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SILICATE BRICK BASED ON LIME-BELITE BINDER SYSTEMS AND DUNE SANDS

Naurizbaev Azamat., Ilyasov Allanazar., Qayratdinova Aynura., Nurymbetov Baxtiyar., Toremuratov Sharibay.
1Karakalpak State University named after Berdakh, Nukus.
2Karakalpak Scientific Research Institute of Natural Sciences of the Karakalpak Division of Academy of Sciences of the Republic of Uzbekistan.

Abstract

The possibility of improving the physicochemical and physico-mechanical properties of silicate materials obtained based on lime-belite binder systems and dune sands, for the first time was revealed in the article. Regulation of the hydration processes of structure-formation of lime-belite binder systems with local mineral microfillers under normal and hydrothermal conditions and the dosages of fillers were determined; the mechanisms of their action were explained.

Introduction

It is known that silicate materials, due to their high technical and economic performance, are widely used in construction. The development of the production of these materials needs to expand the raw material base using local raw materials. In the production of autoclave-silicate materials, various binders are used as a binder - lime, lime-belite, lime-slag, lime-cement, etc. [1].

A large number of publications were devoted to the issue of the theory of hardening of binders, however, so far there is no single generally recognized point of view on the processes of structure-formation during the hardening of silicate materials. Therefore, the study of the processes of hardening and hydration of structure-formation of silicate materials is relevant.

It is customary to call artificial stones molded from homogeneous mixtures of quartz sand, binder and water, taken in strictly defined quantities, and treated with saturated water vapor at a pressure of at least 0.8 MPa as silicate concrete products. Various building materials are made from silicate concrete - bricks, blocks, and panels for external and internal walls of residential and industrial buildings, floor slabs, heat insulating and facade slabs [2-3].

For the production of autoclave building materials, various binders are used, including lime-cement binders, for which Portland cement is preferably used with a maximum content of belite and minimum content of aluminate and aluminoferrite minerals, due to the specificity of hardening of mineral binders during autoclave treatment [4–5].

Objects and Methods of Research

The object of study in this article is silicate brick produced on the base of lime-belite binder systems and dune sands.

Lime-belite binder (LBB) material based on carbonate minerals of Karakalpakstan is close to a high-grade binder, so it can be used to produce building materials and products with high physical and mechanical properties.

The kinetics of hydration structures and the physicochemical properties of lime-belite binders from Akburly and Porlytau marls are studied in the article.

In accordance with the above, this work is devoted to the study of the fundamental possibility of optimal heat treatment, phase composition and properties of the products obtained, to the development of ways to control the processes of obtaining and hydration of structure-formation in the considered LBB, their rational use for the production of some silicate products.

The study of physicochemical and mechanical properties, as well as the brand of lime-belite binders based on carbonate minerals of Karakalpakstan, was conducted according to state standards GOST 23789-04.

From previous experiments conducted by the authors, it was found that the optimal characteristics of autoclaved samples were obtained using a lime-belite binder (LBB) by firing chalk marl at a temperature of 1000˚C [6].

In this article, lime-belite binder (LBB) was studied as a binder for autoclave hardening, obtained by high-speed firing of chalk marl from the Ustyurt deposit of Karakalpakstan with the following mineralogical compositions, %: SiO_2–20,5; R_2O_3–3,87; CaO–38,65; MgO–1,34; p.p.p. -35.46; hydraulic activity – 116 mg CaO/g.

Lime-belite binder was obtained on a sintering machine by quick firing of a mixture of granulated marl and crushed solid fuel (14% of the mass of the charge) of brand ASh. Q p/h =5200 kcal/kg. Clinker ($CaO_{\text{свод}}$ - 1.1%) was crushed to a specific surface Sуд=350 m^2/kg. The calcium lime activity used in the experiments was 89%.

Samples (2x2x2 cm) were molded by casting (W/S-0.35). Autoclaving mode is 2 + 6 + 2 hours at a pressure of 0.8 MPa and a temperature of 174.5°C.

The following mixture was studied: LBB - quartz sand - water. The optimal amount of sand in mixtures was determined: CaO - quartz sand - water, and LBB - quartz sand - water. In the lime–quartz sand–water system, the amount of CaO varied from 20 to 35% with an interval of 5%, and in the system LBB – quartz sand – water, the content of LBB varied from 70 to 85% with an interval of 5%.

Samples made with 25% calcium lime and 75% ground quartz sand showed a maximum strength of 11.8 MPa. In mixtures of LBB - quartz sand - water, the highest strength - 12.9 MPa had samples with 80% of LBB and 20% of quartz sand.

In the LBB–CaO–quartz–water system, the CaO content varied from 10 to 60% with an interval of 10%. The amount of quartz sand was calculated from previously established mixtures in systems LBB - quartz sand and CaO - quartz sand. The maximum strength - 12.4 MPa was found in samples of lime-belite binder (80% of LBB

+ 20% of lime) with the optimal amount of quartz sand - 46%. The optimal content of sand in other mixtures with LBB was determined.

The density of the samples varied within 1500-1700 kg/m^3.

Thus, studies of LBB under hydrothermal conditions show that autoclaving contributes to an increase in strength due to the intensification of the formation of hydrocompounds. In the building materials plant, a pilot batch of silicate brick was manufactured based on LBB, obtained by heat treatment at 1000°C and with a curing time of 90 minutes, of marls from the Ustyurt deposit and dune sand from the Muynak deposit.

After mixing LBB with sand, the mixture was loaded into an SMS-152 molding machine and steamed in an AT 2x17 dead-end autoclave at a pressure of 0.8 MPa. The duration of the cycle was 11 hours.

10 hours after the extraction, the brick samples were tested for bending and compression on a ZIM-P-125 hydraulic press.

The physical and mechanical properties of silicate brick were determined according to GOST 379-95, the results are shown in Table 1.

Table 1 The results of studies of the physical and mechanical properties of silicate brick based on LBB

№	mixture composition, %			water absorption, %	bulk mass	tensile strength, MPa	
	LBB	Dune sand	water			bending	compression
1	10	90	8	11,3	1738	3,12	11,3
2	10	90	7,6	11,5	1743	3,90	11,6
3	15	85	8	12,2	1750	4,30	12,4
4	15	85	7,9	11,9	1747	4,46	12,8
5	20	80	7,7	13,5	1656	4,12	12,7
6	20	80	7,5	10,5	1678	4,30	12,9

According to GOST 379-95, the samples of brick tested for compression correspond to grades 100-125. The density of samples varied within 1500–1700 kg/m^3.

It should be noted that the produced silicate brick is one of the kinds of products based on LBB; on its base it is possible to obtain other silicate products (blocks, cell concrete, etc.) that are not inferior in their properties to products obtained with other binders.

Discussion and Research Results

The processes of hydration of clinker and other minerals that make up binders, as well as hydration structure-formation in these systems are interrelated; the latter usually follows from the former, so their coupled consideration is of scientific and practical interest from the point of view of their role in controlling the properties of binder systems.

In our case, natural marls from the Akburly and Porlytau deposits (Republic of Karakalpakstan) served as the raw material for the production of lime-belite binder (LBB).

There is little information in the literature on the study of hydration and structure-formation of binders in the presence of soluble anhydrite; this may be due to the instability or technical insignificance of the raw material [7].

In the system “lime-belite binder (LBB) - soluble anhydrite – water”, the product of dehydration of gypsum dihydrate – soluble anhydrite obtained by dehydration of gypsum dihydrate in the temperature range of 220-330°C was used as a gypsum filler. For the study, natural gypsum was taken, subjected to heat treatment in a laboratory oven at 300°C to obtain soluble anhydrite. The ratios of lime-belite binder to anhydrite were 90:10 and 85:15%.

Two processes cause the hydration process of structure-formation in the system lime-belite binder - soluble anhydrite - water: hydration of soluble anhydrite and hydration of lime-belite binder in the presence of anhydrite with the formation of calcium hydrosulfoaluminate.

Soluble anhydrite differs from hemihydrate in higher water demand, faster setting and reduced strength. Therefore, when obtaining building gypsum, heating of gypsum to temperatures at which soluble anhydrite formation is possible should be avoided [8].

The setting and hardening of soluble anhydrite when mixed with water are initially due to the rapid formation of a hemihydrate, followed by its transition to a dihydrate according to the reactions:

$$CaSO_4+0,5H_2O \rightarrow CaSO_4 \cdot 0,5H_2O$$

$$CaSO_4 \cdot 0,5H_2O+1,5\ H_2O \rightarrow CaSO_4 \cdot 2H_2O$$

The process of structure-formation of soluble anhydrite has the same character as the process of structure-formation of hemihydrate calcium sulfate. The process of hydration of soluble anhydrite proceeds rapidly in the initial periods of its mixing with water, and already by 60 min, as the author of [9] shows, it ends.

During heat treatment of marl, tricalcium aluminate is formed. When soluble anhydrite is added to LBB, it interacts with tricalcium aluminate (C_3A). Therefore, we will consider the mechanism of the formation of calcium hydrosulfoaluminate in the presence of lime since there is a certain amount of it in the LBB composition.

In the literature, the mechanism of formation of calcium hydrosulfoaluminate (HSAC) is explained in different ways depending on the conditions of the experiment and on the concentration of lime [2-4].

So, the authors of [5] believe that the interaction of gypsum with C_3A in a low-lime system ends after a few days, while some excess of free $CaSO_4$ remains, which did not react due to the lack of lime. At 50% of $Ca(OH)_2$ in the system, the participation of calcium sulfate in the reaction increases. At the same time, the process is inhibited by an excess of $Ca(OH)_2$.

In [6], the inhibitory effect of lime was considered during the interaction of calcium aluminates with gypsum in connection with the magnitude and kinetics of supersaturations created by it in the system. The latter determines the thickness of the screening films on the surface of C_3A from the first portions of HSAC and, consequently, the amount of substance consumed for their formation. An increase in CaO supersaturation causes a rapid sorting of small HSAC crystals and, accordingly, a decrease in the film thickness and the degree of participation of the initial components in the reaction.

Thus, the interaction of gypsum with C_3A in the presence of $Ca(OH)_2$ is accelerated, but the process is inhibited by an excess of calcium hydroxide.

Table 2 presents data indicating the kinetics of structure-formation in the system LBB - soluble anhydrite-water; the structure-formation was judged from the plastic strength of the system.

Table 2 Effect of soluble anhydrite on the kinetics of structure-formation of LBB_1 pastes

LBB	additive %	W/S	Terms of measurement of plastic strength, Pm, MPa											
			minutes			hours				days				
			1	15	30	1	2	4	6	1	3	7	14	28
Akburly	10	1,00	3,94	7,07	9,48	11,44	12,19	12,98	13,96	16,55	19,78	18,22	20,98	25,47
Akburly	15	1,10	5,25	9,19	10,7	12,17	13,05	14,10	16,86	18,20	20,90	19,47	21,56	28,25
Porlytau	10	1,00	3,69	7,60	10,2	11,78	12,34	13,04	14,69	16,95	18,30	17,86	20,95	26,05
Porlytau	15	1,10	5,91	8,60	10,5	12,74	13,69	14,19	17,45	18,36	21,76	20,34	22,45	28,27

As seen from the data in Table 2, with an increase in the amount of filler, the water-to-solid ratio (W/S) of the system increases. This is due to the crystal lattice of soluble anhydrite. The latter has a high specific surface area and porosity, resulting in its increased water demand. This influences the increase in the W/S system.

The introduction of soluble anhydrite causes increased strength at the initial period, that is, up to 7 days. This is due to the rapid hydration of soluble anhydrite with the formation of $CaSO_4{\cdot}2H_2O$ within 1 hour, since the hydration of soluble anhydrite ends at this time. After the formation of dihydrate gypsum, there comes a period of its interaction with tricalcium aluminate with the formation of trisulfate calcium hydrosulfoaluminate. The formation of calcium hydrosulfoaluminate at the initial period of hardening does not cause a decrease in strength due to the plasticity of the hardening system [10].

After 7 days, when the plasticity of the system is lost and its strength increases, the formation of the trisulfate form of calcium hydrosulfate aluminate with 31 water molecules causes a decrease in its strength. The strength properties of the LBB - soluble anhydrite - water system are shown in Table 3.

Table 3. Influence of soluble anhydrite on the mechanical strength during LBB hardening

LBB based on marl	Amount of filler, %	W/S	Compressive strength, MPa					
			moisture-air curing			hydrothermal treatment		
			3 days	7 days	28 days	3 days	7 days	28 days
Akburly	10	1,00	5,51	9,68	13,40	6,15	11,70	17,32
Akburly	15	1,10	7,25	13,56	16,19	8,47	15,73	20,50
Porlytau	10	1,00	6,18	10,26	14,65	7,47	12,70	18,43
Porlytau	15	1,10	8,05	13,76	17,43	9,15	16,10	21,86

The strength of the samples, as shown in Table 3, increases with time. Their initial strength is higher and this is due to the rapid setting of soluble anhydrite. After 7 days, the strength of the system slows down somewhat, but it is still higher after 28 days compared to samples with lime-belite binders without anhydrite additive, by 45% and 41% for the marls from Akburly and Porlytau deposits, respectively.

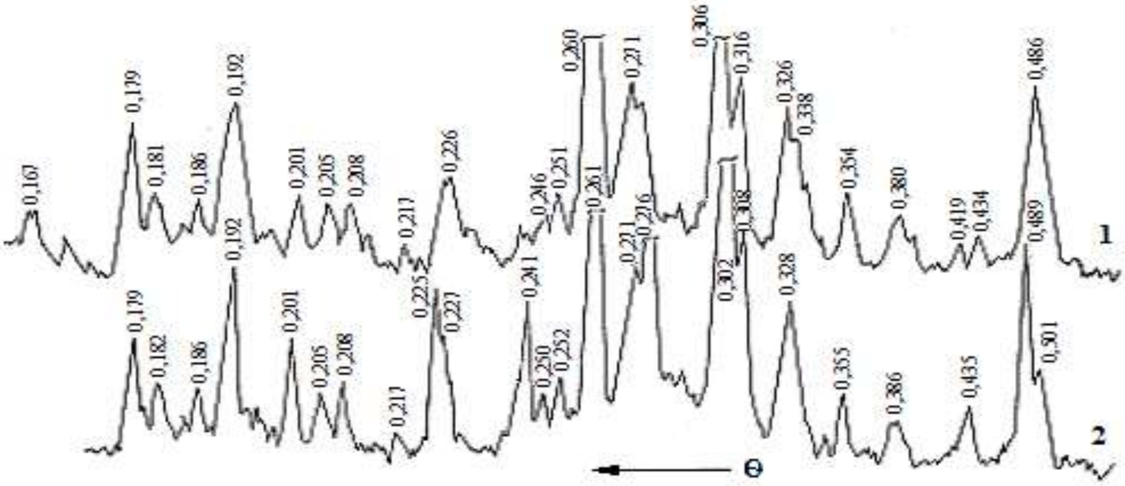

Fig. 1. Diffractogram of LBB_1 hydration products made with marls No. 1 and No. 2.

The influence of soluble anhydrite on the increase in the strength of the system is associated with the degree of supersaturation of the solution with respect to calcium ions and, accordingly, with the intensification of the formation of newgrowths. During thermal wet treatment, the strength of samples gradually increases with time [11].

The results of X-ray phase analysis of LBB_1 hydration products are shown in Figure 1. X-ray diffraction patterns of lime-belite binders hydrated for 28 days are characterized by the presence of more intense lines of gillebrandite (0.302; 0.271; 0.260 nm), portlandite (0.486 and 0.192 nm), calcium hydrosilicate of type C-S-H (II) (0.306 nm), as well as less intense lines of tobermorite $5CaO \cdot 6SiO_2 \cdot 5H_2O$ (0.251 and 0.248 nm). Lines 0.326; 0.205 nm and 0.186; 0.179 nm refer to calcium hydroaluminate and calcium hydroferrite, respectively. Under heat-moisture treatment of the sample, the intensity of the lines found in the air-moisture hardening increases.

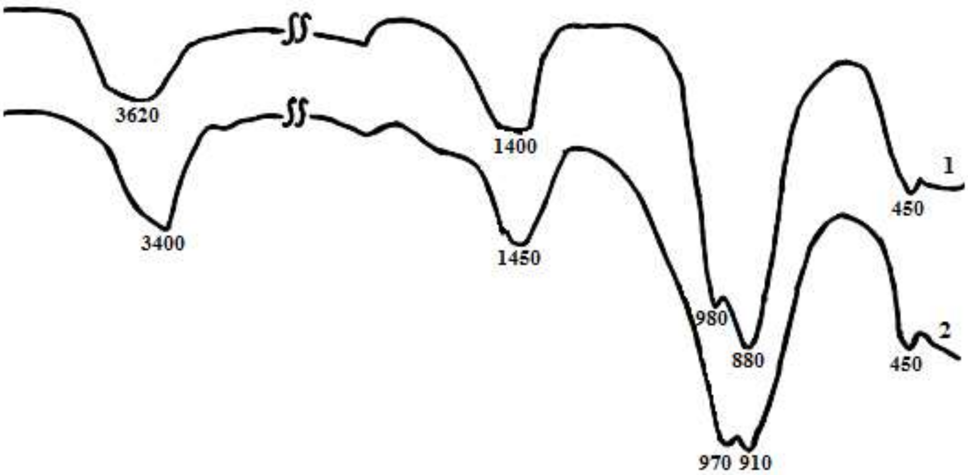

Fig. 2. IR absorption spectra of samples in the system LBB$_1$ - soluble anhydrite - water: LBB$_1$ based on marls from the Akburly (1) and Porlytau (2) deposits.

IR spectroscopic studies (Fig. 2) reveal the presence of intense absorption bands at wavenumbers 3400, 1450 and 970 cm^{-1} and weaker ones at 1640, 980 and 880 cm^{-1}. Vibrations in IR spectra in the range of wavenumbers 1400 - 1640 cm^{-1} indicate the presence of calcium hydroxide; absorption bands of stretching vibrations of calcium hydrosilicates are clearly expressed in the range of wavenumbers 970 - 980 cm^{-1}. Vibrations in the spectra of calcium hydrosulfoaluminate at 880 - 910 cm^{-1} are less pronounced. Detection of wavenumbers 3000...3400 cm^{-1} indicates the presence of water in the sample.

It was determined that the strength formation is associated not only with the formation of different types of structures in the system - coagulation and crystallization and with the transition of the first to the second, but also with various stages of the crystallization structure formation. Studying the physicochemical and physico-mechanical properties of the considered lime-belite binders, the authors of the article showed the possibility of using lime-belite binders for the preparation of high-strength building materials and autoclaved products.

It was established that under hydrothermal treatment (temperature - 175^{0}C, pressure - 0.8 MPa), the strength of the lime-belite binders obtained increases sharply. In the case of LBB$_1$ without filler, it reaches 40.0-42.0 MPa, and with the addition of soluble anhydrite it reaches 45.0-47.0 MPa, and in the case of LBB$_2$, 43.0-45.0 and 50.0-51 .0 MPa, respectively, which is associated with the influence of the filler and the acceleration of the processes of newgrowths formation and hydration structure-formation in the system [11].

Based on lime-belite binders and dune sands, it is possible to obtain autoclaved products with high physical and mechanical properties of the grades from 100 to 300 (10-30 MPa).

Lime-belite binders based on marls are effective binders that can be used to manufacture high-strength autoclave silicate materials and products. Various building materials can be produced from silicate concrete based on LBB - bricks, blocks, and panels for external and internal walls of residential and industrial buildings, floor slabs, heat insulating and facade slabs.

The production of autoclaved silicate materials based on lime-belite binders will bring huge economic benefits due to the production of binders using waste-free technology and silicate materials by low-cost low-temperature hydrothermal treatment.

Thus, the implementation into the production of the technology for autoclave-silicate material manufacturing will provide the construction industry of the Republic with modern building materials.

Conclusions

Thus, based on the data obtained, it can be concluded that the optimal amount of soluble anhydrite microfiller is 15%. Ettringite, formed by the interaction of gypsum with C_3A, gives additional strength to the sample.

It is shown that the effect of soluble anhydrite on an additional increase in the strength of the system is related to the degree of supersaturation of the solution with respect to calcium ions and, accordingly, with the intensification of the process of formation of newgrowths of gillebrandite and tobermorite, C-S-H (II).

The filler also interacts with tricalcium aluminate in the composition of the LBB with the formation of hydrosulfoaluminates, which cause an additional increase in the strength of the system under normal and hydrothermal hardening conditions.

Based on lime-belite binders and dune sands, it is possible to obtain autoclaved silicate materials with high physical and mechanical properties.

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