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MONOLITHIC HEAT-INSULATING LININGS WITH CELLULAR CERAMIC FIBER STRUCTURE

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An optimum composition of mixtures that gives linings with the requisite density and admissible shrinkage is calculated. It is established that a homogeneous mixture is obtained after mixing the components in a double-shaft vibromixer. The effects of the mixture composition and the conditions of heating of the lining on the structure of the material are investigated. The acoustic emission method is used to choose the heat treatment regime and study destructive phenomena in a monolithic lining during the first firing and cooling. The main properties of the cellular ceramic fiber material are described.

A critical analysis of the current state of the methods for producing monolithic linings for industrial furnaces shows that they are mainly produced by guncreting, ramming, vibration molding, and casting [1 – 3]. All these methods have the deficiency of being highly power-consuming and requiring complex equipment, which decreases the efficiency of monolithic linings. This means that the manufacturing technology has to be improved.

In the last 15 – 20 years fiber-reinforced refractory materials have become popular due to their various advantages in service over conventional materials [4]. Therefore, there are substantial possibilities for broad development of the production of monolithic heat insulation. Research and industrial experiments on the use of monolithic heat insulation from fiber-reinforced materials on a clay binder by the pouring in of mixtures with a moisture content of 300% have given positive results. However, the prolonged drying necessitated by the evaporation of a large amount of mixing water has impeded the introduction of this technology into industry.

In this connection much attention is devoted to the method of self-compacting mixtures; here the design of the monolithic lining does not require mechanical means of compaction and may be used in the most complex sites of the heating devices, and the installation of the lining is reduced to preparing a forming mixture, erecting the form, and placing the mixture between the form and the casing of the heating device. In heating a self-compacting mixture the polystyrene foams and the moisture is squeezed out through the perforations of the form (compaction of the lining). We can expect that this technology for preparing a monolithic lining will have the following advantages over existing methods:

– uniform density and strength over the entire volume of the lining at a relatively low pressure created inside the system;

– absence of porosity in the structure;

– the possibility of obtaining large-sized linings with a complicated configuration under the effect of the internal pressure;

– absence of shrinkage in drying due to the squeezing-out of the main portion of water at the moment of foaming of polystyrene grains.

Scientific investigations on creating such a technology concern unilateral heating of the self-compacting mixture, the directed first firing and cooling of the lining, and the structure of the material and destructive processes.

We investigated MKRV mullite-silica fiber (TU 14-8-107 – 74), DN-1 clay, and foaming bead polystyrene of fraction No. 5.

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TABLE 1. Variable Factors and Variation Ranges of the Factors in the Cellular Fiber Ceramic Material

Parameters	Values of variables			
	natural		coded	
	X_1, %	X_2, %	X_1	X_2
Main level	2.5	10	0	0
Lower level	1	6	–	–
Upper level	4	14	+	+
Variation range	1.5	4	–	–

TABLE 2. Results of Statistical Processing of Experimental Data*

Number of experiment	X_1	X_2	Y_1^e, MPa	Y_1^c, MPa	$Y_1^e - Y_1^c$, MPa	Y_2^e, %	Y_2^c, %	$Y_2^e - Y_2^c$, %
1	–	–	1.9	1.967	0.067	1.22	1.139	–0.081
2	+	–	1.3	1.267	–0.033	0.32	0.326	0.006
3	–	+	1.8	1.800	0.000	1.23	1.206	–0.024
4	+	+	1.1	1.100	0.000	0.38	0.392	0.012
5	–	0	2.1	2.033	–0.067	0.92	1.026	0.106
6	+	0	1.3	1.333	0.033	0.23	0.212	–0.016
7	0	–	1.4	1.367	–0.033	0.31	0.386	0.076
8	0	+	1.2	1.200	0.000	0.44	0.452	0.012
9	0	0	1.4	1.432	0.033	0.36	0.272	–0.088

* The index "e" is used for "experimental," the index "c" is used for "calculated."

Computation of the optimum composition of mixtures by the method of mathematical design of experiment. This consists in determining the proportion of the components that would ensure the requisite mobility of the mixture and also the strength and thermal resistance of the lining. It should be taken into account that the requirements on the shrinkage of the material of a monolithic lining are rigorous. The total shrinkage in drying and firing should be at most 3% and the residual linear shrinkage should be 1.5%.

We investigated the effect of the following factors on the properties of a cellular ceramic fiber material: X_1 – the fiber-to-clay ratio, %, X_2 – the content of bead polystyrene, %. The influence of X_1 and X_2 on the ultimate compressive strength of the fired articles (Y_1, MPa) and the firing shrinkage (Y_2, %) was investigated too. The variable factors and the variation intervals are presented in Table 1. The experimental results were processed by the method of multiple regression analysis using a computer.

The algorithm for computer calculation of the coefficients of the regression equation is based on the method of matrix inversion. The results are presented in Table 2.

As a result of the computation we obtained the regression equations

$$Y_1 = 1.982 - 0.789X_1 + 0.167X_2 + 0.111X_1^2 - 0.009X_2^2,$$

$$Y_2 = 2.75 - 1.04X_1 - 0.175X_2 + 0.154X_1^2 + 0.009X_2^2.$$

The significance of the regression coefficients was tested by Student's criterion, and the adequacy of the obtained models of strength (Y_1) and shrinkage (Y_2) was tested by Fisher's criterion.

The results of the statistical analysis of the equations showed that the adequacy of the models is not rejected with a probability of 0.95. The regression equations were used to construct isolines of strength and shrinkage (Fig. 1). Analysis

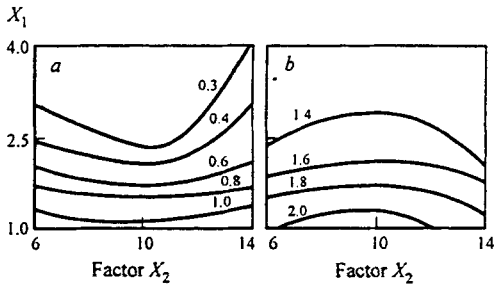

Fig. 1. Level lines of shrinkage (*a*) and ultimate compressive strength (*b*) of the cellular ceramic fiber material as functions of X_1 and X_2.

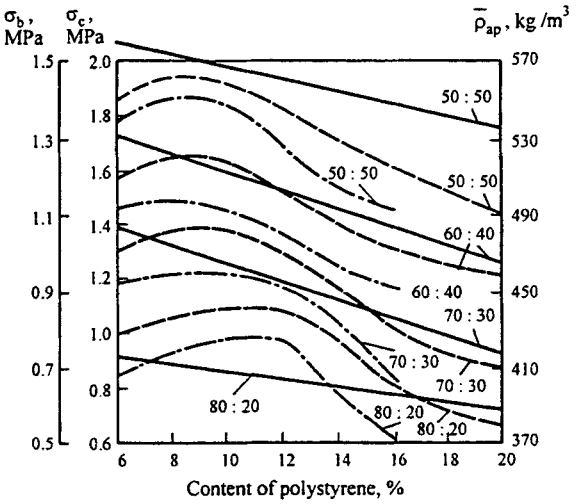

Fig. 2. Variation of the mean apparent density ρ_{ap} (——) and the ultimate bending σ_b (– · – · –) and compressive σ_c (– – –) strengths of fired specimens as a function of the content of bead polystyrene; the numbers at the curves indicate the fiber-to-clay ratio, %.

of the lines of the equations of the response function (Y_1 and Y_2) led us to the following conclusions:

– the strength of the cellular ceramic fiber material is affected by the ratio of the components in the mixture (fiber / clay), i.e., by factor X_1;

– the optimum content of bead polystyrene for the given ratios of fiber and clay is 8 – 12% (Fig. 2);

– the shrinkage phenomena in the cellular ceramic fiber material increase with decrease in the fiber-to-clay ratio, i.e., factor X_1.

After analyzing the lines of the equations of strength and shrinkage the optimum composition of the mixture was chosen to be $X_1 = 2.5$, $X_2 = 8 - 12\%$, which corresponds to a mixture containing 70% MKRV fiber, 30% DN-1 clay, and 8 – 12% (above 100%) bead polystyrene No. 5.

The chosen composition gives articles with sufficient strength and admissible shrinkage.

Figure 2 presents the dependences of the mean apparent density and the ultimate compressive and bending strengths of fired specimens on the amount of bead polystyrene intro-

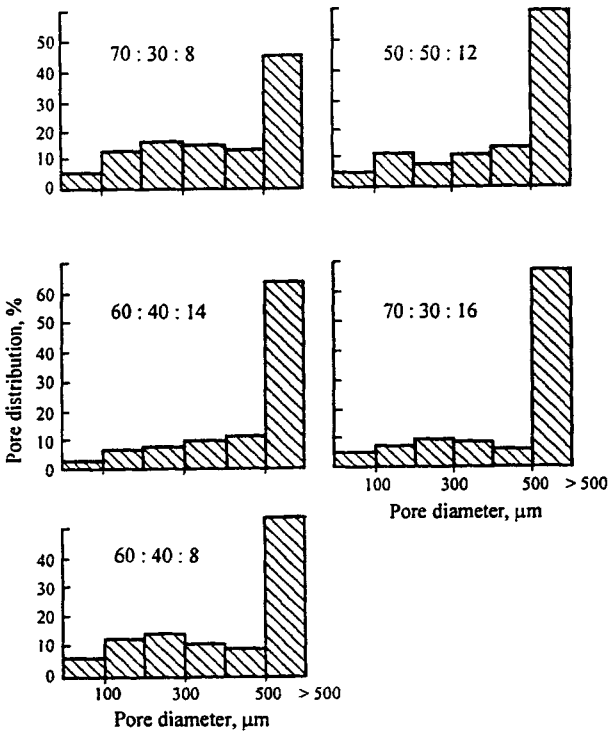

Fig. 3. Size distribution of pores in the cellular ceramic fiber material as a function of its composition.

duced into the charge. These experimental data correspond to the computed content of polystyrene in the forming mixture (8 – 12%). Here the maximum use of its compacting force is attained. The decrease in the strength at a high polystyrene content can be explained by weakening of the free cross-sectional area of the porous body, a decrease in the contact surface of the solid phase, and stress concentration in the pore walls under the load.

It was established that in order to obtain a homogeneous mixture and a uniform distribution of bead polystyrene the components should be agitated in a double-shaft vibromixer at a frequency of 3000 vibrations per min, an amplitude of 1.5 mm, a rotational speed of the shaft of 200 rpm, and a mixing time of 4 – 4.5 min.

Proceeding from the rheological properties of the forming clay-fiber-polystyrene mixtures, their initial moisture content was established to be (180 ± 5)%. Such a mixture fills the form for monolithicic structures by gravity without using additional forces.

The essence of the method for obtaining monolithic linings from self-compacting mixtures consists in the following. Lump wool is loaded into the vibromixer, then the binder (clay), polystyrene, and water are added, and the mixture is agitated. The resulting mixture is placed in a closed perforated volume and subjected to unilateral heating at 190 – 200°C. Investigations show that the process is completed in 5 h. After the form is removed, the moisture content of the lining is nonuniform over its thickness and fluctuates from 20% on the hot side to 100% on the cold side, which leads a difference in the mechanical properties of the material layers over the cross section. However, at 180 – 200°C the lining attains a monolithicic structure to a depth of 10 – 20 mm due to fusing of polystyrene, which makes it possible to conduct further forced drying of the lining without the form. The drying time after removal of the form is 20 – 21 h, during which no air shrinkage is detected.

We investigated the possibility in principle of obtaining a monolithic without a clay binder on the basis of the theoretical assumption that the wool fibers have a developed specific surface and numerous contacts among each other. By compressing such a system from the inside we can obtain a rather strong skeleton. The method developed consists in the following. The refractory fibers, polystyrene, and water are agitated in a vibromixer. The mixture with a moisture content of 250% is placed in a closed perforated volume and heat-treated at 200°C. As a result of polystyrene foaming, the moisture is squeezed out and the fibers are compressed. After completing the regime of the furnace, the polystyrene is burnt off and the material acquires a uniform fiber-cellular structure. Results of this investigation are presented in Table 3.

TABLE 3. Effect of the Mixture Composition on the Structure of the Material

Batch of specimens	Mixture composition,* %	Closed porosity, %	Amount of pores, %		Total porosity, %	Size distribution (%) of pores, μm					
			merged	cracklike		10 – 100	100 – 200	200 – 300	300 – 400	400 – 500	> 500
17	70 : 30 : 8	36.35	17.10	1.01	54.46	5.7	11.8	12.7	12.5	12.3	45.0
16	60 : 40 : 8	35.97	16.01	2.92	54.92	5.1	11.8	12.0	9.4	9.3	52.1
14	50 : 50 : 12	33.70	21.14	4.00	58.84	4.6	9.7	7.6	9.7	11.5	56.9
10	60 : 40 : 14	35.20	23.00	2.50	60.70	2.1	7.2	7.8	10.0	10.2	62.9
11	70 : 30 : 16	37.0	24.06	1.10	62.16	3.7	7.2	8.6	8.4	5.1	67.1
I	70 : 30 : 8	35.96	17.01	0.20	53.17	5.8	11.5	13.0	13.5	14.0	42.2
II	70 : 30 : 8	36.0	17.20	0.86	54.06	5.3	11.8	12.8	13.2	13.5	13.4
III	70 : 30 : 8	30.10	18.00	8.13	56.23	5.2	12.1	11.9	14.0	9.4	47.4
IV	70 : 30 : 8	29.00	17.90	9.00	57.00	5.3	10.7	11.0	12.1	11.6	49.3

* The fiber / clay / polystyrene proportions are presented.

This method can be used for any kind of fiber. Heating devices can be repaired without being shut down by injecting the mixture into poorly accessible places, because the absence of the ceramic component will not cause shrinkage phenomena.

Effect of the mixture composition and the heating conditions for the lining on the structure of the material. The macrostructure of the lining determines the behavior of the material in service. Therefore we should know how the contents of polystyrene, clay, and fiber as well as the rate of temperature increase in the first firing affect its properties. Results of this investigation are presented in Table 3 and in histograms (Fig. 3).

We begin with the effect of polystyrene, whose content in the mixture is 8, 12, 14, and 16%. It should be noted that the porosity (total) of the material with 8% polystyrene does not exceed 55% in all the experiments and that for 12, 14, and 16% polystyrene it amounts to 58.84, 60.70, and 62.16%, respectively. The content of pores with a diameter exceeding 500 μm increases here, which is caused by aggregation of polystyrene granules and pore coalescence. By increasing the clay component from 30 to 50% we increase the number of cracklike pores from 1 to 4% (specimens of batches 17, 16, 14). However, in the specimens of batch 14 the content of polystyrene is somewhat higher (12%) and the number of merged pores in them is also somewhat higher than for the two preceding batches. In the same specimens with an increase in the content of the clay component the amount of fibers decreases from 70% to 60 and 50%, and the closed porosity amounts to 36.35, 37.97, and 33.70%, respectively. The size distribution of the pores in these batches gives histograms of a similar nature.

The effect of the rate of temperature growth on the structure of the lining was investigated on specimens of batches I – IV for the same composition of the mixture, namely, 70% fiber, 30% clay, 8% polystyrene. The rate of temperature growth during firing of specimens of batches I – IV was 5, 10, 15, and 20°C/min, respectively (Fig. 4). The total porosity in the specimens of batches I and II is rather close and amounts to 53.17 and 54.06%. The size distribution of the pores is also identical and is close to that in the specimens of batch 17.

A sharper increase in the firing temperature (to 15 and 20°C/min) leads to a more marked increase in the amount of cracklike pores (8 – 10%) and the total porosity (56 – 57%). The amount of pores over 500 μm in size increases somewhat (see Fig. 4). It should be noted that all the specimens are characterized by a rather uniform pore distribution over the area of the polished section.

Thus we have established that

– the pore distribution in the cellular ceramic fiber material is uniform;

– the pore distribution is similar and differs mainly in the amount of pores with sizes exceeding 500 μm;

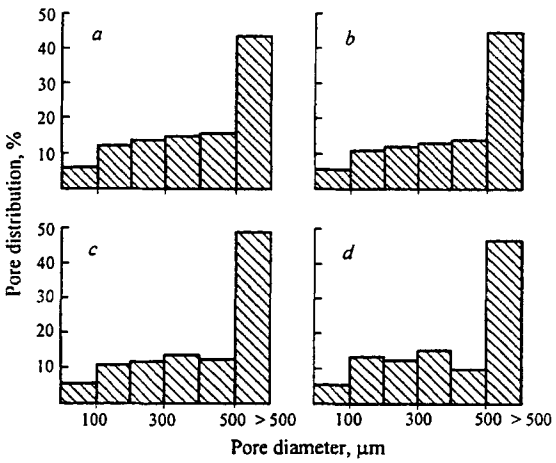

Fig. 4. Size distribution of pores in the monolithic lining as a function of the heating rate: *a*) 5°C/min; *b*) 10°C/min; *c*) 20°C/min; *d*) 15°C/min.

– an increase in the content of polystyrene increases the total porosity and the proportion of merged pores and pores over 500 μm in size;

– an increase in the amount of fiber leads to an increase in the proportion of closed pores and pores with sizes exceeding 500 μm;

– an increase in the content of the clay component leads to the formation of cracklike pores;

– an increase in the rate of growth of the firing temperature above 10°C/min leads to the appearance of a considerable amount of cracks.

Choice of heat-treatment regimes and investigation of destructive phenomena in a monolithic lining during the first firing and cooling (the method of acoustic emission). Stress waves in a monolithic lining are initiated by the effect of high temperatures. This process of physical action is reduced to a mechanical (force) action. The loading is equivalent to heating of the specimen and the unloading is equivalent to its cooling.

The influence of technological factors on the physicotechnical properties of the lining was studied using mathematical modeling of experiment by the method of Greco-Latin square.

We studied the effect of the following factors on the properties of a cellular ceramic fiber lining: X_1 – the heating rate, °C/min; X_2 – the cooling rate, °C/min; X_3 – the lining thickness, mm; X_4 – the lining diameter, mm. The effect of X_1, X_2, X_3, X_4 on the sum of the pulses in heating and cooling (Y_1, Y_2) and the ultimate compressive and bending strengths (Y_3, Y_4, MPa) was determined. The variable factors and the variation intervals are presented in Table 4.

The results were computer-processed by the method of multiple regression analysis. The algorithm of computer calculation of the coefficients of the regression equation is based

TABLE 4. Variable Technological Factors of a Monolithic Cellular Ceramic Fiber Lining and Levels of Their Variation

Levels of factors	Values of variables			
	X_1, °C/min	X_2, °C/min	X_3, mm	X_4, mm
1	20	20	30	120
2	15	15	35	130
3	10	10	40	135
4	5	5	45	140
5	2	2	52	153

on the method of matrix inversion. The computation gave us the following regression equations:

$$Y_1 = 24843.652344 - 36.367233X_1 - 228.380005X_3 - 435.939697X_2 - 192.612X_4 + 1.389202X_1^2 + 0.460648X_1X_2 - 0.266163X_1X_3 + 1.844752X_1X_4 + 1.093581X_2^2 + 1.926677X_2X_3 + 2.345963X_2X_4 + 1.47510X_3X_4 + 0.311114X_4^2,$$

$$Y_2 = 6516.019531 + 377.766846X_1 + 43.957764X_2 - 8.940748X_3 - 80.902557X_4 - 5.724776X_1^2 - 0.729599X_1X_2 - 1.151128X_1X_3 - 0,651463X_1X_4 + 0.879378X_2^2 + 0.954967X_2X_3 - 0.351717X_2X_4 - 0.549130X_3^2 + 0.455628X_3X_4 + 0.264579X_4,$$

$$Y_3 = -8.899843 - 0.115583X_1 + 0.010255X_2 + 0.164169X_3 + 0.106942X_4 - 0.001196X_1^2 + 0.00126X_1X_2 + 0.000188X_1X_3 + 0.000601X_1X_4 - 0.001044X_2X_3 - 0.00155X_3X_4 - 0.000239X_4^2,$$

$$Y_4 = -1.691508 - 0.027463X_1 - 0.030763X_2 + 0.099081X_3 + 0.012586X_4 - 0.001040X_1^2 + 0.001777X_1X_2 - 0.000616X_3^2 - 0.000343X_3X_4.$$

The significance of the regression coefficients was tested by Student's criterion, and the adequacy of the models of the sum of the pulses in heating (Y_1) and cooling (Y_2) and the ultimate compressive (Y_3) and bending (Y_4) strengths was tested by Fisher's criterion. The results of the statistical analysis of the equations showed that the adequacy of the models is not rejected with a probability of 0.95.

Analysis of the regression equations leads to the following conclusions:

– the sum of the pulses in heating (Y_1) depends mainly on the heating rate (X_1) and the diameter of the lining (X_4);

– the sum of the pulses in cooling (Y_2) depends on the heating rate (X_1), the cooling rate (X_2), and the lining diameter (X_4);

– the ultimate compressive strength (Y_3) and the ultimate bending strength (Y_4) depend on the heating rate (X_1), the cooling rate (X_2), and the lining diameter (X_4).

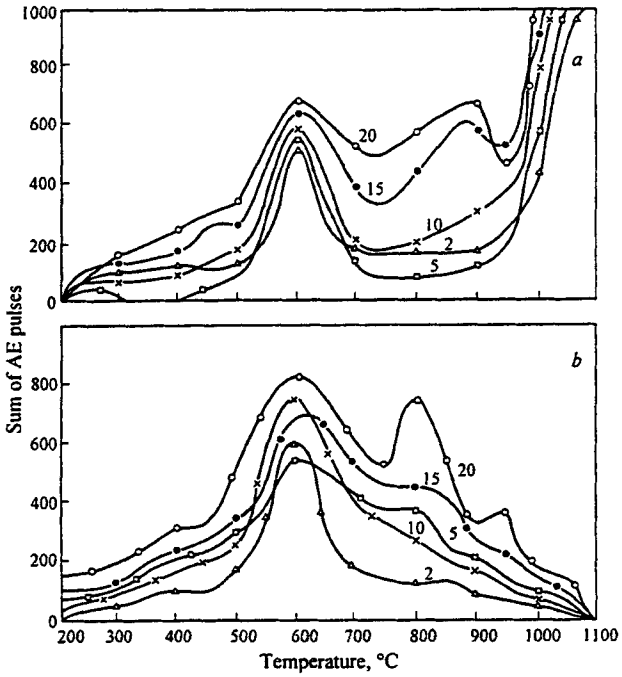

Fig. 5. Variation of the sum of the AE pulses as a function of the firing (*a*) and cooling (*b*) regimes of the monolithic lining; the numbers at the curves indicate the heating (cooling) rates, °C/min.

Based on the obtained acoustic emission (AE) signals we plotted the average variation of the sum of the pulses for different heating and cooling regimes (Fig. 5).

The maxima of the curves indicate that at the corresponding temperatures a specimen undergoes processes having the highest intensity of destructive phenomena. It can be seen that as the heating and cooling rates of the monolithic lining are increased, destructive phenomena intensify.

In order to describe the physical essensc of the processes the detected destructive phenomena were compared with results of x-ray phase analysis, data of differential thermal analysis (DTA), and data of high-temperature optical microscopy. In the monolithic cellular ceramic fiber lining, signals with the highest intensity were detected at 200 – 300, 500 – 700, and 950 – 1100°C in heating and at 500 – 850°C in cooling.

Comparison of the results of the DTA and the x-ray phase analysis allows us to infer that at 200 – 300°C the physically bound moisture is removed. The maximum of destructive phenomena at 500 – 700°C can be associated with removal of chemically bound water and the appearance of cristobalite. The fall in the AE signals at 950°C is caused by the appearance of a liquid phase, which damps down the acoustic emission. This stage is characterized physically by shrinkage. With a further increase in the temperature the material is sintered, which is reflected by the maximum on the curves.

In cooling of the lining in the interval 850 – 750°C clay transforms from a pyroplastic state to a solid state with the

appearance of stresses reduced by local disruptions (cracks). The dangerous region at 500 – 650°C is explained by the "quartz effect" (the transformation of α-quartz into β-quartz).

In order to choose the optimum rate of the first firing and cooling of the monolithic lining we investigated the maximum values of Y_3 and Y_4 by the regression equations. The computation was based on the method of direct exhaustion.

Since we did not observe active destructive changes in the intervals 20 – 500°C and 700 – 900°C in heating and 1100 – 850°C and 500 – 20°C in cooling, we assumed that an increase in the heating and cooling rates in these ranges does not affect the properties of the lining.

Analysis of the AE data allowed us to choose optimum regimes for the first firing and cooling of the monolithic cellular ceramic fiber lining. For the first firing of the lining the recommended regime is as follows:

Temperature range, °C	0 – 500	500 – 700	700 – 900	900 – 1100
Heating rate, °C/min	10	5	10	5

For cooling of the monolithic lining we recommend

Temperature range, °C	1100 – 850	850 – 500	500 – 20
Cooling rate, °C/min	10	5	10

The effect of the scale factor (the lining diameter) is explained by the appearance of thermal stresses in shrinkage. These stresses can be eliminated by envisaging temperature joints.

In the first firing and cooling of the monolithic lining by the recommended regime we did not observe destructive processes. Repeated heating and cooling with the established maximum admissible rates did not affect the physicomechanical properties of the monolithic lining. This allowed us to conclude that further use of the monolithic lining can occur in the operating regime of the furnace. The main properties of the material are presented in Table 5.

TABLE 5. Physicotechnical Properties of the Cellular Ceramic Fiber Material

Parameters	Cellular ceramic fiber material	ShLB-0.4 (prototype)
Mean density, kg/m^3	460 – 470	400
Ultimate strength, MPa:		
compressive	1.35 – 1.45	1.4
bending	0.8 – 0.9	–
Shrinkage, %:		
air	0	15
firing	0.4 – 0.6	11.2
Thermal conductivity, W/(m · K), at an average temperature of, K:		
473	0.12	0.16
1073	0.210	0.280
Heat resistance (1300°C – air), heat cycles	> 75	< 10
Temperature coefficient of linear expansion, 10^{-6} K^{-1}	2.3	5.7

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