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Experiment Planning for Prospective Use of Barium-Containing Alumina Cement for Refractory Concrete Making

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Abstract. The prospect of further use of refractory concrete based on barium-containing alumina cement was justified, with the help of a simplex-lattice method of the experiment planning, the granulometric composition of concrete was selected and its physical, mechanical and technical properties were determined, which exceed the properties of the used analogues.

Introduction

The very limited reserves of natural fuel in Ukraine and the lack of cheap alternative energy sources make it impossible to abandon the operation of nuclear power plants in the near future. Currently, the relevant is creation of structures for biological protection of nuclear power plants and various nuclear installations using new types of refractory concrete, which have high strength, refractory resistance, and resistance to aggressive factors: ionizing radiation, high temperature conditions, corrosive media, etc.

Complex solution of the problems of increasing of durability of various materials for construction of nuclear plants and research reactors, as well as reducing of labor costs for their construction and repair, is provided by refractory and heat-resistant cements and concrete based on them, having high thermomechanical properties [1]. For thermal protection of nuclear power facilities, special-purpose concrete based on alumina cement is increasingly used. However, the experience of creating of sarcophagus over the emergency unit of the Chernobyl nuclear power plant showed that under conditions of elevated temperatures, such materials lose up to 30% of their initial strength, which is associated with the removal of water from calcium hydro aluminates and can lead to formation of cracks and deformation of structures. Therefore, development of new types of materials that would withstand the effects of radiation heating is an actual issue.

Materials and Methods

Cement based on $CaO – BaO – Al_2O_3$ optimal system composition [2], cement : aggregate ratio – 1 : 3 was used for making concretes. Electro fused corundum of specified fractional composition was used as aggregate. Concrete samples of size $(2 \times 2 \times 2) \cdot 10^{-2}$ м (V/T = 0.08) were made by vibration compaction.

To determine the optimal grain size ratio of adjacent aggregate fractions, formula 1 was used [3]:

$$d_n / d_{n-1} = 0,226 \quad (1)$$

where d_n – the average grain diameter of the fine fraction;
d_{n-1} – the average grain diameter of the coarse fraction.

Optimization of quantitative ratio of adjacent aggregate fractions was carried out using simplex-lattice method of experiment planning [4-5].

To determine the dependence of concrete properties on the quantitative ratio of corundum aggregate fractions, a simplex lattice planning method with a polynomial of incomplete third order was used:

$$У = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 + \beta_{123} x_1 x_2 x_3 \quad (2)$$

Based on the results of experimental data, polynomial coefficients expressing the dependence of strength on the quantitative and granulometric ratio of aggregate fractions were calculated. The regression equation is the following:

$$У = 40\ x_1 + 43\ x_2 + 46\ x_3 + 10\ x_1 x_2 + 8\ x_1 x_3 + 62\ x_2 x_3 + 84\ x_1 x_2 x_3 \quad (3)$$

The regression equation was calculated with a variation step of 10%. Based on the results of calculations and mathematical processing of the experiment, a simplex diagram "composition: property" (Fig. 1) and projections of lines of equal level for concrete strength were built.

The experiment-planning matrix is shown in Table 1.

Table 1. Experiment-planning matrix

Polynomial factor	Designation and physical meaning of factors			
	Aggregate fractions			Compressive strength to 28 days, [MPa]
	$BaAl_2O_4$ X_1	$CaAl_2O_4$ X_2	$BaCa_2Al_8O_{15}$ X_3	
η_1	1	0	0	40
η_2	0	1	0	43
η_3	0	0	1	46
η_{12}	0.5	0.5	0	60
η_{13}	0.5	0	0.5	45
η_{23}	0	0,5	0,5	44
η_{123}	0.33	0.33	0.33	55

Concrete samples $(2 \times 2 \times 2) \cdot 10^{-2}$ m were prepared to determine the heat resistance and tested after 28 daily hardening at 850 ^{0}C и 1300 ^{0}C followed by air cooling. Because of tests, it was found that concrete samples withstood more than 20 thermal cycles, while maintaining more than 80% of the initial strength.

One of the important performance characteristics of refractory concrete is the degree of its softening during heating. For the study of the mechanical strength of concrete under the influence of elevated temperatures, samples were made with a size of $(2 \times 2 \times 2) \cdot 10^{-2}$ m, which were tested after 28 days of hardening. Isothermal exposure – 2 hours at the specified temperature (temperature interval – 200-1300 ^{0}C), the results of concrete tests under the influence of increased temperatures are given in Table 2 and Figure 2.

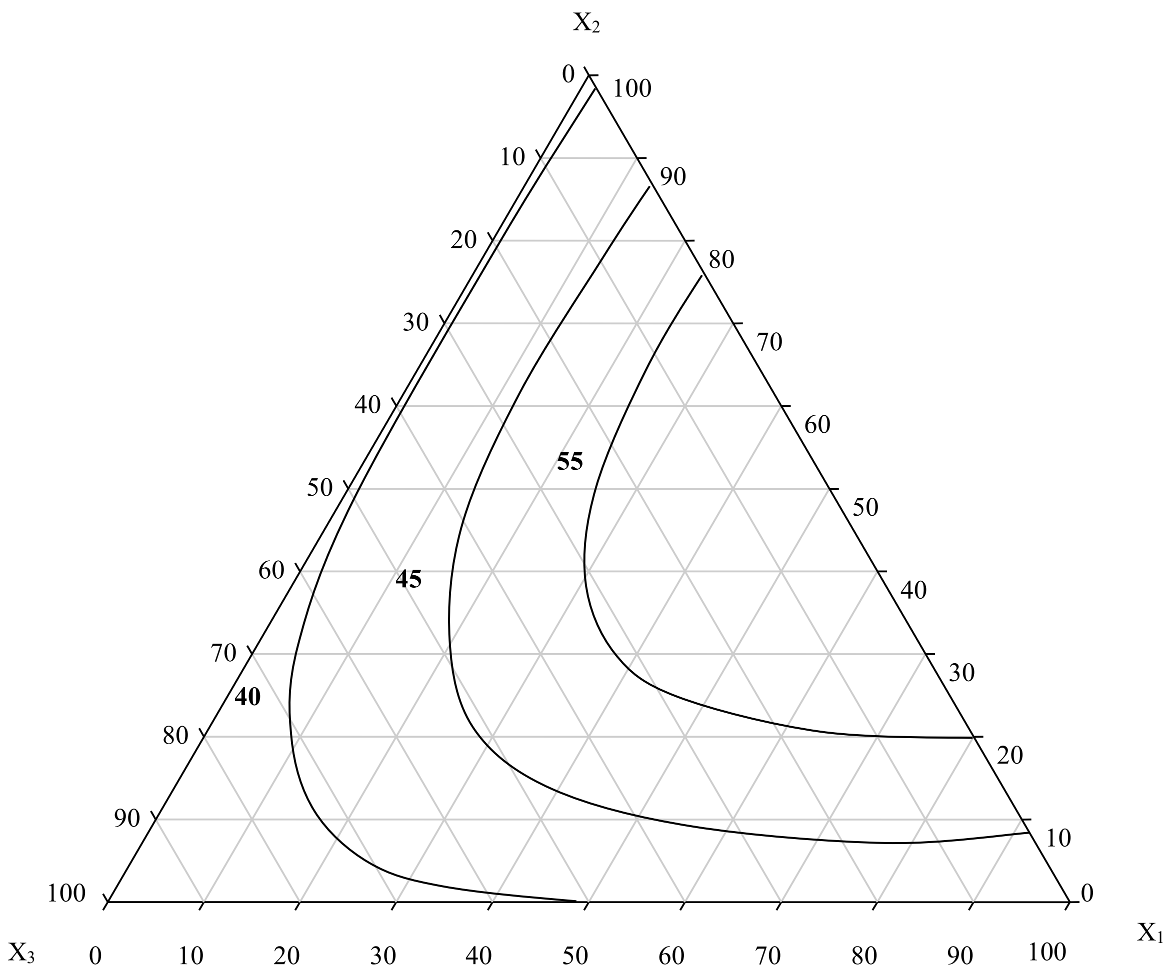

Fig 1. Barium-containing concrete strength diagram based on refractory cement, taking into account particle size distribution

As can be seen from the results presented, the greatest softening of concrete is observed in the temperature range from 600 to 1000 ^{0}C. Above 1000 ^{0}C, sintering of the material begins to obtain a strong ceramic structure, as evidenced by increase in strength.

Concrete tests were carried out for refractory resistance and strain under load. The refractory strength of concrete was over 1780 ^{0}C. At this temperature, there is no change in the surface of the pyroscope and there is no liquid phase, which indicates that the refractory strength of the sample is close to 2000 ^{0}C.

Table 2. Effect of elevated temperatures on concrete strength characteristics

Heat treatment temperature, [^{0} C]	Concrete on cement BCA -1 composition		Concrete on cement BCA - 2 composition	
	Compressive strength, [MPa]	Degree of softening,[%]	Compressive strength, [MPa]	Degree of softening,[%]
20	60.0	-	56	-
200	58.2	-3.0	55.2	-1.43
400	56.1	-6.5	52.1	-6.96
600	54.5	-9.17	48.3	-13.75
800	52.3	-12.83	47.4	-15.36
1000	51.8	-13.67	46.8	-16.43
1300	53.2	-11.33	47.9	-14.46

During testing of concrete for deformation under load, it was established that the temperature of the beginning of destruction of the concrete sample on cement of BCA - 1 composition is 1370 ^{0}C (the temperature corresponds to a decrease in the height of the sample by 1 mm, which corresponds to 2%). The temperature of start of concrete sample destruction on cement of BCA - 2 composition is 1310 ^{0}C (temperature corresponds to decrease of sample height by 0.3 mm, which corresponds to 0.6%).

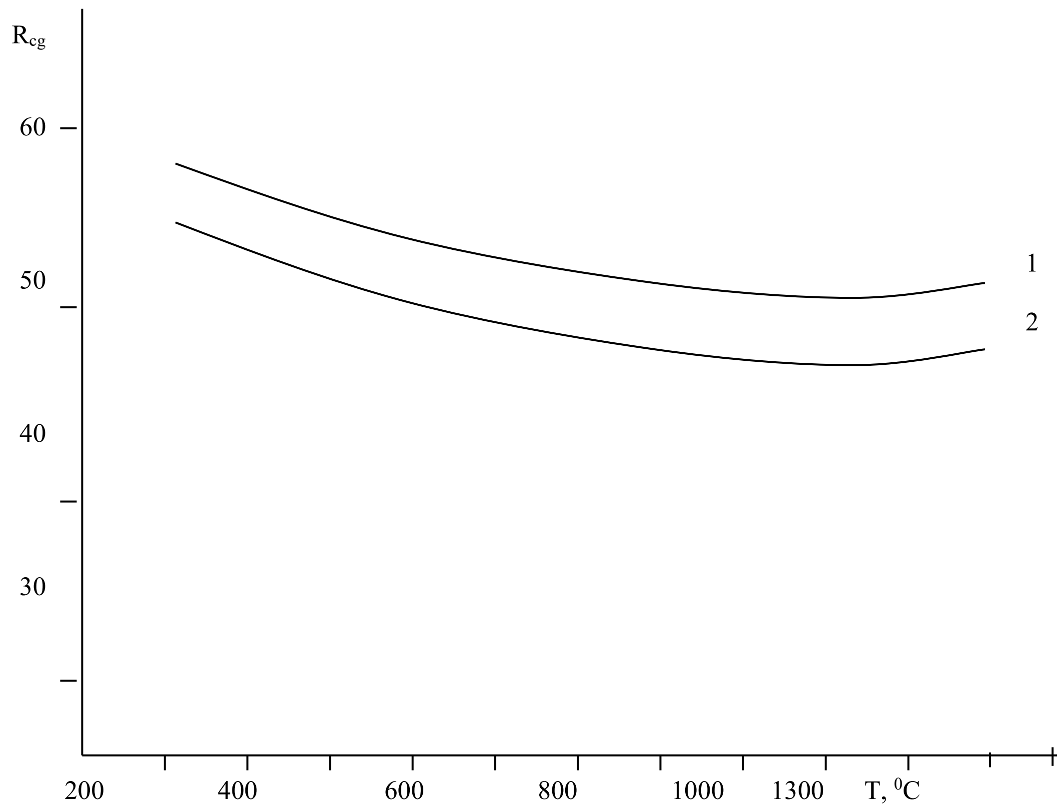

Fig. 2. Dependence of concrete softening degree on temperature:
1 – concrete on cement BCA - 1 composition,
2 – concrete on cement BCA - 2 composition

Table 3 shows the physical, mechanical and technical properties of concrete on cement of two optimal compositions.

The petrographic studies of special concrete based on barium-containing alumina cement of BCA-2 composition and electro fused corundum were carried out.

Because of the carried out petrographic studies, it was found that the structure of the material is uneven (Figure 3). Larger angular areas (grains) of isothermal and irregular shape with a size of 0.1 – 0.65 mm, maximum of 1 mm (~75%) and a more fine-grained binder mass are distinguished. Aggregate areas (grains) are fused corundum, colorless, with cleavage cracks.

Corundum of fine fraction with size of 20-100 mcm, aggregates with size of 20-40 mcm, maximum of 100 mcm, consisting of anisotropic crystals with size of 4-12 mcm, maximum of 20 mcm, preferably $BaAl_2O_4$, are distinguished in binding mass.

Corundum grains and "residues" of cement phases are cemented with colorless (~ 10-15%) isotropic films with N_{cp} ~ 1.540 ± 0.005, in some areas with N_{cp} ~ 1.520 ± 0.005, which corresponds in the first case to hydrates of type $BaAl_2O_4 \cdot 6H_2O$, in the second case – to $Ca_2Al_2O_5 \cdot 8H_2O$. In unit sections N_{cp} raised to 1.600-1.605, possibly due to the presence of $Ca_3Al_2O_6 \cdot 6H_2O$ or $Al(OH)_3$.

The contacts of aggregate grains (corundum) with the binder mass are in most cases dense.

Table 3. Physical, mechanical, and technical properties of concrete

Indicators	Concrete on cement BCA - 1 composition	Concrete on cement BCA - 2 composition
Compression strength at the age of 28 days, [MPa]	60	56
Heat resistance, quantity of heat shifts, 850 ^{0}C – air 1300 ^{0}C – air	› 20 › 20	
Degree of softening in temperature range 20 – 1300 ^{0}C, [%]	3.00 – 13.67	1.43 – 16.43
Fire resistance, [^{0}C]	1780 - 2000	
Strain start temperature under load, ^{0}C: - 2 % - 0,6 %	1370	1310

There are rare cracks on the contact of corundum grains with the ligament and in the ligament 7-23 micrometers wide, maximum – 40 micrometers. Pores are rare rounded in size 0.1-1.3 micrometers, maximum – 2.0 micrometers. Figure 3 shows large areas (fragments) of grains of fused corundum and binding mass.

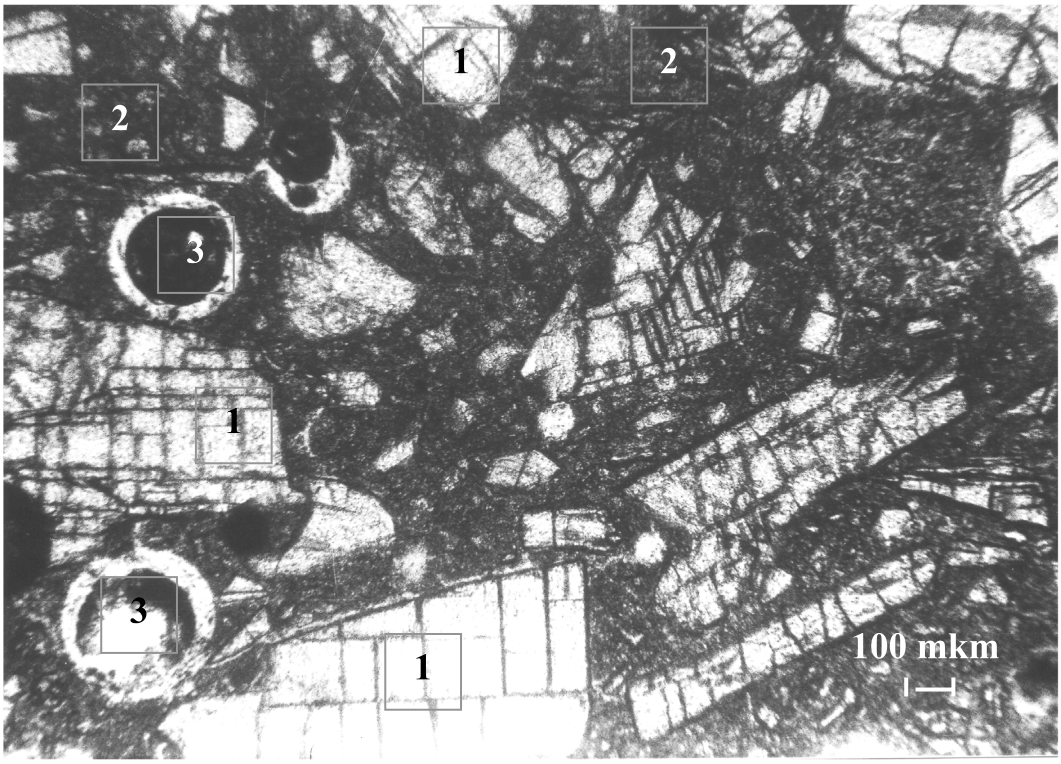

Fig. 3. Total concrete microstructure (increase x 28,8):
1 – fused corundum with characteristic solidity;
2 – binder mass (cement residues + hydrate forms); 3 – pores

Conclusions

To obtain protective concretes, it is proposed to use electro-fused corundum as aggregate, which has matrix affinity for cement. Concrete with two optimal compositions of cements was tested, granulometric composition of aggregate was selected, polynomial coefficients were calculated and mathematical equation expressing dependence of strength on quantitative and granulometric ratio of aggregate fractions was obtained. It was established that the obtained concretes are characterized by high compression strength after 28 days of hardening of 40-60 MPa depending on the aggregate fractions, heat resistance of more than 20 heat shifts, while maintaining more than 80% of the initial strength, have a low degree of softening when heated to 16.5%. The temperature of the beginning of deformation of the concrete sample on cement of BCA - 1 composition is 1370 ^{0}C (the temperature corresponds to a decrease in the height of the sample by 1 mm, which corresponds to 2%). The temperature of the beginning of deformation of the concrete sample on cement of BCA - 2 composition is 1310 ^{0}C (the temperature corresponds to a decrease in the height of the sample by 0.3 mm, which corresponds to 0.6%).

The results of petrographic studies found that the structure of refractory concrete is dense, uneven, the nature of porosity is mainly closed, larger angular areas (grains) of isothermal and irregular shape with a size of 0.1-0.65 mm, maximum of 1 mm (~ 75%) and a finer-grained binder mass are distinguished.

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