UDC 666-123 DOI: 10.31650/2786-6696-2024-8-50-64

OPTIMIZATION OF ACCELERATED CARBONIZATION HARDENING EFFECTS OF EXPANDED CLAY CONCRETE

[1]**Gara O.A.,** Ph.D., Professor,
gara@odaba.edu.ua, ORCID: 0000-0002-2413-1860
[1]**Kolesnykov A.V.**, Ph.D., Associate Professor,
kolesnikov_himek@odaba.edu.ua, ORCID: 0000-0001-8737-0933
[1]**Semenova S.V.**, Ph.D., Associate Professor,
semenova@odaba.edu.ua, ORCID: 0000-0002-5309-5854
[1]**Oliinyk T.P.**, Ph.D., Associate Professor,
oleynik_himek@odaba.edu.ua, ORCID: 0000-0002-4469-6797
[1]*Odessa State Academy of Civil Engineering and Architecture*
4, Didrikhson str., Odesa, 65029, Ukraine

Abstract. The paper examines the data obtained during the study of accelerated carbonization hardening of expanded clay concrete. The mechanisms and methods of carbonization hardening, the influence of recipe and technological factors on strength characteristics are considered. The most significant of them were the consumption of the binder, the relative amount and granulometry of the filler (ground limestone) and aggregate (keramzite), the concentration of the plasticizing additive (SYM) and the technological parameters of carbonization – the maximum pressure and time of carbonization. The nature of the influence of the relevant properties on the strength characteristics is considered. The results of the planned and implemented experiment were studied using the methodology of response surfaces. Experimental-statistical models of compressive strength were built for 1 hour, 28 days and 180 days from the end of the carbonation procedure. The degrees of influence of various factors are determined. Constructed dependencies were displayed using a series of diagrams of isolines of strength at the limit values of three factors of weak influence. The proposed hypothesis is that in experimental-statistical models of the properties of materials closely related to their structure, a tendency to simplification is created when the corresponding materials pass through the stage of structure formation. Insignificant components of the experimental-statistical model, mainly related to the interaction of factors determining the structure of the material, are reduced. Corresponding transformations arise, in particular, for constructed strength models. Two tasks of optimizing the long-term strength of expanded clay concrete are formulated. The first task is related to the elimination of hard additional constraints during optimization, while the long-term strength reaches its maximum. The second engineering-based task contains additional requirements for the economical use of binder and the convenience of the carbonization process. The distributions of the parameters of the desirability function were determined for both tasks. As a result of the optimization, two main sets of recipe-technological factors and corresponding properties of composite materials were obtained.

Keywords: accelerated carbonization hardening, experimental planning, optimality according to the combined criterion, desirability function.

Introduction. Accelerated carbonation hardening (ACH) is a promising solution to the problems of increasing the durability and strength of concrete structures. Traditional methods of curing concrete, such as steam curing or the use of chemical additives often require long curing periods and may have environmental or economic disadvantages. Thus, more than 10 million tons of conventional fuel, including more than 3 billion cubic meters of gas, is spent annually on heat treatment in the European Union. The coefficient of use of heat and fuel due to the imperfection of the technological process is 5...11%. In contrast, accelerated carbonisation offers a sustainable and effective approach to improving the properties of concrete. Its application has a significant ecological component related to the fixation of atmospheric carbon dioxide CO_2. ACH technology is associated with the injection of CO_2, which reacts with non-hydrated components of the cement composite [1], which leads to the transformation of absorbed CO_2 into various forms of stable carbonates [2]. This method is better than natural or weathered carbonation, because the natural diffusion of atmospheric CO_2 through the pores of concrete or mortar occurs very slowly [3]. ACH technology accelerates the rate of carbonization and increases the net amount of sequestered CO_2 [4]. In addition, ACH process conditions can be adjusted in a controlled environment (usually in a carbonization chamber) [5]. Thus, the main task of promoting ACH concrete as a carbon dioxide sequestration strategy is to ensure conditions under which carbonation does not impair mechanical strength, as well as other operational properties. The explanation of the environmental and economic efficiency of ACH is clearly visible in the following examples.

Consider the construction of a reinforced concrete bridge in harsh environmental conditions. ACH can significantly reduce the time required for the concrete to reach design strength, which allows shortening the construction period and minimizing traffic disruptions. Moreover, the increased durability provided by carbonation can extend the life of the bridge, reducing maintenance costs and ensuring long-term structural integrity.

In urban areas where space and resources are limited, accelerated carbonation techniques can be particularly useful. For example, in the construction of high-rise buildings, the use of ACH can accelerate the hardening process, which will allow developers to meet a tight project deadline and optimize the use of resources. In addition, the increased durability of carbonized concrete can increase the resistance of high-rise structures to the effects of environmental factors and seismic phenomena, ensuring the safety of passengers and reducing the risk of expensive repairs or upgrades in the future. Accelerated carbonation also provides environmental benefits due to the binding of carbon dioxide within the concrete matrix. This ability to capture and store carbon dioxide contributes to the reduction of greenhouse gas emissions, which meets the goals of sustainable development and regulatory requirements in the construction industry.

Thus, the implementation of ACH concrete serves several purposes, including reducing construction time, increasing structural strength, optimizing resource use, and promoting environmental sustainability. ACH optimization methods allow to achieve the corresponding goals in the most effective way.

Analysis of recent research and publications. Existing studies have shown that accelerated carbonization can significantly improve the mechanical properties of concrete, including compressive strength, flexural strength, and abrasion resistance [6]. Research results indicate that carbonized concrete exhibits higher compressive strength compared to conventionally cured concrete, with improvements ranging from 10% to 50% depending on carbonation conditions and concrete mix composition. It was also shown that ACH increases the durability of concrete by reducing permeability and increasing resistance to the penetration of chlorides, the action of sulfates, and the alkali-silica reaction [7]. In addition, the improvement of the long-term characteristics of carbonized concrete structures, the reduction of maintenance requirements and the increase of service life observed in the field [8] are shown.

ACH of cement composites contains complex mechanisms that occur both at the macroscopic and microscopic levels inside the concrete matrix [9]. Understanding these mechanisms is necessary to optimize the efficiency and effectiveness of accelerated carbonation methods. The mechanisms causing ACH have several key processes:

1. Diffusion of carbon dioxide (CO_2). The process of accelerated carbonation begins with the diffusion of CO_2 into the structure of the porous concrete matrix. The presence of moisture inside the pores facilitates the transport of CO_2, allowing it to penetrate deeper into the concrete [10].

2. Chemical reactions. Carbon dioxide upon contact with calcium hydroxide ($Ca(OH)_2$) present in hydrated cement paste undergoes a number of chemical reactions. The primary reaction [11] includes the formation of calcium carbonate ($CaCO_3$) and water H_2O:

$$Ca\,(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$

In the course of this reaction, calcium hydroxide is consumed and calcium carbonate is formed, which leads to the deposition of calcite crystals in the pores of concrete.

3. Propagation of the carbonization front. As carbonation progresses, a carbonation front forms inside the concrete matrix, delineating the boundary between carbonized and non-carbonated areas. The advancement of this front is influenced by factors such as CO_2 concentration, moisture content, temperature, and pore structure [12].

4. Formation of calcium carbonate crystals ($CaCO_3$). Precipitation of calcium carbonate crystals occurs in the pores of the concrete matrix. These crystals contribute to compaction of the microstructure by filling voids and internodal spaces. The re-formed calcium carbonate acts as a binder, binding the cement particles and aggregates together, thereby increasing the overall strength and durability of the concrete.

5. Formation of calcium silicate hydrate (CSH). In addition to calcium carbonate, accelerated carbonation also contributes to the formation of a calcium silicate hydrate (CSH) gel. This gel serves as the main binding phase in cement hydrated dough and is responsible for the mechanical properties of concrete [13]. The formation of CSH caused by carbonation contributes to the compaction and strengthening of the concrete matrix.

6. Thinning of pores and microstructural changes. The deposition of calcium carbonate and the formation of CSH gel lead to the crushing of the porous structure within the concrete matrix. Such grinding of pores reduces the permeability of concrete, limiting the ingress of harmful substances such as chlorides and sulfates. In addition, microstructural changes caused by accelerated carbonization contribute to increased resistance to chemical action and environmental degradation.

7. Decrease in alkalinity and decrease in pH. As carbonation progresses, the alkalinity of the concrete matrix decreases due to the consumption of calcium hydroxide. Such a decrease in pH can have the effect of increasing the susceptibility to corrosion of collateral steel reinforcement [14]. However, the formation of a protective layer of calcium carbonate on the surface of the reinforcement can reduce the risk of corrosion of carbonized concrete.

8. Long-term effects and aging. The effectiveness of accelerated carbonation in the long term depends on factors such as exposure conditions, concrete composition and maintenance methods [15]. Monitoring the performance of carbonized concrete structures over time is necessary to assess their durability and ensure structural integrity.

Thus, the mechanisms of accelerated carbonation of cement composites contain a number of interconnected processes, including CO_2 diffusion, chemical reactions, pore crushing, microstructural changes, and alkalinity changes. Together, these mechanisms contribute to increasing the strength, durability and stability of concrete, which makes accelerated carbonation a promising method of improving the characteristics of binding materials in various construction applications [16].

The purpose of the work is an analysis of previously obtained experimental data on the effect of binder consumption, plasticizing additive content, limestone content, carbonation pressure (CO_2), carbonation process time on compressive and tensile strength at the age of 1 day, 28 days and 180 days, as well as the construction of ES-models, their conceptual interpretation and optimization by the methods of the desirability function, taking into account the technologically justified distribution of degrees of importance and the technological conditions of conducting ACH.

Research methodology consists in the application to the previously obtained experimental data of the planning of the experiment [17], in particular, the methodology of ES-modeling and response surfaces, as well as the apparatus of the desirability function [18], intended for the quantitative optimization of recipe and technological factors. All applied methods were implemented using the Design Expert software package [19].

Research results. Accelerated carbonation hardening (ACH) of concrete-based composites is a process that stimulates an accelerated reaction between carbon dioxide (CO_2) and hydrated cement in concrete, resulting in the formation of calcium carbonates and strengthening of the concrete matrix. From a technological point of view, the ACH technique can be divided into the following stages:

1. *Surface preparation*. Before starting the process of accelerated carbonation, the surface of the concrete must be cleaned of impurities and processed to ensure uniform penetration of CO_2.

2. *Installation of a device for accelerated carbonation*. For the process of accelerated carbonation, special equipment is used, such as a chamber with controlled CO_2 content or a CO_2 injection system.

3. *Regulation of environmental conditions*. The temperature and humidity of the environment can significantly affect the rate of the carbonation reaction. Therefore, it is important to maintain

optimal conditions to ensure effective penetration of CO_2 into the concrete matrix.

4. *CO_2 injection*. CO_2 is injected into the middle of the concrete structure using special equipment. This can happen through holes located on the surface of the concrete, or with the use of special channels and pipelines laid inside the concrete structure.

5. *Process control and monitoring*. When performing accelerated carbonation, it is important to control and adjust process parameters such as CO_2 content, temperature, humidity, and exposure time. This helps to ensure uniform and effective penetration of CO_2 throughout the thickness of the concrete and achieve the desired hardening characteristics.

6. *Evaluation of results*. After the accelerated carbonization process is completed, the results are evaluated, which includes an analysis of the mechanical properties of concrete, its structure, and the degree of carbonization.

In order to ensure high de-decking strength of concrete within 30...60 minutes, it is necessary to use carbonation regimes characterized by excessive pressure. According to the theory of heat and mass transfer, a significant acceleration of the carbonization process can be achieved by intensively supplying a gaseous reagent to the reaction zone. From this point of view, preliminary vacuuming of freshly formed concrete allows to create a porous capillary system that is under rarefaction. After that, pressure drops in the initial period of hardening caused by the removal of vacuum and the creation of excess pressure during the supply of carbon dioxide cause stress relaxation in the capillary structure of concrete, which leads to effective self-absorption and intensification of the carbonation process. The process of "carbonate" dissolution of the original minerals is accelerated in proportion to the pressure of carbon dioxide. At the same time, the use of modes with a high pressure value allows you to control the processes of structure formation of cement compositions.

Based on the results of previous studies and literature data [20], expanded clay concrete with an optimal aggregate structure and consumption of binder 300 kg/m^3 of concrete was chosen as the basic composition. The properties of the concrete mixture, as well as the structure and properties of hardened concrete depend most on the relative content of fine fractions in the aggregate (up to 5 mm). It was established that by changing the relative content of small fractions, it is possible to vary all the main properties of lightweight concrete within certain limits – strength, density, modulus of elasticity, thermal conductivity, etc.

The structure of carbonized concrete and its strength characteristics should be considered in relation to three main factors:

- aggregate structure, which depends on the location of the porous aggregate in the concrete structure;
- the concentration of the binder, which depends on the type and on the regularity of the properties of cement and its additives;
- consumption of mixing water, taking into account the features of the carbonization technology.

In connection with the influence on the structure formation process of a complex of recipe-technological and physico-chemical factors, the construction and technical properties of concrete can vary widely. Under conditions of carbonization, as a result of the simultaneous action of a number of structure-forming and destructive factors, the hardening process can develop with different speed and completeness of the reactions. In order to control the technological process, which allows to ensure optimal conditions of carbonization hardening, it is necessary to identify and study the degree and nature of the influence of the main recipe and technological parameters on the initial strength of after expanded clay concrete carbonization and for the strength during the period of operation of the products. In this case, the criterion for optimizing the recipe and technological conditions for the preparation of carbonized-hardening expanded clay concrete can be taken to be the minimum consumption of cement, which ensures the specified properties of the material, as the most expensive and scarce component.

The factors that largely determine the regulated physical and mechanical properties of expanded clay concrete carbonization hardening were adopted as variable recipe-technological factors, namely: binder consumption, sulfite-yeast mash (SYM) additive content, percentage content of ground limestone in the binder, as well as carbonization parameters.

The binder consumption was varied in the range from 300 to 500 kg/m^3. Such a narrow interval of variation in the consumption of the binder is due to the fact that the percentage content of ground limestone in the binder, which was changed in the interval from 0 to 30%, was taken as one of the factors. In this regard, the actual consumption of cement in the experiment varied from 200 to 500 kg/m^3, which covers almost the entire strength range of light concrete within the limits of permissible norms of cement consumption.

The introduction of crushed limestone into the binding composition is due to the need to reduce the consumption of cement, in addition, it should lead to the intensification of the carbonation process and cause structural changes that improve the mechanical characteristics of the system.

The introduction of the SYM plasticizing additive into the concrete mixture was considered as a technological technique that ensures the reduction of the total water content of the concrete mixture at the stage of its preparation, which is one of the ways of intensifying the carbonation process. The content of the SYM additive was varied in the experiment in the range from 0 to 0.4% of the weight of the binder.

Carbonation of freshly formed concrete samples was carried out in the mode of gradually increasing pressure, the effectiveness of which was shown in [20]. At the same time, the amount of carbon dioxide pressure at the main stage of the regime was varied in the range from 0.6 to 1.2 MPa. The choice of the upper limit is dictated by the nominal value of pressure for which industrial autoclaves used in the construction industry are designed.

The duration of the processing regime was varied from 30 to 60 minutes, based on technological considerations. An increase in the carbonization time contradicts the formulated goal of research related to the development of an effective mobile technology for the production of wall products, subject to a sharp reduction in the duration of hardening.

In order to build mathematical models that reflect the influence of the specified factors on the strength of carbonized expanded clay concrete in the design terms of the tests, a five-factor plan of the second order of the "Hartley-5" type with six points in the center of the plan was implemented. The factors and their levels of variation are given in the Table 1.

Table 1 – Factors and their levels of variation

Factors		Unit measurement	Code	Levels of variation		
				-1	0	+ 1
A	Binder consumption	kg/m^3	Knitt	300	400	500
B	Contents of the SYM addditive	%	SYM	0	0.2	0.4
C	The content of ground limestone binder	%	GL	0	15	30
D	The amount of carbonation pressure	MPa	PCO_2	0.6	0.9	1.2
E	The duration of the carbonization process	min	tCO_2	30	45	60

In the experiment, expanded clay concrete of the optimal fractional composition, determined for the conditions of carbonization hardening, were used. The matrix of planning and consumption of expanded clay concrete mixture components is presented in [20].

The research was carried out on cube samples with the size of an edge 10 cm. Molded samples were subjected to carbonization according to the appropriate modes. When testing the samples, the density of concrete, the consumption of carbon dioxide for the carbonation of the samples (according to the change in the weight of the samples before and after carbonization), and the compressive strength of the samples during control test periods.

Study of compressive strength Rc (MPa) of the material after carbonation 1 hour (R1h), 28 days (R28) and 180 days (R180) led to the following results (Table 2).

Table 2 – Compressive strength of carbonized lightweight aggregate concrete

	Factors					Rc, MPa				Factors					Rc, MPa		
No	A	B	C	D	E	R1h	R28	R180	No	A	B	C	D	E	R1h	R28	R180
1	-1	-1	-1	-1	-1	3.3	7	8.3	17	-1	0	0	0	0	6.1	8.2	9.4
2	1	-1	-1	-1	1	10.5	18.6	22.5	18	1	0	0	0	0	13.5	20	23
3	-1	1	-1	-1	1	4.5	7.8	10.3	19	0	-1	0	0	0	7.8	11.2	12
4	1	1	-1	-1	-1	10.9	21.8	26.1	20	0	1	0	0	0	9.6	13.4	14.2
5	-1	-1	1	-1	1	2.1	4.6	6.5	21	0	0	-1	0	0	8.8	13.3	15
6	1	-1	1	-1	-1	8.5	15.4	18.7	22	0	0	1	0	0	7.8	10.9	12.4
7	-1	1	1	-1	-1	4.5	7	8.5	23	0	0	0	-1	0	7	12.7	14.2
8	1	1	1	-1	1	12.5	19	22.3	24	0	0	0	1	0	9.6	12.3	13.4
9	-1	-1	-1	1	1	7.3	8.2	8.9	25	0	0	0	0	-1	8	12.1	13.7
10	1	-1	-1	1	-1	12.5	18.2	22.3	26	0	0	0	0	1	9	12.3	13.9
11	-1	1	-1	1	-1	5.7	7	8.5	27	0	0	0	0	0	8.8	12.7	13.9
12	1	1	-1	1	1	16.5	20.6	23.5	28	0	0	0	0	0	8.4	12.2	14.2
13	-1	-1	1	1	-1	4.1	3.8	5.9	29	0	0	0	0	0	9.2	13.2	13.6
14	1	-1	1	1	1	11.7	14.6	18.1	30	0	0	0	0	0	9.1	13.1	13.4
15	-1	1	1	1	1	6.1	7	8.7	31	0	0	0	0	0	8.5	12.7	13.9
16	1	1	1	1	-1	13.7	18.2	20.9	32	0	0	0	0	0	8.8	12.3	14.4

The value of concrete strength in the experiment varies from 2.1 to 16.5 MPa 1 hour after carbonization, from 4.6 to 21.8 MPa at the age of 28 days and from 5.9 to 26.1 MPa at the age of 180 days. The obtained data made it possible to build ES-models of the properties of the composite material, for which the *Design Expert* package was used [19]. One of the useful features of this program is the ability to download "historical" data, that is, data obtained according to an experiment plan built and implemented previously outside of this package. The response surface methodology was used in data analysis. The type of obtained models, as well as some of their statistical characteristics, is shown in Table 3. The selection of statistically significant components was carried out at $\alpha = 0.1$ by splitting in the reverse order (Backward elimination).

All models are statistically significant. The predicted coefficients of determination (Predicted R^2) are consistent with the adjusted coefficient of determination (Adjecent R^2). The signal-to-noise ratio corresponding to Adequate Precision is much greater than the desired value of 4, which indicates the adequacy of the signal. High values of R^2 indicate the acceptability and high quality of the models. Let's consider and interpret each model separately.

*Compressive strength R1h (MPa) at the age of 1 hour after carbonization.*We will first assess the degree of influence of each factor on the analyzed strength characteristic at the central point of the plan using the perturbation diagram (Fig. 1).

A perturbation diagram allows you to compare the effects of all factors at a certain point in plan space and search for those factors that have the greatest effect on the response. The feedback shown in the diagram is constructed by varying only one factor over its range while holding the other factors fixed. A steep slope or curvature of the curve of the corresponding factor shows that the response is sensitive to changes in that factor. So, from Fig. 1, it can be seen that the consumption of cement (Knitt, A), the pressure of the main degree of carbonation (pCO_2, D), with a positive tangent of the angle of inclination in the center of the plan, and the content of limestone rock (GL, C) with a negative influence are most strongly influenced. Lines almost parallel to the base show insensitivity to changes in the corresponding factors, which is characteristic of factors B, E in Fig. 1, a.

Table 3 – ES-strength models of material characteristics and their statistical characteristics

R1h, MPa	R 28, MPa	R 180, MPa
R1h = +8.80 +3.70 * A +0.90 * B -0.50 * C +1.30 * D +0.50 * E +0.40 * A * B +0.20 * A * D +0.20 * A * E +0.40 * B * C -0.100 * B * D +0.10 * B * E -0.30 * C * D -0.30 * C * E +0.20 * D * E +0.98 * A2 -0.52 * C2 -0.52 * D2 -0.32 * E2	R28 = +12.69 +5.88 * A +1.12 * B -1.22 * C -0.22 * D +0.12 * E +0.47 * A * B -0.28 * A * C -0.18 * A * D +0.47 * B * C -0.13 * B * D +0.27 * D * E +1.36 * A2 -0.44 * B2 -0.64 * C2 -0.54 * E2	R180 = +13.87 +6.80 * A +1.10 * B -1.30 * C -0.40 * D +0.10 * E +0.30 * A * B -0.50 * A * C -0.20 * A * D -0.30 * A * E +0.30 * B * C +0.30 * B * D +2.13 * A2 -0.97 * B2
R-squared 0.9983 Adj R-squared 0.9960 Preq R-squared 0.9979 Adeq Precision 93.028	R-squared 0.9984 Adj R-squared 0.9967 Preq R-squared 0.9928 Adeq Precision 90.276	R-squared 0.9985 Adj R-squared 0.9973 Preq R-squared 0.9922 Adeq Precision 106.701

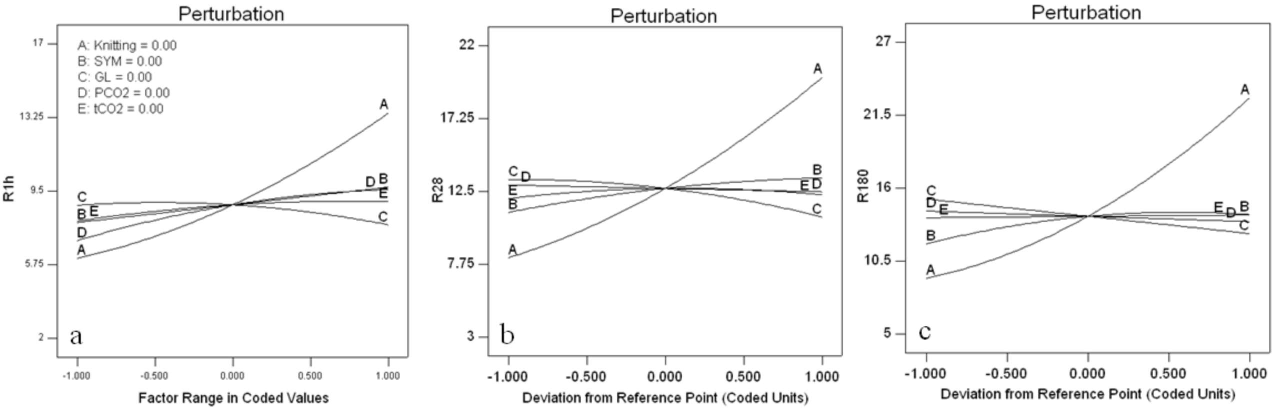
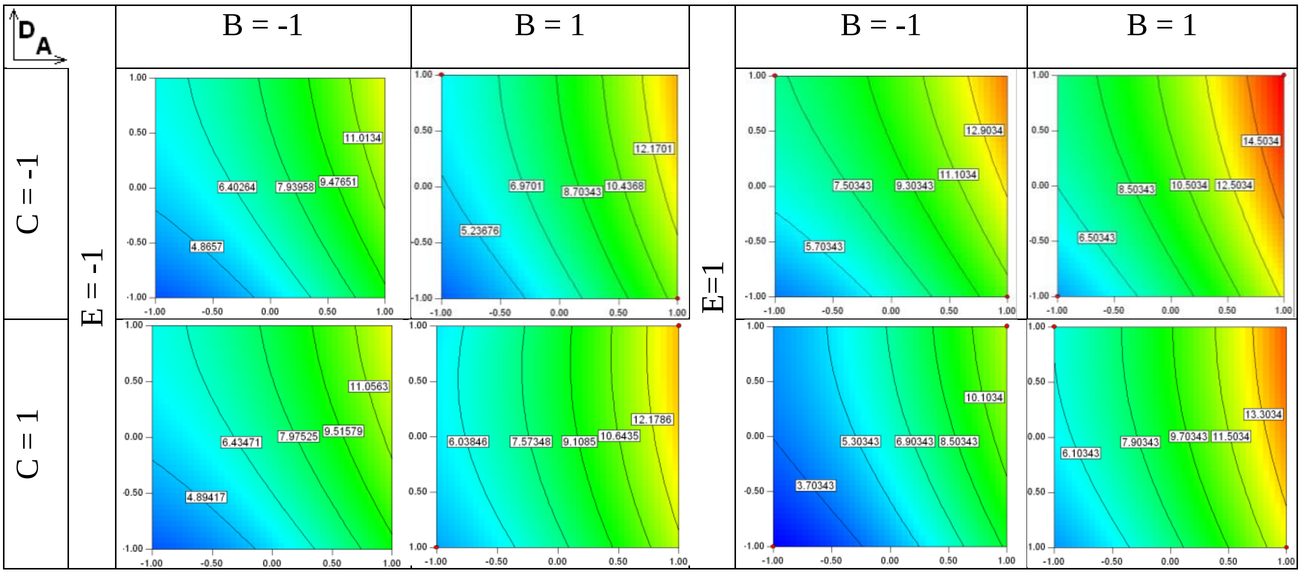


Fig. 1. Diagram of disturbances of strength properties at the central point of the plan:
a – for R1h; b – for R28; c – for R180

It is difficult to get a visual representation of the field of material properties directly due to the multidimensionality of the model under consideration. However, an idea of the geometric characteristics of the propertie's field can be obtained from a series of line diagrams of the level of the desired property (strength R1h, MPa), in the coordinates of the two main factors A and D at fixed values of factors B, C and E. Given the low sensitivity of the model for the latter properties B, C, E it is convenient to consider the map of isolines at the vertices of the unit cube of the factorial subspace BCE: a total of 8 sections shown in Table 4.

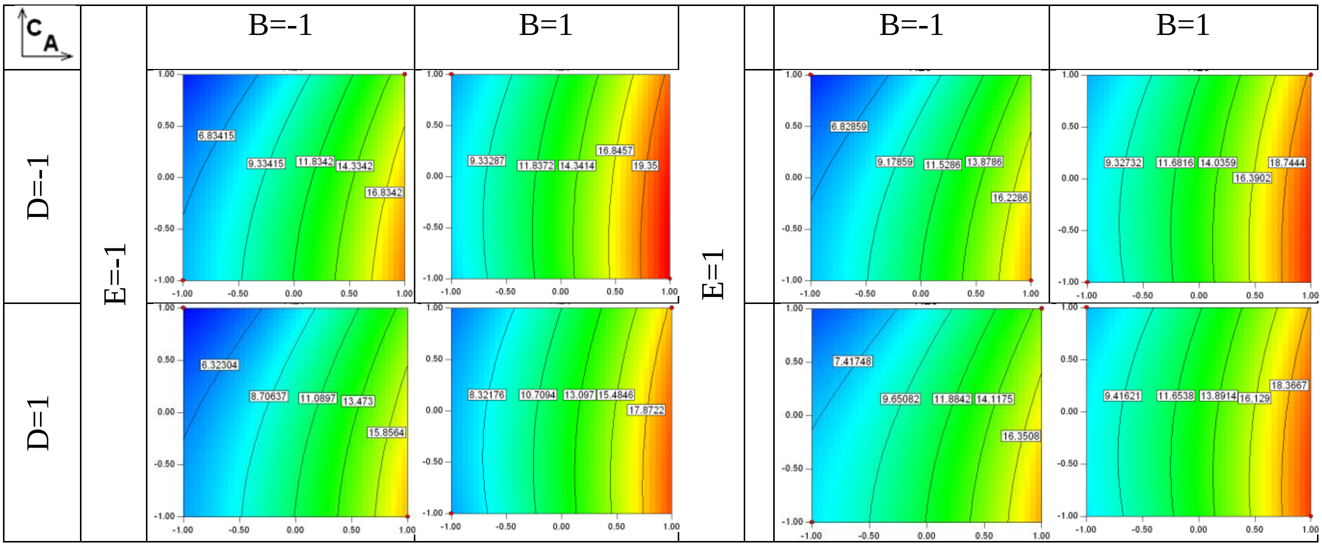
Table 4 – Graphic representation of the strength field of the material at the age of 1 hour from the end of carbonization Rh1, MPa



Since the strength values are actually consecutive elements of the same time series, a single systematic interpretation is appropriate for these data. It is possible only after considering all available results, in analytical and graphical form.

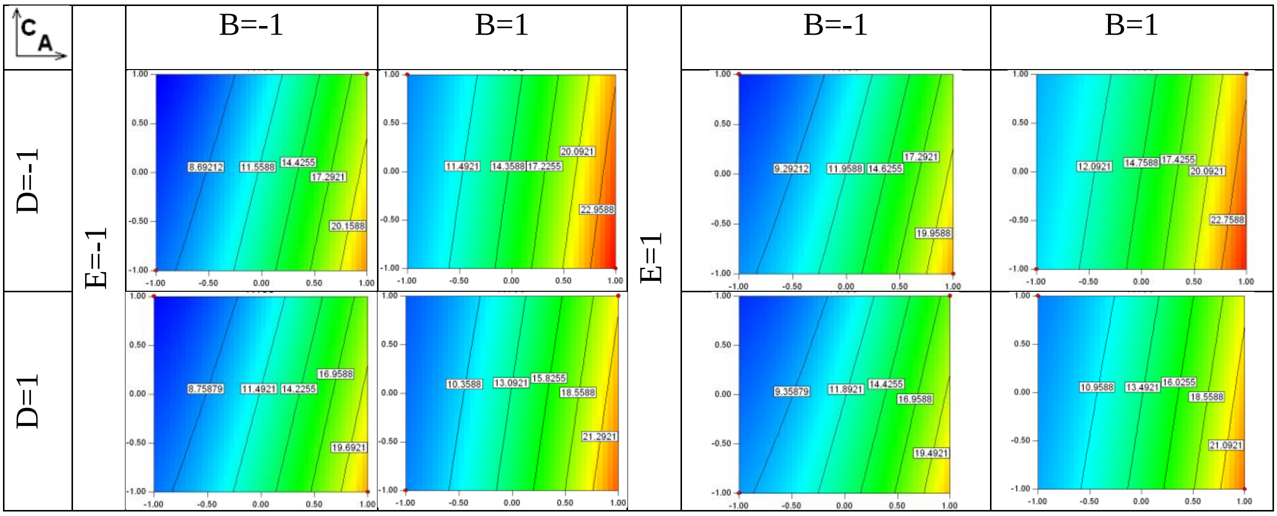
Compressive strength within 28 days after carbonization. The perturbation diagram for the considered model is shown in Fig. 1, b. From the diagram in Fig. 1, b shows that the main influencing factor is the consumption of cement (Knitt, A), with a positive tangent of the tangent angle. Next is the limestone content (GL, C) with a negative tangent of the tangent angle. Factors B, D and E are characterized by a smaller influence. Since the graphical presentation of the model is implemented using maps of isolines, as factors corresponding to the axes of the diagram, it is appropriate to choose A and C. Other factors with a relatively small effect on the strength of R28 are presented in discrete form. As for R1h, they correspond to the vertices of the unit cube of the factor subspace BDE. The appearance of the corresponding diagrams is shown in Table 5.

Table 5 – Graphic representation of the strength field of the material at the age of 28 days, after the end of R28 carbonation, MPa



*Compressive strength within 180 days after carbonization.*The perturbation diagram for the considered model is shown in Fig. 1, c. It can be seen from the diagram that the main influencing factor is again the consumption of cement (Knitt, A), with a positive tangent of the tangent angle. Other factors have a rather weak effect. The dependence for the SYM concentration factor (SYM, B) is characterized by a significant curvature, however, the content of limestones (GL, C) should again be distinguished from the tangent of the angle of inclination, which belongs to the center of the plan with a negative contribution value. Factors B, D and E are presented in discrete form, the axes of the diagrams – A and C. The construction scheme (Table 6) is similar to the previous one, shown in Table 6.

Table 6 – Graphic representation of the strength field of the material at the age of 180 days, after the end of carbonization R180, MPa



Interpretation of the role of factors in the formation of the strength of characteristics. The change in the degree of influence of the consumption of binder A by 3.2 MPa 1 hour after carbonization is a consequence of the change in the levels of factors B, D and E. At this age, the maximum increase in the strength of lightweight aggregate concrete is observed at the maximum content of the SYM additive (SYM, B) and the maximum values of parameters characterizing the carbonation regime. In contrast, at the age of 28 and 180 days, the maximum increase in strength occurs with the maximum content of SYM additive, with the use of a binder without ground limestone rock and with minimum values of pressure and carbonation time. The dependence of the strength of carbonized expanded clay concrete on the consumption of the binder is described by a function of the second order. With an increase in the consumption of the binder, the degree of influence of the factor increases. Thus, when the binder consumption is increased from 300 to 400 and from 400 to 500 kg/m^3, the strength of concrete increases on average by 2.7 and 4.7 MPa one hour after carbonization, by 4.2 and 7.2 MPa at the age of 28 days and at 4.5 and 9.5 MPa at the age of 180 days.

The second most influential prescription factor is the factor (GL, C) – percentage content in binding ground limestone. This factor is ambiguous in terms of its influence. At the age of one hour after carbonation, a change in the nature of exposure causes a change in the content of SYM and the amount of pressure and time of carbonation. Thus, in the absence of SYM additive and at the maximum values of pressure and carbonation time, an increase in the binder content of ground limestone leads to a decrease in strength by 3.0 MPa. With the content of SYM in the amount of 0.4% of the weight of the binder and at the minimum values of pressure and carbonation time, increasing the content of the binder of ground limestone rock from 0 to 30% leads to an increase in strength by 1.0 MPa.

With the increase in the age of concrete, the change in carbonation regimes has practically no effect on the degree and nature of the influence of the content of binding ground limestone on the strength of concrete. In this case, the consumption of the binder and the content of the SYM additive have a significant effect. Thus, with a binder consumption of 500 kg/m^3 and the absence of SYM additive in the concrete mixture, the change in the content of ground limestone in the studied interval leads to a decrease in strength by 4.0 MPa at the age of 28 days and by 4.1 MPa at aged 180 days With a binder consumption of 300 kg/m^3 and the content of the SYM additive in the amount of

0.4% of the weight of the binder, replacing up to 30% of cement with ground limestone has practically no effect on the strength of concrete at the age of 28 and 180 days after carbonization.

Since the values of strength characteristics studied by modeling are elements of a time series, it is of interest to study the transformation of strength models in the hardening process. It should be noted that all factors (AE) of the constructed regression ES-models affect the strength characteristics due to the material structure. When considering the models presented in Table 4, it is easy to determine the trend in the transformation of the dependences of the strength properties of the material on the studied factors affecting the structure. It basically boils down to the following – when moving from early hardening times to normative ones (28 days) and further up to 180 days, the models are significantly simplified. Thus, in the R180 model, many components are statistically excluded (exclusion is based on the p-value [19]) due to insignificance, partial simplification is evident for R28. Such exclusion reflects objective reality. In connection with these observations, the following hypothesis is of interest. The process of structure formation that occurs during hardening is, especially in the late stages, a process of a relaxation nature, and the amount of free energy associated with the structure of the material (structural potential [21]) approaches its minimum value, at least in some directions. Near the minimum of the structural potential, mainly linear interactions associated with factors directly related to the structure of the material remain significant. Thus, there is a tendency to simplify ES-models of a number of properties that are significantly dependent on the structure, for example, strength characteristics, mainly due to the elimination of interaction components.

The final formulation of the proposed hypothesis is as follows. In experimental-statistical models of structure-dependent properties (for example, strength), during the process of structure formation, there is a tendency to simplify due to the elimination of a number of components, mainly members that describe the interaction of factors that determine the structure of the material.

At the relatively late stages of hardening, stepwise splitting of the components can be observed – if any components of ES-models were split at the previous stages of structure formation, they do not occur in the following stages. This effect is manifested, in particular, for R28 and R180. The described display of the course of structure formation in the form of a stepwise transformation of ES-models corresponds to the concept of the material structure development process and its properties "from what has been achieved" [22].

The proposed hypothesis is preliminary in nature and its verification needs to be investigated by time series data of structurally dependent properties, for example, strength, which is the subject of future research.

Optimization of material properties by methods of desirability functions. Since the optimization of the set of recipe-technological factors is carried out in the considered version of ES-modeling according to three types of strength – R1h, R28 and R180, it belongs to the multi-criteria optimization methods based on the idea of compromise solutions.

One of the approaches in the analyzed case is the transition to a single criterion-convolution of partial criteria (1):

$$\Psi\left(\varphi_1\left(x_1,\ldots x_m\right),\varphi_2\left(x_1,\ldots x_m\right),\ldots\varphi_n\left(x_1,\ldots x_m\right)\right)\to max. \tag{1}$$

The convolution method found an engineering embodiment in the form of desirability functions. It can be viewed as an apparatus for interaction with a decision-maker whose task is to qualitatively determine the degree of importance of partial criteria and the type of function itself using weight indicators for each criterion.

The capabilities of the *Design Expert* package in were used to optimize the composite material, which allow, in particular, carrying out multi-criteria optimization of the research result and the selection of optimal solutions. This method is based on the multiplicative nature of the desirability function D, which is determined by formula (2) [18]:

$$D=\left(d_1^{r_1},d_2^{r_2},\ldots,d_n^{r_n}\right)^{\frac{1}{\sum_i r_i}}, \tag{2}$$

where d_i reflect the degree of desirability of each of the partial criteria $0 \le d_i \le 1$.

When maximizing the target level of X_i at the selected upper and lower limits of the interval of changes of this factor $High_i$ and Low_i correspondingly expressed as (3):

$$d_i = \left[\frac{X_i - Low_i}{High_i - Low_i} \right]^{wt_i} \qquad Low_i < X_i < High_i \,. \tag{3}$$

In the case of minimization of the level X_i, the corresponding value of desirability is (4):

$$d_i = \left[\frac{High_i - X_i}{High_i - Low_i} \right]^{wt_i} \qquad Low_i < X_i < High_i \,, \tag{4}$$

where wt_i – indicator of the "influence" of the desirability function (weight factor), at $wt_i = 1$, di-linear function; $wt_i > 1$ exerts a greater influence on the displacement of di towards the target value; $wt_i < 1$ – a less important degree of influence.

In the original task, not all criteria are equally important. The degree of importance r (2) is expressed in the form of integers from 1 ("insignificant criterion") to 5 ("extremely important criterion"). The method discussed above, embodied in the form of a program, represents a convenient and flexible engineering tool for solving the problems of optimizing composites and, in particular, lightweight aggregate concrete of accelerated carbonate hardening.

For the constructed ES-models (Table 3), it is possible to set a large number of optimization tasks that differ in indicators of importance and weight. Let's consider two of them, which have a direct engineering meaning.

Optimizing the result of long-term hardening. The obvious goal of creating a composite material is to achieve maximum strength during the entire long-term hardening process of 180 days. But strength in the regulatory control period of 28 days is also important. To achieve appropriate results, other restrictions and requirements (except, of course, fixed interval restrictions) are removed. The appropriate conditions allow us to construct partial optimization criteria and determine the distribution of degrees of importance for the desirability function (Table 7).

Table 7 – Parameters of the desirability function for optimization of long-term hardening

Factors and properties	Objectives	Lower limit	Upper border	Lower weight	Upper weight	Importance
Knitt	in the interval	-1	1	1	1	1
SYM	in the interval	-1	1	1	1	1
GL	in the interval	-1	1	1	1	1
PCO_2	in the interval	-1	1	1	1	1
tCO_2	in the interval	-1	1	1	1	1
R1h	maximum	2.1	16.5	1	1	2
R28	maximum	3.8	21.8	1	1	2
R180	maximum	5.9	26.1	1	1	5

The desirability function as a convolution criterion was optimized, and the following solutions were obtained in coded values (Table 8).

Table 8 – Results of optimization of long-term hardening

No	Knitt	SYM	GL	PCO_2	tCO_2	R1h	R28	R180	Desirability
1	**1**	**0.89**	**-1**	**0.08**	**0.52**	**14.8 8**	**21.2 1**	**24.6 3**	**0.926811**
2	1	0.93	-0.99	0.06	0.51	14.87	21,23	24.60	0.926516
3	1	0.99	-0.98	0	0.54	14.84	21.25	24.60	0.926229
4	1	0.98	-1	-0.17	0.23	14.25	21.48	24.84	0.925759

The first of them is the most profitable, it should be accepted. It is displayed graphically in the form of arc diagrams (Fig. 2).

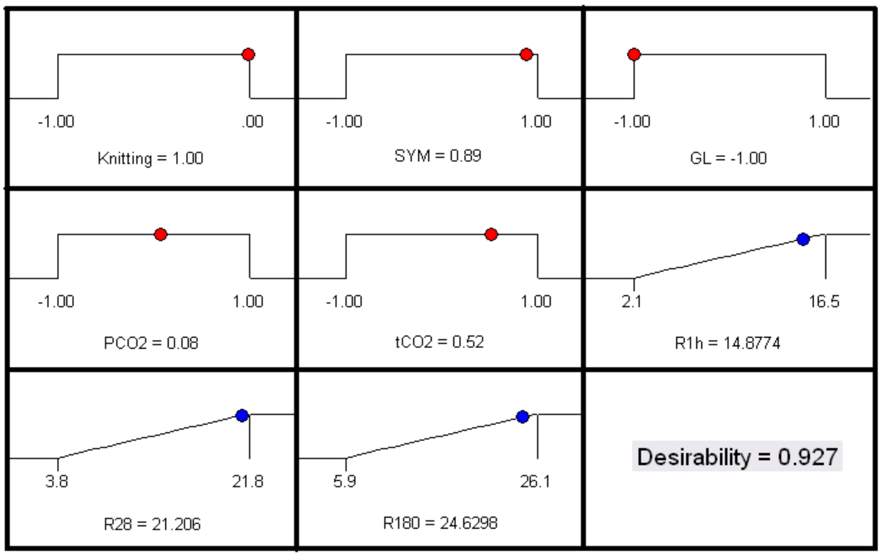


Fig. 2. The main result of multi-criteria optimization of long-term hardening

Optimization of the composition of the composite material taking into account additional conditions. Such additional conditions are the saving of the binder – cement, as well as technological conditions of low pressure and short carbonation time (this corresponds to safety conditions and reduction of construction costs for the carbonization chamber). Normative characteristics of the material at the 28-day hardening period are also important. But the main goal is still long-term strength. The formulated optimization problem allows you to construct partial optimization criteria and determine the distribution of degrees of importance for the desirability function (Table 9).

Table 9 – Parameters of the desirability function for optimization of long-term hardening

Factors and properties	Objectives	Lower border	Upper border	Lower weight	Upper weight	Importance
Knitt	in the interval	-1	1	1	1	4
SYM	in the interval	-1	1	1	1	3
GL	in the interval	-1	1	1	1	3
PCO_2	in the interval	-1	1	1	1	2
tCO_2	in the interval	-1	1	1	1	2
R1h	maximum	2.1	16.5	1	1	4
R28	maximum	3.8	21.8	1	1	5
R180	maximum	5.9	26.1	1	1	5

The desirability function was optimized, and the following solutions were obtained in coded values (Table 10).

Table 10 – Results of optimization of long-term hardening

No	Knitt	SYM	GL	PCO_2	tCO_2	R1h	R28	R180	Desirability
1	**0.57**	**1**	**-0.49**	**-1**	**-1**	**9.29**	**17.89**	**20.23**	**0.583693**
2	0.57	1	-0.48	-1	-1	9.32	17.91	20.24	0.58369 1
3	0.57	1	-0.48	-1	-0.98	9.33	17.91	20.2 3	0.583678
4	0.57	1	-0.52	-1	-1	9.28	17.92	20.29	0.583666

The first of them is the most profitable, it should be accepted. It is displayed graphically in the form of arc diagrams (Fig. 3).

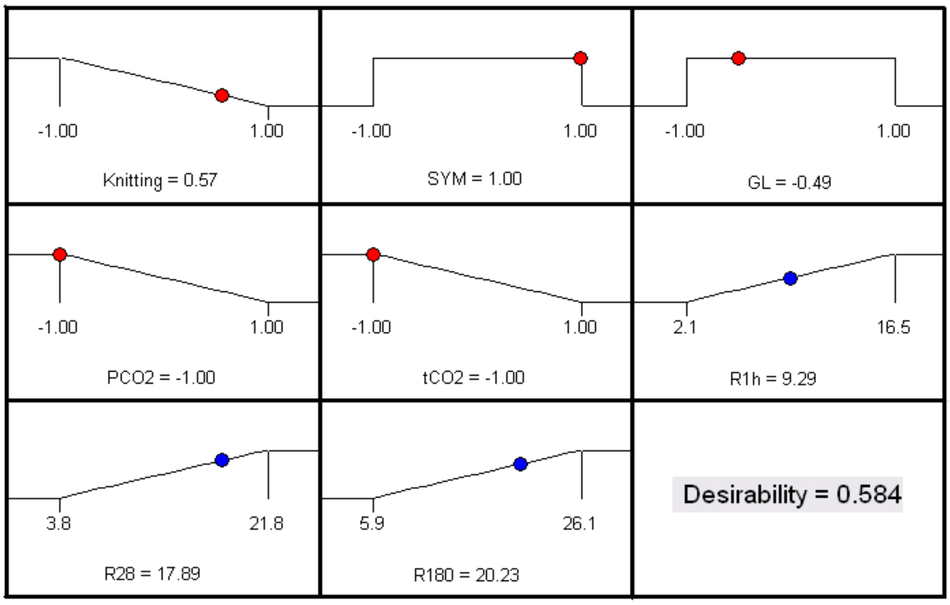


Fig. 3. The main result of multi-criteria optimization of long-term hardening in the presence of additional conditions and requirements

As can be seen from the comparison of the results in Tables 8 and 10, the imposition of additional conditions made it possible to facilitate the carbonation regime and save part of the cement, at the cost of losing 6 MPa of long-term strength. A significant decrease in the values of the desirability function in Table 10 indicates the significant impact of new requirements and restrictions, as well as the conditions of a hard compromise of partial criteria.

The determined optimal modes indicate the possibility of optimization under significant restrictions, which demonstrates flexibility in the application of the apparatus of the desirability function. The approach to quantitative optimization, which is based on the desirability function, is especially useful in those cases when the large dimension of the original problem does not allow to visually display the factor space, as well as when the interpretation of individual features of ES-models is difficult. These useful properties are fully manifested in the optimization of the recipe and technological factors for the production of expanded clay concrete of accelerated carbonization hardening.

Conclusions. The study of the strength data of expanded clay concrete of carbonization hardening made it possible to build ES-models of the strength characteristics of the material. The experimental strength values range from 2.1 to 16.5 MPa 1 hour after carbonization, from 4.6 to 21.8 MPa at the age of 28 days, and from 5.9 to 26.1 MPa at the age of 180 days. Models are graphically displayed and interpreted. The proposed hypothesis is that in experimental-statistical models of structure-dependent properties (for example, strength), during the process of structure formation, there is a tendency to simplify due to the elimination of a number of components, mainly members that describe the interaction of factors that determine the structure of the material. In particular, there is a gradual elimination of the constituent ES-models of the properties related to the material's structure. The formulation of two optimization problems of a set of recipe-technological factors based on long-term strength is proposed. The first optimization task concerns long-term strength in the absence of significant additional requirements and restrictions. The second task is loaded with additional requirements to reduce cement consumption and facilitate carbonation technology. In connection with the defined tasks, the parameters of the desirability function were determined, optimization was carried out and optimal sets of recipe-technological factors were obtained. So, for the second task, the composition and set of properties was as follows: Knitt = 0.57, SYM = 1, GL = -0.49, PCO_2 = -1, R1h = 9.29 MPa, R28 = 17.89 MPa, R180 = 20.23 MPa. The imposition of additional requirements made it possible to save cement consumption and improve the indicators of the carbonization technological regimes at the cost of reducing the long-term strength by approximately 6 MPa.

Reference

[1] S. Teir, S. Eloneva, R. Zevenhoven, "Production of precipitated calcium carbonate from calcium silicates and carbon dioxide", *Energy Convers Manage*, vol. 46, pp. 2954–2979, 2005.

[2] G. Groves, D. Rodway, I. Richardson, "The carbonation of hardened cement pastes", *Advances in Cement Research*, vol. 3(11), pp. 117-25, 1990.

[3] S. Kashef-Haghighi, S. Ghoshal, "Physico-Chemical Processes Limiting CO_2 Uptake in Concrete during Accelerated Carbonation Curing", *Ind. Eng. Chem. Res.*, vol. 52 (16), pp. 5529-5537, 2013.

[4] D. Suescum-Morales, K. Kalinowska-Wichrowska, J.M. Fernández, J.R. Jiménez, "Accelerated carbonation of fresh cement-based products containing recycled masonry aggregates for CO_2 sequestration", *J CO_2Util*, vol. 46, 101461, 2021.

[5] O.A. Gara, "Osoblyvosti karbonizatsiynoho tverdinnya keramzytobetonu", *Visnyk ODABA*, vol. 62, pp. 22-27, 2016.

[6] V. Rostami, Y.Shao, AJ. Boyd, Z. He, "Microstructure of cement paste subject to early carbonation curing", *CemConcr Res*, vol. 42(1), pp. 186–193, 2012.

[7] B. Lagerblad, *Carbon dioxide uptake during concrete life cycle-state of the art*. Swedish Cem.Concr. Res. Inst., 2005.

[8] A. Junior, R. Filho, E. Fairbairn, J. Dweck, "A study of the carbonation profle of cement pastes by thermogravimetry and its efect on the compressive strength", *J Therm Anal Calorim*, vol. 116, pp. 69–76, 2014.

[9] B. Šavija, M. Luković, "Carbonation of cement paste: Understanding, challenges, and opportunities", *Construction and Building Materials*, vol. 117, pp. 285-301, 2016.

[10] L. Parrott,"Carbonation, moisture and empty pores", *Advances in Cement Research*, vol. 4(15), pp. 111-118, 1992.

[11] C. Shi, Z. Tu, M.Z. Guo, D. Wang, "Accelerated carbonation as a fast curing technology for concrete blocks", *Sustain. Nonconv. Constr. Mater. Using Inorg. Bond. Fiber Compos., WoodheadPublishing*, pp. 313-341, 2017.

[12] M. Thiery, G. Villain, P. Dangla, G. Platret, "Investigation of the carbonation front shape on cementitious materials: effects of the chemical kinetics", *Cement and concrete research*, vol. 37(7), pp. 1047-1058, 2007.

[13] A. Morandeau, M. Thiery, P. Dangla, "Investigation of the Carbonation Mechanism of CH and C-S-H in Terms of Kinetics, Microstructure Changes and Moisture Properties", *Cement Concrete Res.*, vol. 56, pp. 153-170, 2014.

[14] L. Bertolini, B. Elsener, P. Pedeferri, E. Redaelli, RB. Polder, *Corrosion of steel in concrete: prevention, diagnosis, repair*. John Wiley & Sons, 2013.

[15] J. Matthews, *Carbonation of ten years old concretes with and without PFA*. ProcAshTech, 1984.

[16] L. Barcelo, J. Kline, G. Walenta et al, "Cement and carbonemissions", *Mater. Struct*, vol. 47(6), pp. 1055–1065, 2014.

[17] V.A. Voznesenskiy, T.V. Lyashenko, *ES-modeli v komp'yuternom stroitel'nom materialovedenii*. Astroprint, 2006.

[18] R.H. Myers, D.C. Montgomery, C.M. Anderson-Cook, *Response surface methodology: process and product optimization using designed experiments*. John Wiley & Sons, 2009.

[19] F. Grant, *Design Expert 7.1*. Scientific Computing World, 2007.

[20] A.A. Gara, *Razrabotka karbonizatsionnoy tekhnologii keramzitobetonnykh stenovykh izdeliy*, dis…. k.t.n.: 05.23.05. Odesskiy inzhenerno-stroitel'nyy institute. Odessa, 1990.

[21] A.V. Kolesnykov, S.V. Semenova, O.O. Makovets′ka, "Modelyuvannya protsesiv

strukturoutvorennya kompozytiv metodamy teoriyi katastrof", *Suchasne budivnytstvo ta arkhitektura*, vol. 6, pp. 90-98, 2023.

[22] V.N. Vyrovoy, V.H. Sukhanov, O.A. Korobko, *Struktura materyala v structure konstruktsyy*. Polyhraf, 2016.

[23] I. G. Grankovskiy, *Strukturoobrazovaniye v mineral'nykh v'yazhuchikh sistemakh*. Nauk. dumka, 1984.

[24] Ye.S. Shinkevich, *Razrabotka nauchnykh osnov polucheniya izvestkovo-kremnezemistykh stroitel'nykh kompozitov neavtoklavnogo tverdeniya* : diss…dokt. tekhn. nauk., OGASA. 2008.

ОПТИМІЗАЦІЯ ЕФЕКТІВ ПРИСКОРЕНОГО КАРБОНІЗАЦІЙНОГО ТВЕРДНЕННЯ КЕРАМЗИТОБЕТОНУ

[1]**Гара О.А.**, к.т.н., доцент,
gara@odaba.edu.ua, ORCID: 0000-0002-2413-1860
[1]**Колесников А.В.**, к.т.н., доцент,
kolesnikov_himek@odaba.edu.ua, ORCID: 0000-0001-8737-0933
[1]**Семенова С.В.**, к.т.н., доцент,
semenova@odaba.edu.ua, ORCID: 0000-0002-5309-5854
[1]**Олійник Т.П.**, к.т.н., доцент,
oleynik_himek@odaba.edu.ua, ORCID: 0000-0002-4469-6797
[1]*Одеська державна академія будівництва та архітектури*
4, вул. Дідріхсона, Одеса, 65029, Україна

Анотація. У роботі досліджуються дані, одержані при дослідженні прискореного карбонізаційного тверднення керамзитобетону. Розглядається механізми та методика здійснення карбонізаційного тверднення, вплив на міцнісні характеристики рецептурних та технологічних факторів. Найбільш значущими з них були витрата в'яжучого, відносна кількість і гранулометрія наповнювача (змеленого вапняку) та заповнювача (керамзиту), концентрація пластифікуючої добавки (СДБ) та технологічні параметри карбонізації – максимальний тиск та час карбонізації. Розглядається природа впливу відповідних властивостей на міцнісні характеристики. Досліджувалися результати спланованого та здійсненого експерименту за допомогою методології поверхонь відгуку. Будувалися експериментально-статистичні моделі міцності при стиску на строках 1 година, 28 діб та 180 діб від кінця процедури карбонізації. Визначені ступені впливу різних чинників. Побудовані залежності відображалися за допомогою серії діаграм ізоліній міцності при граничних значеннях трьох факторів слабкого впливу. Запропонована гіпотеза, згідно з якою в експериментально-статистичних моделях властивостей матеріалів, тісно зв'язаних з їхньою структурою, створюється тенденція до спрощення під час проходження відповідними матеріалами стадії структуроутворення. Зникають складові, переважно пов'язані з взаємодією факторів, визначаючих структуру матеріалу. Відповідні трансформації виникають, зокрема, для побудованих моделей міцності. Сформульовані два завдання оптимізації довгострокової міцності керамзитобетону. Перша задача пов'язана з усуненням жорстких додаткових обмежень при оптимізації, при цьому довгострокова міцніть досягає свого максимуму. Друге інженерно обґрунтоване завдання, містить додаткові вимоги економного витрачання в'яжучого та зручності технологічного режиму карбонізації. Для обох завдань були визначені розподіли параметрів функції бажаності. В результаті проведеної оптимізації одержані два основних набори рецептурно-технологічних факторів і відповідних властивостей композиційних матеріалів.

Ключові слова: прискорене карбонізаційне тверднення, планування експерименту, оптимальність за комбінованим критерієм, функція бажаності.

Стаття надійшла до редакції 24.05.2024