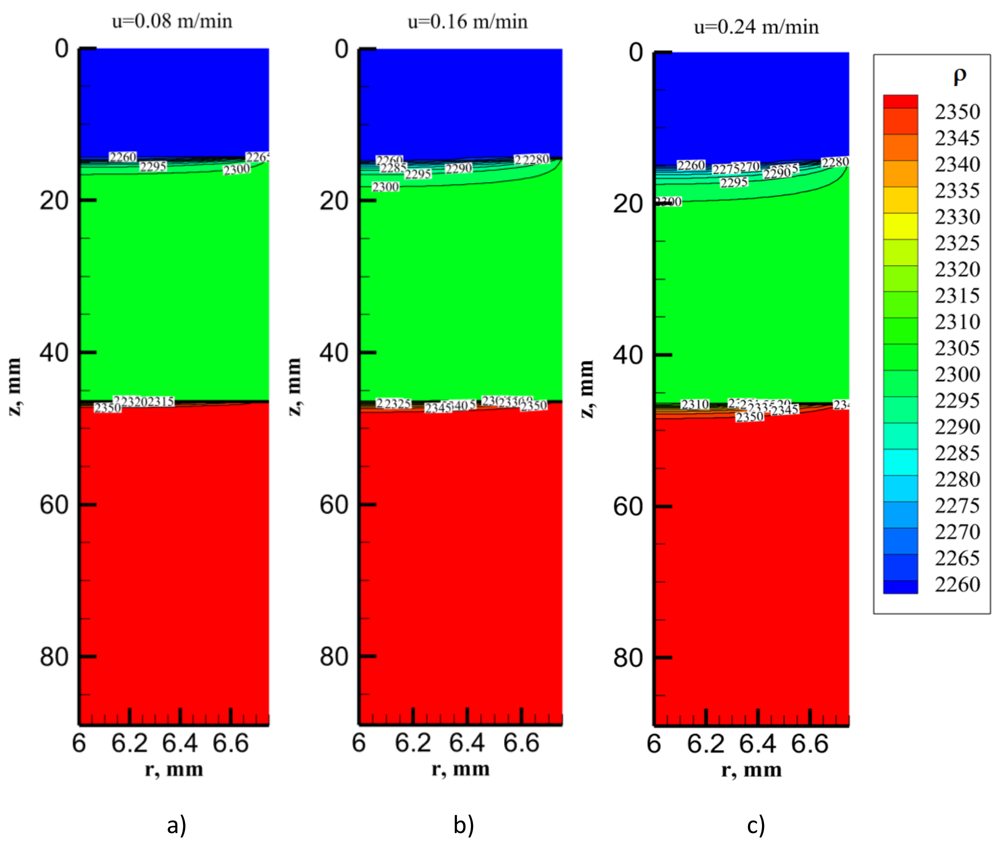
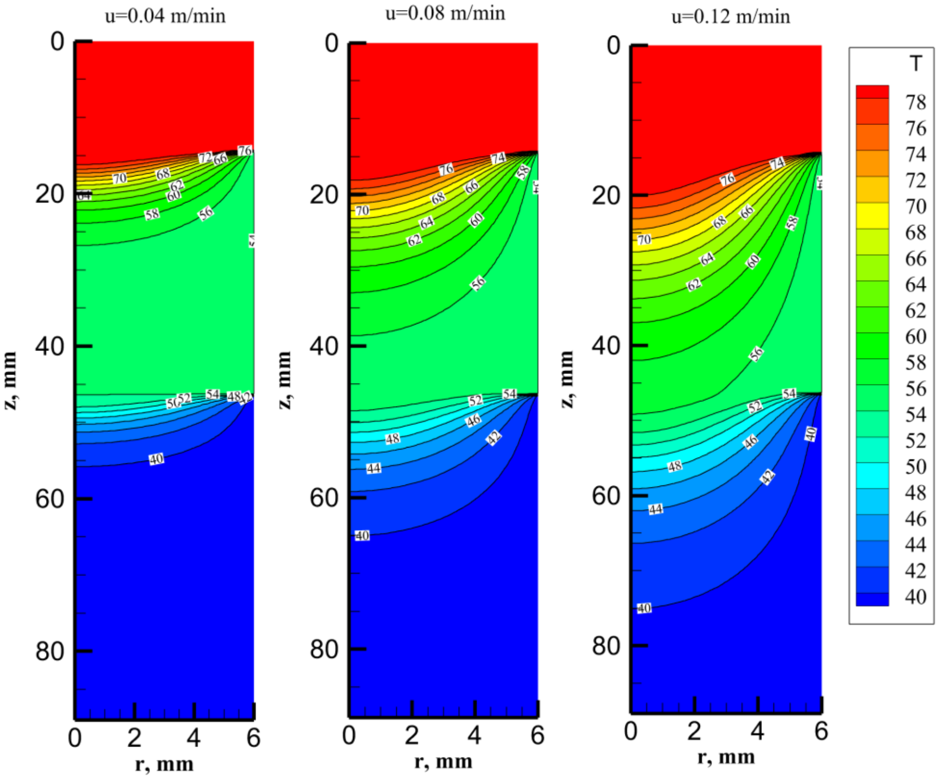
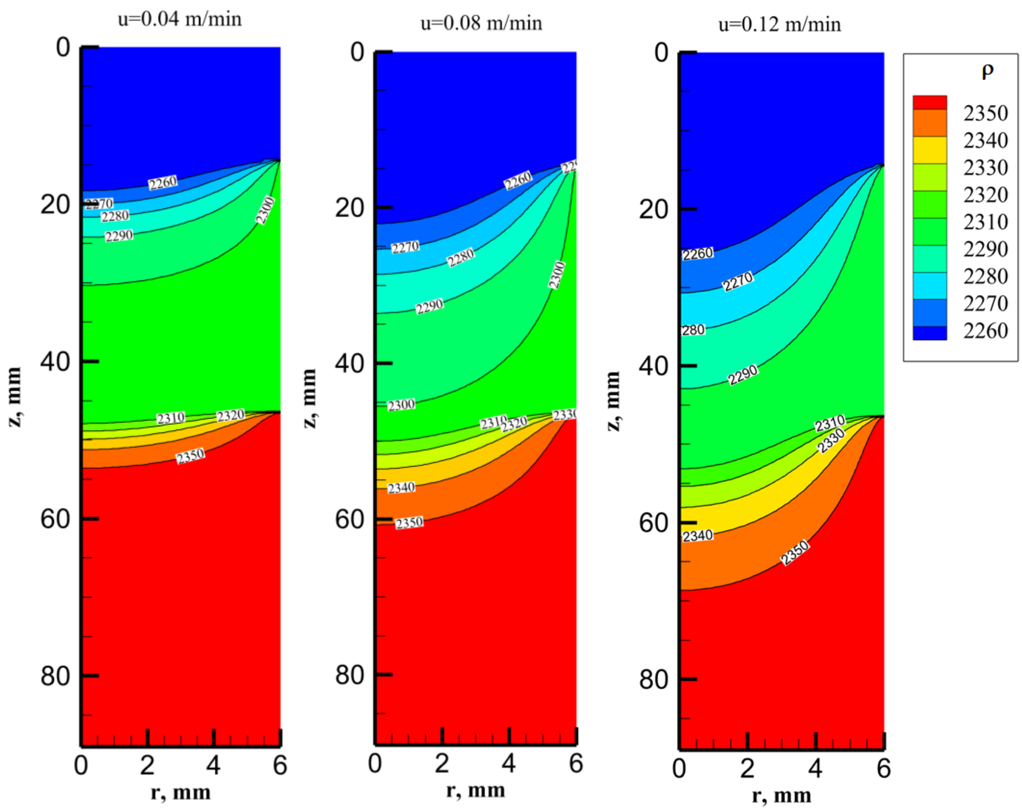


Figure 5 – Beryllium oxide slurry density field in the ring-shaped cavity at different casting speeds and a binder mass fraction of ω=0.117



a) b) c)

Figure 6 – Temperature field of BeO slurry in the round cavity at different casting speeds and a binder mass fraction of ω=0.117



a) b) c)

Figure 7 - Density field of BeO slurry in the round cavity at different casting speeds and a binder mass fraction of ω=0.117

The computed temperature data in the round cavity are shown in Figure 6 at casting speeds of u = 0.04 m/min, 0.08 m/min, and 0.12 m/min. The radius of the cavity is 0.006 m, and the length is 0.089 m. The temperature of the slurry at the inlet of the round cavity is t_0=80°C. In the cooling circuits, the water temperatures were as follows: θ_1= 80°C; θ_2= 56°C; θ_3= 40°C.

The casting speeds in the round cavity are lower than those in the ring-shaped cavity. In the ring-shaped cavity, a tube with a thin wall is formed, while in the round cavity, a rod is formed. The structural dimensions of the cavity determine the casting speed regimes for BeO ceramic products.

At a casting speed of u = 0.04 m/min, the temperature distribution in the contact zones causes only minor thermal deformation of the casting (Figure 6).

However, temperature distributions in the contact zones at casting speeds of u = 0.08 m/min and 0.12 m/min can result in significant thermal deformation of the casting (Figure 6).

The results of experimental studies [[6], [8], [13]] have shown that the increase in density (specific volume) during the cooling process in the liquid state amounts to 5-6%. However, during the cooling of slurry in amorphous and viscoplastic state there is a 70-80% of density (specific volume) increase due to temperature shrinkage [[6], [8], [13]].

From this, it can be inferred that the speed and temperature factors, along with the structural characteristics of the mold cavity, determine the changes in density (specific volume) in the amorphous and viscoplastic states of the slurry.

In the ring-shaped cavity, the increase in density represents only a small part of the amorphous and viscoplastic states of the slurry. Therefore, it can be considered that the flow rate of slurry with feeding provides compensation for thermal shrinkage.

In the round cavity, at a casting speed of u = 0.04 m/min, the density increase occupies a small part of the amorphous and viscoplastic states of the slurry (see Figure 7). The slurry flow rate with feeding can provide compensation for thermal shrinkage.

At casting speeds of u = 0.08 m/min and 0.12 m/min, density increases occupy a significant part of the amorphous and viscoplastic states of the slurry (see Figure 7). In these cases, it may be difficult to compensate for thermal shrinkage by supplying a slurry flow.

Conclusion

The calculation results illustrate the entire process of forming beryllium oxide slurry, taking into account the changes in its aggregate state. Ultrasonic treatment improves the rheological properties and enhances the flowability of the slurry in the mold cavity.

The calculations demonstrate the influence of casting speed, temperature factors, and the structural parameters of the mold cavity on the cooling-solidification process of the casting. In the annular cavity, at casting speeds of u = 0.08 m/min, 0.16 m/min, and 0.24 m/min, the increase in density occupies only a small part of the amorphous and solid-plastic state of the slurry. The flow of liquid slurry with feeding in the annular cavity compensates for the thermal shrinkage of the slurry.

In the round cavity, compensation for the thermal shrinkage of the slurry was achieved only at a casting speed of u = 0.04 m/min. However, at casting speeds of u = 0.08 m/min and 0.12 m/min, significant areas of density change due to thermal deformation exist. In these cases, it may be difficult to compensate for thermal shrinkage by supplying a slurry flow.

In the calculations, it is possible to determine the conditions for forming a slurry with solidification shrinkage using the hot casting method, which allows obtaining a cast product with a homogeneous structure of beryllium oxide.

Conflict of interest. On behalf of all the authors, the correspondent author declares that there is no conflict of interest.

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[1] Жапбасбаев Ұ.Қ., [1*] Рамазанова Г.І., [2] Терехов В.И.,[3] Саттинова З.Қ.

[1]Сәтбаев университеті, Алматы, Қазақстан
[2]С.С. Кутателадзе атындағы Жылуфизика институты РҒА СБ, Новосібір, Ресей
[3]Л.Н. Гумилев атындағы Еуразия ұлттық университеті, Астана, Қазақстан

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ТҮЙІНДЕМЕ
Мақалада бериллий оксидінің керамикасын ыстық құю әдісімен қалыптау кезіндегі термиялық шөгуді бағалау нәтижелері берілген. Термопластикалық шликер – дисперсиялы ортамен (бериллий оксиді) салыстырғанда өте төмен жылу өткізгіш дисперсті фазасы (байланыстырушы) бар композициялық жүйе. Ультрадыбыстық өңдеу шликердің тұтқырлығын төмендетеді және оның құйылу қасиетін жақсартады. Бериллий оксидінің шликерін қалыптастыру жүйенің тұтастығын бұзбай жүзеге асырылады және құюдың жылдамдығы мен температуралық факторларына байланысты. Осы факторлардың бірлескен әсері шликердің құю қасиеттерін анықтайды. Құйма қалыптағы шликердің салқындау-қатуы фазалық ауысумен құйманың сұйық, аморфты және тұтқырлы пластикалық күйінде кезең-кезеңімен өтеді. Барлық кезеңдердегі құйманы салқындату жылдамдығы қуыстың конструкциясына, шликердің реологиялық қасиеттеріне, сонымен қатар құйманың жұмыс параметрлеріне байланысты. Бұл жағдайда құйманың температуралық шөгуіне байланысты оның тұтастығын сақтау маңызды.

Түйін сөздер: термопластикалық шликер, қалыптау, шөгу, құю қасиеті, бериллий оксиді, ультрадыбыстық өңдеу.

Авторлар туралы ақпарат:

Жапбасбаев Ұзақ Қайырбекұлы — *Техника ғылымдарының докторы, профессор, "Энергетикадағы модельдеу" ғылыми-өндірістік зертханасының меңгерушісі, Сәтбаев университеті, Сәтбаев көшесі, 22 үй, 050013 Алматы, Қазақстан. Email: uzak.zh@mail.ru*

Рамазанова Гаухар Ізбасарқызы — *Физика-математика ғылымдарының кандидаты, жетекші ғылыми қызметкер, «Энергетикадағы модельдеу» ғылыми-өндірістік зертханасы, Сәтбаев университеті, Сәтбаев көшесі, 22 үй, 050013 Алматы, Қазақстан. Электрондық пошта: g.ramazanova@satbayev.university*

Терехов Виктор Иванович — *Техника ғылымдарының докторы, профессор, РҒА Сібір бөлімі С.С. Кутателадзе атындағы Жылуфизика институтының бас ғылыми қызметкері. Ресей, 630090, Новосібір қ., Ак. Лаврентьев даңғылы, 1. Email: terekhov@itp.nsc.ru*

Саттинова Замира Қанайқызы — *Физика-математика ғылымдарының кандидаты, қауымдастырылған профессор, Л.Н. Гумилев атындағы Еуразия ұлттық университеті, Сәтбаев көшесі, 2 үй, 010008, Астана, Қазақстан. Email: sattinova.kz@gmail.com*

Расчет формования термопластичного шликера оксида бериллия с ультразвуковой активацией

[1]Жапбасбаев У.К., [1*]Рамазанова Г.И., [2] Терехов В.И.,[3]Саттинова З.К.

[1]Satbayev University, Алматы, Казахстан
[2]Институт теплофизики им. С.С. Кутателадзе СО РАН, Новосибирск, Россия
[3]Евразийский национальный университет имени Л.Н. Гумилева, Астана, Казахстан

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АННОТАЦИЯ
В статье приводятся результаты оценки термической усадки при формовании керамики оксида бериллия способом горячего литья. Термопластичный шликер представляет собой композиционную систему с дисперсионной фазой (связующее вещество), имеющей очень низкую теплопроводность, по сравнению с дисперсной фазой (оксид бериллия). Ультразвуковая обработка снижает вязкость шликера и улучшает его литейное свойство. Формования шликера оксида бериллия проводится без нарушения сплошности системы и зависит от скоростных и температурных факторов литья. Совокупное влияние этих факторов определяет литейное свойство шликера. Охлаждение-отвердевание шликера в литьевой форме происходит поэтапно в жидком, аморфном состояниях с фазовым переходом и вязкопластичном состоянии отливки. Темп охлаждения отливки на всех этапах зависит от конструкции полости, реологических свойств шликера, а также от режимных параметров литья. При этом важным является соблюдение сплошности отливки из-за температурной усадки.

Ключевые слова: термопластичный шликер, формование, усадка, литейное свойство, оксид бериллия, ультразвуковая обработка.

Жапбасбаев Узак Каирбекович	***Информация об авторах:*** *Доктор технических наук, профессор, заведующий научно-производственной лабораторией "Моделирование в энергетике", Satbayev University, ул. Сатпаева 22, 050000, Алматы, Казахстан. Email: uzak.zh@mail.ru*
Рамазанова Гаухар Избасаровна	*Кандидат физико-математических наук, ведущий научный сотрудник. Научно-производственная лаборатория "Моделирование в энергетике", Satbayev University, ул. Сатбаева, 22, 050013 Алматы, Казахстан. E-mail: g.ramazanova@satbayev.university*
Терехов Виктор Иванович	*Доктор технических наук, профессор, главный научный сотрудник Института теплофизики им. С.С. Кутателадзе СО РАН. Россия, 630090, Новосибирск, пр. Ак. Лаврентьева, 1. Email: terekhov@itp.nsc.ru*
Саттинова Замира Канаевна	*Кандидат физ.-мат. наук, Ассоциированный профессор, Евразийский национальный университет имени Л.Н. Гумилева, Казахстан, 010008 Астана, ул. Сатпаева, 2. E-mail: sattinova.kz@gmail.com*

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