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Siliceous rocks as modifiers of structure of photocatalytic self-cleaning concrete. Assessment of the effect on the phase composition of cement stone

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ABSTRACT

Introduction. Currently one of the focus areas for the development of construction material science is the creation of self-cleaning concretes characterized by polydisperse multicomponent composition with the presence of nanoscale photocatalytic additives, primarily based on TiO_2 . These photoactive modifiers give the material a number of positive properties, including the ability to decompose atmospheric pollutants, to self-clean the surface, etc. The promising method for improving the functional characteristics of titanium oxide photocatalysts is the creation of nanostructured systems with 'core (substrate) – shell (photocatalyst)' architecture. Previous research results show that the final efficiency of the synthesized composite photocatalytic modifiers largely depends on the level of substrate reactivity in the cement system. The purpose of this study is to investigate the impact of three types of siliceous rocks (diatomite, trepel, and opoka) on cement stone formation processes and to identify the most effective raw materials for use as photocatalytic carriers in self-cleaning concrete compositions. **Methods and materials.** The methods of Kozeny-Karman, laser diffraction and X-ray fluorescence spectrometry were used to determine the specific surface area and parameters of granulometric and chemical compositions of silicite samples. The phase composition of siliceous rocks and modified cement systems was studied by X-ray powder diffractometry. **Results and discussion.** The main parameters of granulometric composition of diatomite, trepel and opoka were determined. The predominance of reactive modifications of free silica (47.6–78.0 wt. %), represented by amorphous opal-A or cryptocrystalline OCT-phase (opal-CT), were revealed in the structure of silicites. It was found that increasing the dosages of silica-containing additives from 0 to 10% resulted in decreased by 10–27% in the quantity of portlandite in the phase composition of cement stone aged 28 days, while the content of high-strength low-basic calcium hydrosilicates (C–S–H (I)) increased by 11–27%. **Conclusion.** The chemical and mineralogical composition peculiarities of silicites, as well as the nature of the impact of silica-containing modifiers on the structure formation processes of cement systems, determine the prospects of using opal-cristobalite rocks as dispersed photocatalyst carriers for self-cleaning concrete.

KEYWORDS: self-cleaning concrete, composite photocatalyst, siliceous rock, silica, pozzolanic reactivity, Portland cement, supplementary cementitious material, hydration, cement stone, microstructure, phase composition, X-ray diffraction.

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INTRODUCTION

Today one of the focus areas for the development of building materials science is the creation of high performance cement concretes characterized by polydisperse multicomponent composition with the presence of nano-

and micro-sized modifiers of various types (chemical, mineral, organo-mineral), giving the opportunity to directly influence the structure formation processes and properties of the material [1–10].

The nomenclature of modern modified concretes is quite diverse, including reactive powder concrete [11, 12],

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high-strength concrete [13–15], fiber reinforced concrete [16, 17], self-compacting concrete [18, 19], self-prestressing concrete [18, 20, 21], etc. A separate group contains photocatalytically active cement systems, which acquire a number of positive properties during light irradiation, including the ability to decompose atmospheric pollutants, to self-clean the material surface, etc. [22–27].

The studies [22, 23, 27–33] revealed that the use of photocatalytic additives based on anatase TiO_2 , characterized by non-toxicity, chemical stability and photoactivity under conditions of exposure to ultraviolet radiation, allowed to achieve the required level of special properties of self-cleaning concrete. The research results [34, 35] showed that the efficiency of photocatalytic process when using titanium dioxide in the composition of cement material could be reduced due to:

- reducing the amount of absorbed light and free surface for photochemical reactions because of the high content of micro- and macro-sized components in the concrete;
- narrowed spectral action range of TiO_2 , characterized by band gap of the rutile and anatase forms equal to 3.0 and 3.2 eV, respectively, which requires solving the problem of its sensitization to visible light;
- difficulties in ensuring uniform distribution of the photocatalytic nanomodifier in the cement material volume when its dosage is low, etc.

The promising method for improving the functional characteristics of titanium oxide photocatalysts is the creation of nanostructured photocatalytic systems with ‘core – shell’ architecture, the synthesis of which is by precipitation of TiO_2 on dispersed carriers (mineral substrates). Using this technology allows to increase the active surface area of titanium dioxide, and also promotes its more more equal distribution in the structure of complex modifier, thereby increasing the photoactivity of ‘carrier-photocatalyst’ compositions in the cement material [35, 36].

The study results by domestic and foreign authors [35–38] indicated that the final characteristics of the synthesized photocatalytic systems with ‘core – shell’ architecture significantly depended on the type of mineral substrate used.

It is known that siliceous raw materials of natural and man-made origin, in particular opal-cristobalite rocks (diatomite, opoka, trepel), are among the most efficient substrates upon obtaining the composite/hybrid photocatalytic modifiers for cement materials [35, 38–40]. The prospect of using silicites in the composition of self-cleaning concretes lies in their widespread occurrence, high affinity to crystalline hydrates of cement stone, as well as increased pozzolanic activity, owing to the predominance of amorphized varieties of silica in the structure [41, 42].

Notably, the effectiveness of using various opal-cristobalite rocks as photocatalyst carrier for self-cleaning

concrete can vary substantially depending on the composition and properties of silicites, in particular on the level of their reactivity in cement systems.

The aim of this study was to install the laws of the impact of three types of opal-cristobalite rocks (diatomite, trepel and opoka) from deposits of the Republic of Mordovia on the structure formation processes of cement stone and to find the most effective siliceous raw materials for use as photocatalyst carriers in the composition of self-cleaning concrete.

The following tasks were solved to achieve the aim of this study:

- 1) the chemical and mineralogical composition of opal-cristobalite rocks was established;
- 2) the main parameters of granulometric composition of silicites (specific surface area, particle size distribution) were determined;
- 3) the influence of siliceous rocks on the cement stone phase composition at the project age (28 days) was installed;
- 4) the most efficient types of silicites were identified for use as carriers of photocatalytic agents in the composition of self-cleaning concrete.

METHODS AND MATERIALS

Materials

Portland cement CEM I 42.5R (PC) that meets the requirements of the Russian State Standard GOST 31108-2020 was used as the main component of binder in the composition of cement materials. The researched siliceous rocks were diatomite (DMT), trepel (TPL) and opoka (OPK) from the Atemarsky, Dubensky and Alekseevsky field of the Republic of Mordovia, respectively.

The effect of mineral additives of silicites (MA) on the phase composition of cement stone at the project age (28 days) was studied in the process of experimental study. The variable prescription factors were:

- water-binder ratio ($x_1 = W/(PC+MA)$);
- type and content of mineral additives based on opal-cristobalite rocks: w_1 (DMT), w_2 (TPL), w_3 (OPK).

The variation levels of the studied prescription factors are presented in Table 1.

Methods

The methods of study silicites and cement systems are given in Table 2.

The crystallinity degree of structure of opal-cristobalite rocks was calculated based on the analysis of diffraction patterns of samples as the ratio of area of all crystalline peaks to the total area of crystalline peaks and amorphous halo.

Table 1

Variation levels of the prescription factors

Factors			Variation levels		
			-1	0	+1
Water-binder ratio, rel. units	x_1	W/(PC+MA)	0.33	0.35	0.37
Type and content of mineral additives based on silicites, % by weight of binder (PC+MA)	w_1	DMT	0	5	10
	w_2	TPL	0	5	10
	w_3	OPK	0	5	10

Note. The plan for experimental research was compiled taking into account fulfillment of the following conditions: $x_1 = -1, 0, +1$; $w_i = -1, 0, +1$ ($i = 1, 2, 3$).

Table 2

Methods for studying silicites and cement systems

Studied parameter	Used research method
Parameters of granulometric composition and specific surface area of siliceous rock powders	Laser diffraction and Kozeny-Karman methods using the Shimadzu Sald-3101 analyzer and the PSKh-12 device.
Chemical composition of silicites	X-ray fluorescence spectrometry using the ARL Perform'X 4200 spectrometer (Rh K_α radiation).
Phase composition of opal-cristobalite rocks and cement stone	X-ray powder diffraction on the PANalytical Empyrean multipurpose diffractometer with the Bragg-Brentano focusing geometry in the θ – 2θ scanning mode. The survey was performed using $CuK\alpha$ radiation characterized by weighted average wavelength $\lambda = 1.54 \text{ \AA}$. Phase identification was carried out on the basis of the Powder Diffraction File (PDF-2) from the International Center for Diffraction Data (ICDD).

When studying the phase composition of cement stone with additives of silicites, the main controlled parameters were the relative amounts of portlandite, low-basic calcium hydrosilicates (C–S–H(I)) and high-basic calcium hydrosilicates (C–S–H(II)). These parameters were determined as the ratio of intensities of the main reflexes $Ca(OH)_2$ ($d = 4.92$ – 4.96 \AA and 2.63 – 2.65 \AA), α - $CaO \cdot SiO_2$ ($d = 3.23$ – 3.25 \AA) / β - $CaO \cdot SiO_2$ ($d = 2.97$ – 3.00 \AA) and β - $2CaO \cdot SiO_2$ ($d = 2.78$ – 2.79 \AA) for samples of modified and control (additive-free) compositions, respectively. At the same time, the intensities of α -CS, β -CS and β - C_2S peaks were determined according to the diffraction patterns of cement stone powders calcined at 980 – $1000^\circ C$.

RESULTS AND DISCUSSION

Granulometric composition of silicites

The main parameters of the granulometric composition of opal-cristobalite rock samples were determined based on the study results using the laser diffraction method (Figure 1, Table 3). In particular, the particle size

range of diatomite, trepel and opoka was 0.3 – 123.4 \mu m , 0.6 – 153.8 \mu m and 0.8 – 171.7 \mu m ; the median particle size distribution (d_{50}) by volume was 11.1 \mu m , 7.9 \mu m and 7.3 \mu m ; the mean particle diameter was 10.3 \mu m , 9.6 \mu m and 10.9 \mu m , respectively.

It was found that the specific surface area of siliceous rock powders increased in the row of trepel → opoka → diatomite with values of $1.2 \text{ m}^2/\text{g}$, $1.3 \text{ m}^2/\text{g}$ and $2.0 \text{ m}^2/\text{g}$, respectively. With comparable average particle size of silicites, the increase by 1.5 – 1.7 times in the specific surface area of diatomite was due to the significantly higher porosity of its particles compared to opoka and trepel.

Chemical composition of silicites

Table 4 shows the chemical composition of the studied siliceous rocks.

The analysis of experimental data obtained by X-ray fluorescence spectrometry revealed the predominance of silicon, aluminum and iron oxides in the chemical composition of silicites. The content of SiO_2 , Al_2O_3 and Fe_2O_3 was 75.98 – $82.13 \text{ wt.}\%$, 5.34 – $9.62 \text{ wt.}\%$ and 3.11 – $4.11 \text{ wt.}\%$, respectively (Table 4). It is worth noting

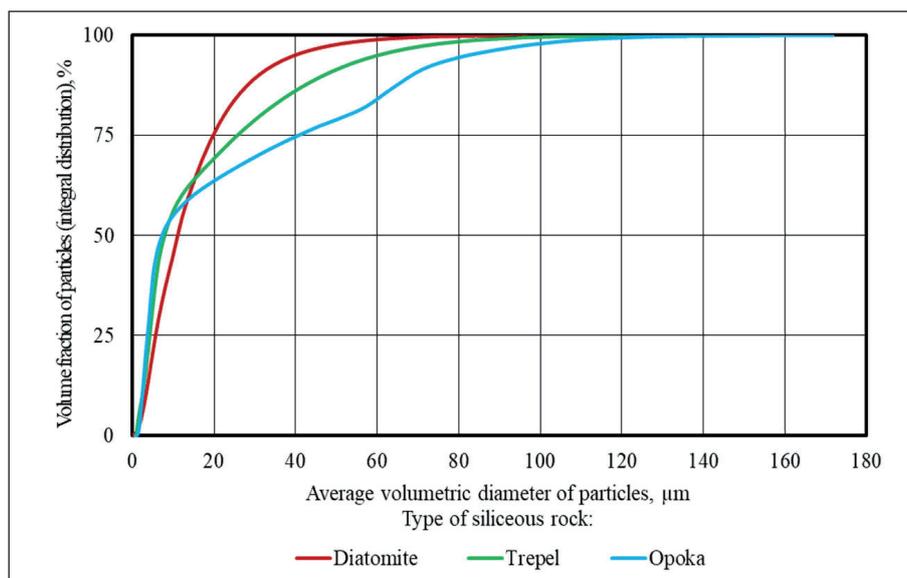


Fig. 1. Integral curves of the volumetric particle size distribution for silicite samples (according to laser diffraction)

Table 3

Main parameters of the granulometric composition of silicite powders

Parameter of granulometric composition	Type of siliceous rock			
	Diatomite	Trepel	Opoka	
Specific surface area, m ² /g	2.0	1.2	1.3	
Particle size range, μm	0.3–123.4	0.6–153.8	0.8–171.7	
Particle size, μm	d 25 %	5.6	4.2	3.6
	d 50 %	11.1	7.9	7.3
	d 75 %	19.8	25.6	40.5
	median	11.1	7.9	7.3
	mean	10.3	9.6	10.9

Table 4

Chemical composition of siliceous rocks (in recalculation on oxides)

Chemical composition	Siliceous rock		
	Diatomite	Trepel	Opoka
	Content, wt. %		
SiO ₂	81.47	75.98	82.13
Al ₂ O ₃	5.34	9.62	6.05
Fe ₂ O ₃	3.11	4.11	3.48
CaO	0.91	1.59	1.05
MgO	0.67	1.01	0.84
K ₂ O	0.97	1.49	1.16
Na ₂ O	0.43	0.50	0.48
TiO ₂	0.37	0.29	0.32
SO ₃	0.12	0.31	0.30
other (loss on ignition)	6.61	5.10	4.19

that the higher concentration of alumina recorded for the trepel sample (9.62%) were indicative of the increased content of clay minerals in it compared to diatomite and opoka. At the same time, increased weight losses during calcination of diatomite (6.61%) were associated with the growth of opal content in its structure, the amount of water in which can reach 10–20 wt.%.

Phase composition of silicites

Figure 2 and Table 5 show the results of qualitative and quantitative X-ray diffraction analysis of siliceous rock samples.

The phase composition of silicites consisted (Table 5) mainly of various polymorphic modifications of free silica (66.0–84.5 wt.%), in particular, amorphous opal (opal-A) and crystalline quartz for diatomite; crystalline quartz and quasi-crystalline (structurally disordered) opal-cristobalite-tridymite (OCT) phase for trepel and opoka. The OCT-phase (opal-CT) is the crystallization product of opal-A, and is the single mineral formation in the form of complex of amorphized/cryptocrystalline and crystalline varieties of silica (opal, cristobalite and tridymite).

It was found that despite significant differences in the opal-CT content of trepel and opoka (47.6 wt.% and 69.0 wt.%, respectively), the OCT-phase has a close ratio of components, in particular, opal/cristobalite/tridymite = 68–69/17/14–15 (Table 5). It is worth noting that according to [43], the fixed ratio of the OCT-phase components with almost equal concentrations of cristo-

balite and tridymite is specific to siliceous rocks located in platform areas.

It was revealed (Table 5) that the content of amorphous phase (opal) increased in the row of trepel → opoka → diatomite (33.0 wt.%, 47.0 wt.% and 78.0 wt.%), while the crystallinity degree of the studied siliceous rocks correspondingly decreased (67.0%, 53.0% and 22.0%). At the same time, decrease of 1.7 and 2.4 times in the amount of X-ray amorphous opal for opoka and trepel compared to diatomite was associated with increase in the content of quartz in their phase composition by 2.1 and 2.8 times (from 6.5 wt. % to 13.7 wt.% and 18.4 wt.%, respectively).

It was established that clay minerals in the amount of 11.6–17.7 wt.% (Table 5), represented mainly by micas and hydromicas from subgroups of muscovite and illite, were the second structural component of the studied silicites after free silica in terms of content. The insignificant amounts of smectites from the montmorillonite subgroup were also present in diatomite and trepel, as evidenced by the presence of typical reflex with $d = 15.6–15.0 \text{ \AA}$ and $2\theta = 5.7–5.9^\circ$ (Figure 2).

X-ray diffraction analysis showed the presence in the siliceous rock structure of feldspars in the amount of 1.9–5.7 wt.%. These phases are group of framework aluminosilicates of potassium (microcline, orthoclase), sodium (albite) and calcium (anorthite). In addition, the mineralogical composition of trepel contained 12.3 wt.% of zeolites from the subgroups of clinoptilolite and heulandite (Table 5).

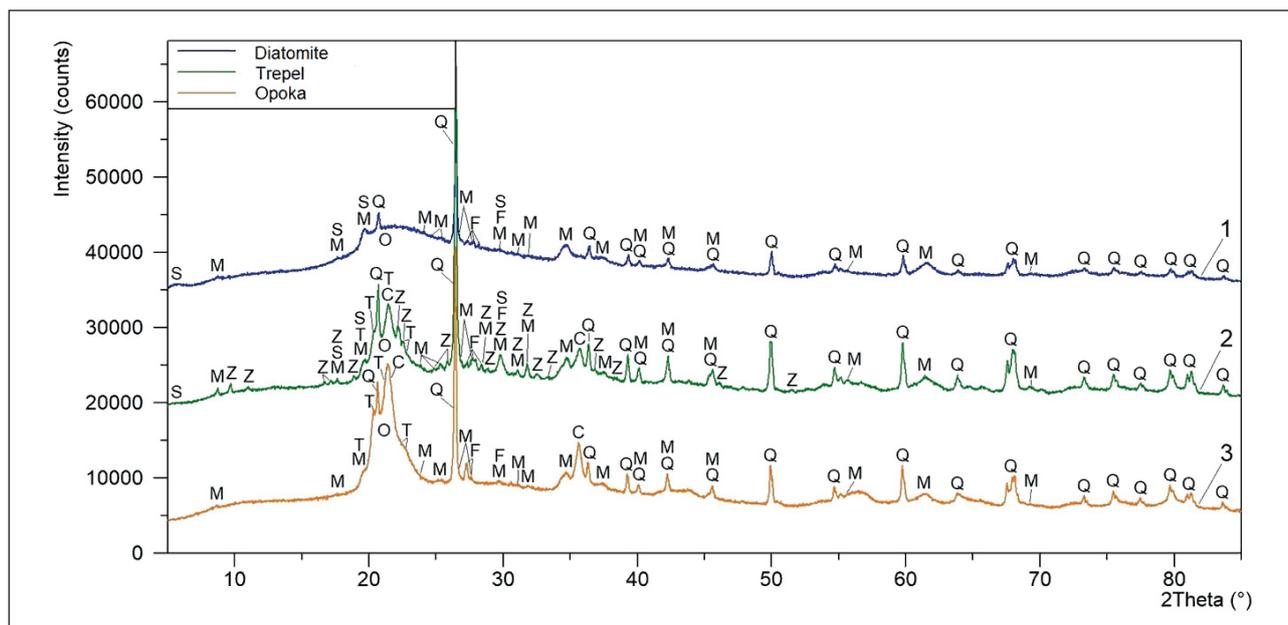


Fig. 2. Results of qualitative X-ray diffraction analysis of samples of diatomite (1), trepel (2) and opoka (3): opal (O); cristobalite (C); tridymite (T); quartz (Q); micas and hydromicas (M) in the form of muscovite and illite; smectites (S) in the form of montmorillonite; feldspars (F); zeolites (Z) in the form of clinoptilolite and heulandite

Table 5

Results of quantitative X-ray diffraction analysis of siliceous rock samples

Description of phases			Position of the main / typical reflexes d , Å (2θ , °)	Siliceous rock			
Type	Group	Subgroup, name		Diatomite	Trepel	Opoka	
<i>Phase composition, wt. %</i>							
Amorphous phase	Modifications of free silica	OCT-phase / Opal-CT (for trepel and opoka)	Opal	halo corresponding to 4,9–3,6 Å (18–25°) maximum in the area of 4,1 Å (21,6°)	78.0	33.0	47.0
			Cristobalite	4.13; 2.51 Å (21.5; 35.7°)	–	8.0	11.8
Crystalline phases	Modifications of free silica		Tridymite	4.36; 4.13; 3.94–3.88 Å (20.4; 21.5; 22.6–22.9°)	–	6.6	10.2
			Quartz	4.28; 3.36; 1.82 Å (20.7; 26.5; 50.0°)	6.5	18.4	13.7
		Clay minerals	Micas and hydromicas (muscovite, illite)	10.10; 5.02; 4.50; 2.58 Å (8.7; 17.6; 19.7; 34.7°)	13.6	17.7	11.6
	Smectites (montmorillonite)		15.6–15.0; 5.02; 4.50 Å (5.7–5.9; 17.6; 19.7°)				
		Zeolites	Clinoptilolite, heulandite	9.10; 8.02; 5.16; 4.00 Å (9.7; 11.0; 17.2; 22.2°)	–	12.3	–
	Feldspars		Alkaline feldspars (microcline, orthoclase)	3.26; 3.22–3.18 Å (27.3; 27.7–28.0°)	1.9	4.0	5.7
Plagioclases (albite, anorthite)			3.26; 3.22–3.18; 3.16; 3.14 Å (27.3; 27.7–28.0; 28.2; 28.4°)				
<i>Characteristics of the opal-cristobalite-tridymite (OCT) phase</i>							
Content, wt.%				–	47.6	69.0	
Ratio of opal/cristobalite/tridymite in the OCT-phase, %				– (opal is 100 %)	69/17/14	68/17/15	

Thus, despite the close total SiO₂ content (75.98–82.13 wt.% (Table 4)), the studied silicites had different ratios of structure-forming phases, primarily polymorphic modifications of free silica (opal, OCT-phase, quartz) and clay minerals. The phase composition variation predetermined the complex of the main functional properties of siliceous rocks, including their chemical activity in cement systems.

Influence of silicites on the phase composition of cement stone

It is known that calcium silicate hydrates (C–S–H) of different basicity and portlandite (Ca(OH)₂) are the main hydrate phases of cement stone due to the predominance

of alite (3CaO•SiO₂) and belite (β-2CaO•SiO₂) in the mineralogical composition of Portland cement clinker.

During the hardening of cement systems, the resulting calcium silicate hydrates have different composition and degree of crystallization (amorphization), which vary in wide ranges. Taking into account the chemical and mineralogical features, calcium silicate hydrates are conditionally divided into two main groups [44–46]:

1) high-strength low-basic calcium silicate hydrates (C–S–H (I)) with CaO/SiO₂ molar ratio less than 1.5, characterized by crystalline hydrates of colloidal size (less than 100 nm);

2) high-basic calcium silicate hydrates (C–S–H (II)) that are larger and less durable than C–S–H(I) and that have CaO/SiO₂ molar ratio at least 1.5.

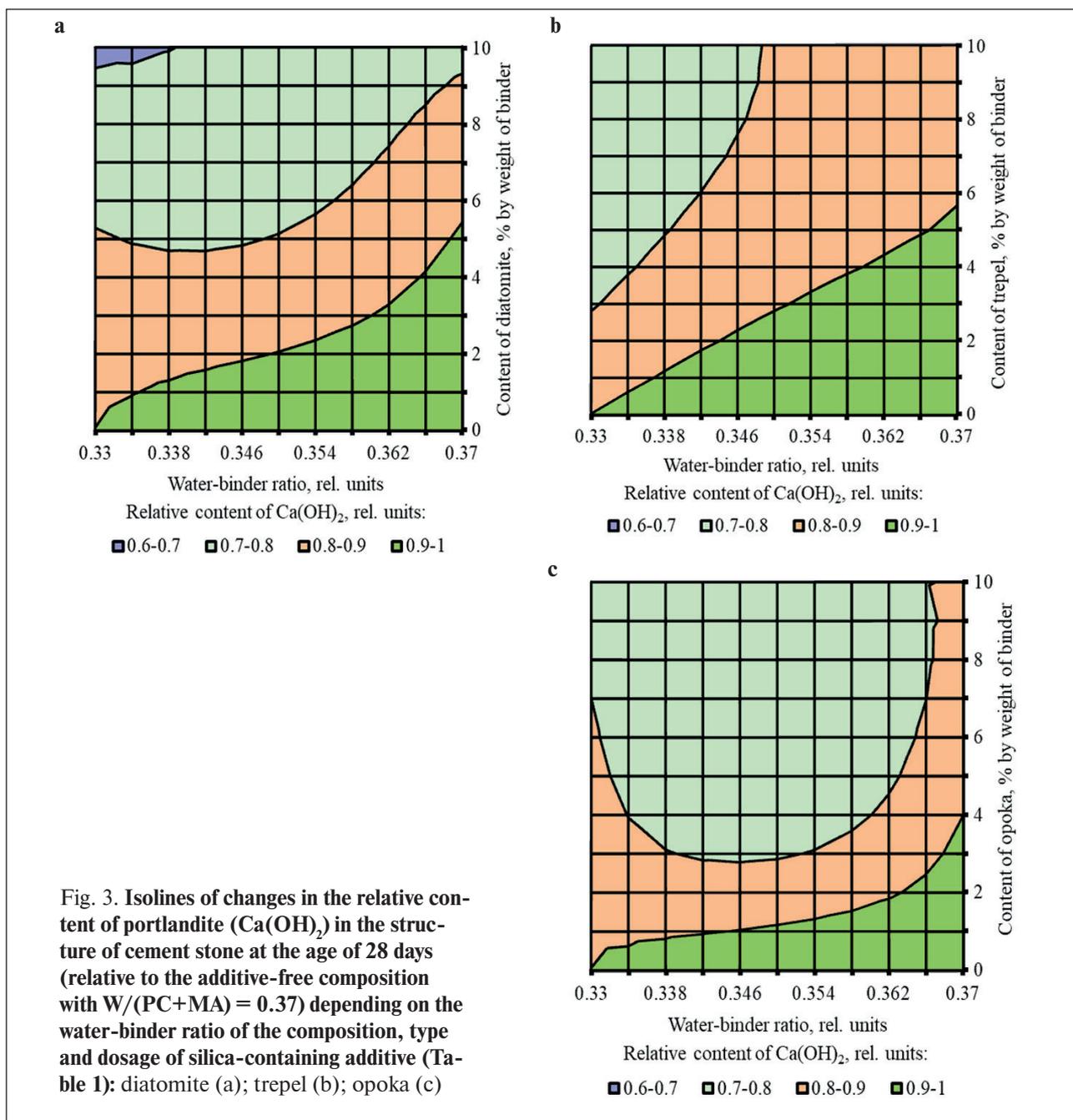
Typical representatives of low-basic calcium silicate hydrates of C–S–H (I) type are:

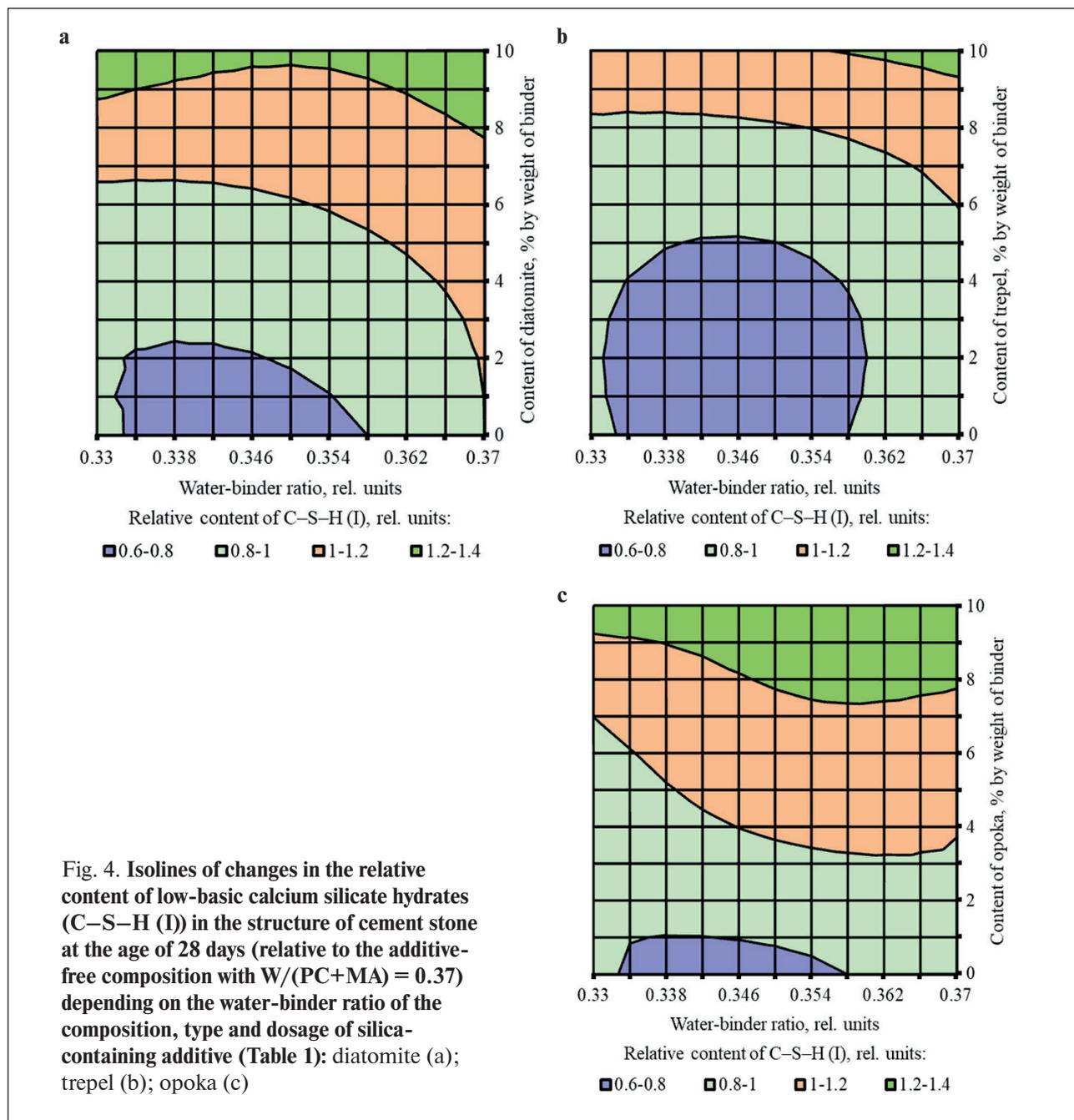
- tobermorite group with the CaO/SiO_2 molar ratio from 0.8 to 1.0, in particular, 9.3 Å tobermorite (riversideite, $5\text{CaO} \cdot 6\text{SiO}_2 \cdot 3\text{H}_2\text{O}$), 11.3 Å tobermorite ($5\text{CaO} \cdot 6\text{SiO}_2 \cdot 5\text{H}_2\text{O}$) and 14 Å tobermorite (plombierite, $5\text{CaO} \cdot 6\text{SiO}_2 \cdot 10.5\text{H}_2\text{O}$);
- compounds with the CaO/SiO_2 molar ratio from 0.5 to 1.3 that are similar to the wollastonite structure, in particular, nekoite ($3\text{CaO} \cdot 6\text{SiO}_2 \cdot 8\text{H}_2\text{O}$), okenite ($\text{CaO} \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$), xonotlite ($6\text{CaO} \cdot 6\text{SiO}_2 \cdot \text{H}_2\text{O}$), foshagite ($4\text{CaO} \cdot 3\text{SiO}_2 \cdot 1.5\text{H}_2\text{O}$);

- gyrolite group with the CaO/SiO_2 molar ratio from 0.5 to 0.7, in particular, gyrolite ($2\text{CaO} \cdot 3\text{SiO}_2 \cdot 2.5\text{H}_2\text{O}$), truscottite ($\text{CaO} \cdot 2\text{SiO}_2 \cdot 0.67\text{H}_2\text{O}$).

The most common high-basic calcium silicate hydrates of C–S–H (II) type are afwillite ($3\text{CaO} \cdot 2\text{SiO}_2 \cdot 3\text{H}_2\text{O}$), hillebrandite ($2\text{CaO} \cdot \text{SiO}_2 \cdot 1.17\text{H}_2\text{O}$), α -hydrate C_2S ($2\text{CaO} \cdot \text{SiO}_2 \cdot \text{H}_2\text{O}$), dellaite ($6\text{CaO} \cdot 3\text{SiO}_2 \cdot \text{H}_2\text{O}$), calcium chondrodite ($5\text{CaO} \cdot 2\text{SiO}_2 \cdot \text{H}_2\text{O}$).

Figures 3 and 4 show the study results of the effect of silicite and water dosages on the content of portlandite and low-basic calcium silicate hydrates of C–S–H (I) type in the structure of cement stone relative to the con-





trol additive-free composition with water-binder ratio of 0.37.

There was direct relationship between the water content of composition and the concentration of portlandite in the structure of the cement stone at the project age (28 days). It was established that reduction in the water-binder ratio from 0.37 to 0.33 rel. units led to decrease in the relative amount of $Ca(OH)_2$ in the phase composition of cement stone: by 10% in the control additive-free compositions (without silicites); by 12%, 16% and 3% in the modified compositions with dosage of diatomite, trepel and opoka equal to 10%

by weight of the binder, respectively (Figure 3). The increase in the content of portlandite with the growth of the water-binder ratio of cement systems in the accepted range of variation (0.33–0.37 relative units) was largely due to increase in the degree of Portland cement hydration in compositions with higher water content, as evidenced by the results of numerous studies, in particular [44, 45].

It was found that increase in the dosages of siliceous rocks in the modified cement systems from 0 to 10% by weight of the binder led to the growth in the relative content of low-basic calcium silicate hydrates of C–S–H (I)

type by 1.22–1.35 times, 1.11–1.26 times and 1.26–1.40 times in the compositions with diatomite (DMT), trepel (TPL) and opoka (OPK), respectively (Figure 4). At the same time, replacing 10 % of Portland cement with DMT, TPL and OPK helped reduce the relative amount of portlandite in the phase composition of cement stone by 22–27%, 10–19% and 13–21%, respectively (Figure 3). Thus, the study results of the $\text{Ca}(\text{OH})_2$ content in the structure of cement systems indicated an increase in the pozzolanic activity of opal-cristobalite rocks in the row of trepel → opoka → diatomite.

To summarize, the use of siliceous rocks made it possible to significantly change the quantitative relationship between the main hydrate phases of cement stone such as portlandite and calcium silicate hydrates of different basicity. The established shift in the balance towards an increase in the volume of high-strength low-basic calcium silicate hydrates of C–S–H (I) type (so-called secondary phases with CaO/SiO_2 molar ratio less than 1.5) instead of primary crystalline hydrates of $\text{Ca}(\text{OH})_2$ and high-basic calcium silicate hydrates of C–S–H (II) type, characterized by lower dispersion, strength and corrosion resistance, was the manifestation of the chemical effect [46, 47] in the action mechanism of silica-containing modifiers. This chemical effect was associated with the pozzolanic activity of silicites and was due to the predominance of reactive varieties of silica in their mineralogical composition, in particular, amorphous opal-A for diatomite and quasi-crystalline (structurally disordered) OCT-phase (opal-CT) for trepel and opoka.

CONCLUSION

The following scientific results were obtained in the paper:

1. The chemical and mineralogical composition of opal-cristobalite rocks was established.
2. The main parameters of granulometric composition of silicites (specific surface area, particle size distribution) were determined.
3. The influence of siliceous rocks on the phase composition of cement stone at the project age (28 days) was installed.
4. The most effective types of silicites were identified for use as carriers of photocatalytic agents in the composition of self-cleaning concrete.

The use of opal-cristobalite rocks made it possible to purposefully change the phase composition of cement stone by:

- decreasing the number of the $\text{Ca}(\text{OH})_2$ crystals, characterized by low strength and reduced corrosion resistance;
- increasing the density and strength of calcium silicate hydrates by shifting the balance towards increasing the content of highly dispersed low-basic phases of the C–S–H (I) type instead of larger high-basic compounds of C–S–H (II) type.

The established nature of the influence of silicites on the phase composition of cement systems indicates their increased chemical activity due to the predominance in the structure of reactive modifications of silica (opal-A, opal-CT). It determines the prospects of using siliceous rocks as dispersed carriers of photocatalytic agents for self-cleaning concrete.

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Tatyana A. Nizina – scientific advice; development of the concept and development of study methodology; analysis of research results; drawing up conclusions.

Vladimir M. Kyashkin – X-ray diffraction analysis of siliceous rocks and cement stone; collection and systematization of experimental data; drawing up conclusions.

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