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циклических нагрузках в химически агрессивных средах. Проведено исследование структурной неоднородности наплавленного металла и его влияние на работоспособность пресс-форм. Проанализированы все варианты наплавки способом плазма-МИГ наплавки и на основе проведенных исследований принято оптимальное решение, позволяющее существенно повысить скорость кристаллизации наплавленного металла.

Ключевые слова: плазма-МИГ наплавка, порошковая проволока, структурная неоднородность наплавленного металла, химическая неоднородность.

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INVESTIGATION OF THE INFLUENCE OF ORGANOMINERAL ADDITIVES ON THE COLLOID-CHEMICAL PROPERTIES OF GEOCEMENT DISPERSION

Наведено результати впливу органомінеральної добавки на колоїдно-хімічні властивості геоцементної дисперсії. Оптимізовано склад органомінеральної добавки і визначено області допустимих концентрацій її складових. Встановлено, що зміни величин інших вихідних параметрів прив'язані до зміни умовної в'язкості. Кількісне вираження вихідних параметрів знаходиться в межах: ρ =1,571–1,766 г/см^3, cosΘ=0,50–0,67, поверхневий натяг σ =114–128 мН/м, роботи адгезії, змочування і когезії, відповідно, 184–204 мН/м, коефіцієнти змочування і розтічності –0,77–(–)0,84, –37–(–)55 мН/м.

Ключові слова: *геоцементна дисперсія, оптимізація складу органомінеральної добавки, колоїдно-хімічні властивості.*

1. Introduciton

In recent years, materials based on alkali-activated binders – geocements or alkaline hydroaluminosilicates – have become increasingly popular. In general, the main properties of the binders themselves are considered: the processes of structure formation, strength, frost resistance, etc. [1–12]. Occasionally, publications aimed at studying special properties (heat resistance, fire resistance, corrosion resistance, etc.) of geocement materials [13–23], including properties of protective coatings – adhesion, technological viscosity, etc., can be found occasionally [24]. However, in these publications, the authors do not at all concern the study of the colloidal-chemical properties of geocement binders, as the basis for obtaining highly filled composites. Therefore, the studies considered in this paper are relevant and are aimed at studying the influence of mineralogical on the colloidal-chemical properties of geocement dispersions.

2. The object of research and its technological audit

The object of research is the geocement dispersion of the heulandite-clinoptilolite composition of the structural formula $Na_2O{\times}Al_2O_3{\times}6SiO_2{\times}20H_2O$, modified with an organomineral additive. This additive consists of:

– redispersible in water dispersion powder of the terpolymer of ethylene, vinyl laurate and vinyl chloride – Vinnapas RI-551Z (Vinnapas 8031 H, Wacker Polymer Systems, Germany);
– microcalcite fraction of 2 microns (Nigtas, Turkey);
– aluminate cement Istra 40 (HeidelbergCement, Germany), taken in certain quantitative relationships.

Positive from the use of organomineral additives, in addition to elasticity, hydrophobicity and strength, is the improvement of the colloidal-chemical properties of geocement dispersion, namely:

– wetting contact angle;
– surface tension;
– works of adhesion, cohesion and wetting;
– coefficients of wetting and spreading.

For investigations in this work, a geocement dispersion of the composition $Na_2O{\times}Al_2O_3{\times}6SiO_2{\times}20H_2O$, obtained on the basis of metakaolin, microsilica, and sodium soluble glass is used. Optimization of the composition of the organomineral additive that affects the colloidal-chemical properties of the geocement dispersion is carried out using a three-factor simplex-center method of experiment planning in the mathematical environment of Statistica 12.0.

The number of additives is chosen as the variable factors, %: RI-551Z (factor X1), $CaCO_3$ (factor X2) and AC (factor X3), the changes of which are given in Table 1. As the output parameters are chosen: conditional viscosity, density, wetting angle, surface tension, work of adhesion, cohesion and wetting, wetting and spreading coefficients.

The results of the experiment planning are given in Table 2.

Table 1

Factors of variation

Factors, view	Natural	Coded	Variation levels		Variation interval
			0	1	
RI-551Z	%	X1	2	6	4
$CaCO_3$	%	X2	1.1	3.1	2
AC	%	X3	2.8	7.8	5

Note: RI-551Z (Vinnapas 8031 H) a water-redispersible dispersion powder of a terpolymer of ethylene, vinyl laurate and vinyl chloride; CaCO3 – microcalcite fr. 2 μm; AC – aluminate cement Istra 40.

Table 2

Experimental design matrix

№	The matrix of the plan in coded units			The matrix of the plan in natural units		
	X1	X2	X3	RI-551Z, %	$CaCO_3$, %	AC, %
1	0.00	1.00	0.00	2.00	3.10	2.80
2	0.33	0.33	0.33	3.33	1.77	4.47
3	1.00	0.00	0.00	6.00	1.10	2.80
4	0.50	0.50	0.00	4.00	2.10	2.80
5	0.00	0.00	1.00	2.00	1.10	7.80
6	0.50	0.00	0.50	4.00	1.10	5.30
7	0.00	0.50	0.50	2.00	2.10	5.30

The rheokinetic properties of the constituent organomineral additives have been separately studied. But there is no data on the complex or joint organomineral additive on the colloidal-chemical properties of geocement dispersion, as the basis for obtaining protective coatings, adhesives and other types of composite materials.

3. The aim and objectives of research

The aim of research is investigation of the colloidal-chemical properties of a geocement dispersion modified with a complex organomineral additive.

To achieve this aim it is necessary to accomplish the following tasks:

1. To optimize the composition of the complex organomineral additive.
2. To determine the physical and colloidal-chemical properties of geocement dispersion.

4. Research of existing solutions of the problem

Among the main ways to eliminate the gap in the study of the colloidal-chemical properties of geocement variances revealed in the resources of the world scientific periodicals, the following can be singled out:

– the quality of the binder, which is characterized by viscosity, surface tension, the presence of gels and microgels, particle size, colloidal stability and adhesion strength. The consistency of the binder itself and the strength, hardness and durability of materials based on it depend on the type and composition of these indicators [1–12];

– dependence of the surface tension of uncured mineral binders on the nature of the solvent. With an increase in the concentration of solvents, the surface tension of unconfined binders decreases, and the greater, the less the surface tension of the solvents [25–27].

In particular, works [28–31] are devoted to the study of adhesion phenomena, wettability and spreading at the interface of condensed phases. The work of *Wa* adhesion was calculated on the basis of the experimental values of the surface tension at the liquid-gas interface (σ) and the wetting contact angle (θ) by the Dupre-Young equation:

$$Wa = \sigma'(1+\cos\theta). \quad (1)$$

The work of cohesion was determined from the cost of energy for a reversible isothermal discontinuity of the binder in a section equal to a unit area. Since, a surface forms during the rapture in two units of area, the work of cohesion is equal to twice the value of the surface tension at the boundary with the gas:

$$Wc = 2\sigma. \quad (2)$$

The work of wetting was calculated by the formula:

$$Ww = \sigma'\cos\theta. \quad (3)$$

The coefficients of wetting and spreading were determined from the ratio of adhesion to cohesion and their difference:

$$S = Wa/Wc = (1+\cos\theta)/2, \quad (4)$$

$$f = Wa - Wc = \sigma'(\cos\theta - 1). \quad (5)$$

Spreading of liquid along a solid substrate can't be a positive quantity, since $\cos\theta - 1 < 0$, as noted in [7, 16, 17, 30, 31].

The authors of [27] showed that the deepening of knowledge on the colloidal properties of binders greatly simplifies the flow of technological processes for the manufacture of materials, as well as an understanding of the degree of their filling.

An alternative solution to the problem, presented in [7, 16, 17, 29], does not give a clear picture on the determination of the colloid characteristics of binders in a wide range of concentrations of additives that affect these properties.

In the opinion of the authors of the works [1, 7, 18, 25–27], it is possible to obtain more correct colloidal characteristics of binders only with the use of modern instruments [32].

In works [7, 24–29], devoted to the study of the viscosity of dispersions, the main drawback of capillary viscometers determining the parameters of the structuring of suspensions is shown.

The ways to improve the approaches to calculating the colloidal properties of suspensions were considered in [25, 29].

Thus, the results of the analysis allow to conclude that an understanding of the nature of the manifestation of the colloidal-chemical characteristics of suspensions leads to the production of high-quality and durable composite materials, increases the degree of their filling and, as a consequence, the field of application.

5. Methods of research

Traditional methods of colloid chemistry were used to determine the surface tension and the contact angle of wetting [25, 26, 29–31] – the stalagmometric method (the droplet counting method) and the sessile drop method. It is assumed that at the moment of separation, the surface tension force is equal to:

$$F = 2\pi' R'\sigma, \quad (6)$$

where *R* – the radius of the hole from which the drops flow; Σ – the surface tension of the liquid.

At the same time, the surface tension force is equal to the force of gravity:

$$P = m'g, \quad (7)$$

where *m* – the mass of one drop. So,

$$2\pi' R'\sigma = m'g. \quad (8)$$

It is practically impossible to measure the exact radius of the capillary hole, so a comparative method was used with the known surface tension of a standard liquid, for example, water:

$$\sigma_0/m_0 = \sigma_1/m_1 = g/(2\pi' R) = \text{const}, \quad (9)$$

where σ_0, σ_1 – the surface tension of water and the liquid, respectively, m_1 и m_0 – the mass of one drop of water and the liquid, respectively.

The cosine of the contact wetting angle was calculated by the formula:

$$\cos\theta = ((d/2)2-h2)/(d/2)2+h2). \quad (10)$$

The error of the methods is 10 %. Plate of black metal protruded from the rust deposit was the substrate.

6. Research results

As a result of optimization, mathematical models that characterize the influence of the concentrations of the constituent organomineral additives on the changes are obtained:

relative viscosity, η, s(B3-5):

$v = 185x1+93x2+101x3-56x1x2-80x1x3-$
$-36x2x3+345x1x2x3+0;$

– density of geocement dispersion, ρ, g/cm^3:

$v = 1.575x1+1.571x2+1.766x3+0.06x1x2-0.274x1x3-$
$-0.09x2x3-0.123x1x2x3+0;$

contact wetting angle, cos θ:

$v = 0.3436x1+0.6746x2+0.509x3+0.296x1x2+$
$+0.2904x1x3-0.2068x2x3-2.3934x1x2x3+0;$

– surface tension σ, mN/m:

$v = 114.5x1+114.21x2+128.39x3+4.38x1x2-19.9x1x3-$
$-6.56x2x3-8.91x1x2x3+0;$

– work of adhesion, *Wa*, mN/m:

$v = 153.84x1+191.26x2+204.14x3+40.88x1x2-$
$-17.64x1x3-53.64x2x3-261.15x1x2x3+0;$

– work of cohesion *Wk*, mN/m:

$v = 229x1+228.42x2+256.78x3+8.76x1x2-39.8x1x3-$
$-13.12x2x3-17.82x1x2x3+0;$

– work of wetting *Ww*, mN/m:

$v = 39.34x1+77.05x2+65.35x3+36.5x1x2+23.06x1x3-$
$-26.28x2x3-283.44x1x2x3+0.$

Coefficients:

– wetting s:

$v = -0.67x1-0.84x2-0.79x3-0.14x1x2-$
$-0.08x1x3+0.18x2x3+1.11x1x2x3+0;$

– spreading *f*, mN/m:

$v = -75.16x1-37.16x2-52.64x3+32.12x1x2+$
$+22.16x1x3-40.52x2x3-243.33x1x2x3+0.$

Analysis of mathematical models shows that the changes in the values of the output parameters displayed in Table. 3, namely: conventional viscosity; density; wetting angle; surface tension; works of adhesion, cohesion and wetting of geocement dispersions, factors *X*1...*X*3 have an influence that are significant. Also, the joint effect of factors, respectively, *x*1*x*2*x*3, *x*1*x*2, *x*1*x*2 and *x*1*x*3 has a significant effect.

Table 3

Colloid-chemical properties of uncured geocement dispersions

№	η, s	ρ, g/cm^3	cos θ	σ, mN/m	*Wa*, mN/m	*Wc*, mN/m	*Ww*, mN/m	*s*	*f*, mN/m
1	93	1.571	0.6746	114.21	191.26	228.42	77.05	–0.84	–37.16
2	120	1.599	0.4626	116.25	170.03	232.50	53.78	–0.73	–62.47
3	185	1.575	0.3436	114.50	153.84	229.00	39.34	–0.67	–75.16
4	125	1.588	0.5831	115.45	182.77	230.90	67.32	–0.79	–48.13
5	101	1.766	0.5090	128.39	204.14	256.78	65.35	–0.79	–52.64
6	123	1.602	0.4989	116.47	174.58	232.94	58.11	–0.75	–58.36
7	88	1.646	0.5401	119.66	184.29	239.32	64.63	–0.77	–55.03
K	72	1.564	0.6894	113.70	192.08	227.40	78.38	–0.85	–34.60

Note: η – conventional viscosity; ρ – the average density; cos θ – the cosine of the contact wetting angle; σ – surface tension; *Wa* – work of adhesion; *Wc* – work of cohesion; *Ww* – work of wetting; *s* – wetting coefficient; *f* – spreading factor; *K* – control composition.

The coefficients of wetting and spreading of geocement dispersions are significantly influenced only by the joint action of the factors *x*1*x*2*x*3.

Graphical interpretation of the models is shown in Fig. 1.

When analyzing the ternary surfaces of the influence of the concentrations of the constituents of the complex organomineral additive on the properties of the output parameters (Fig. 1, 2), the relationship between the conditional viscosity and the wetting coefficient is traced. And also between the wetting angle, the work of adhesion, wetting and the spreading coefficient and between density, surface tension and work of cohesion.

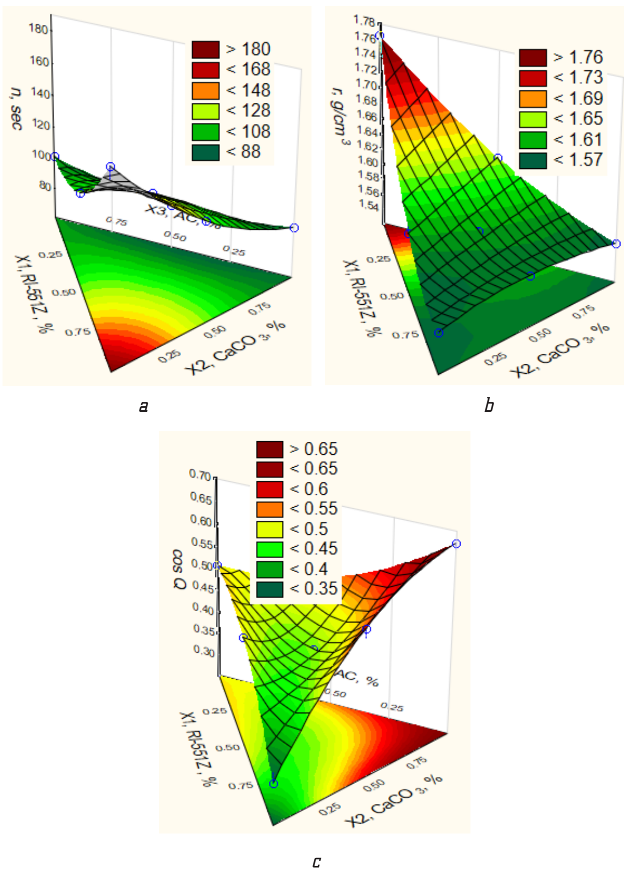

Fig. 1. Ternary surfaces of changes: *a* – conditional viscosity; *b* – density; *c* – angle of wetting of alkaline aluminosilicate dispersion, depending on changes in concentrations of variable factors

The defining index for assessing the colloidal-chemical properties of non-cured geocement dispersion is the conditional viscosity index, which should not exceed a value greater than 100 s.

The change in the conditional viscosity (Fig. 1, *a*) is more affected by the concentration of the polymer RI-551Z, the optimum amount of which is in the range from 2 to 2.31 %. Naturally, changes in the values of other output parameters will be tied to the change:

– the conditional viscosity and their values are in the following limits:

ρ =1.571–1.766 g/cm^3, cosΘ=0.50–0.67;

surface tension σ =114–128 mN/m;

works of adhesion, wetting and cohesion, respectively, 184–204 mN/m;

– coefficients of wetting and spreading –0.77– (–)0.84, –37–(–) 55 mN/m (Fig. 1, *b*, *c* and Fig. 2, *a*–*f*).

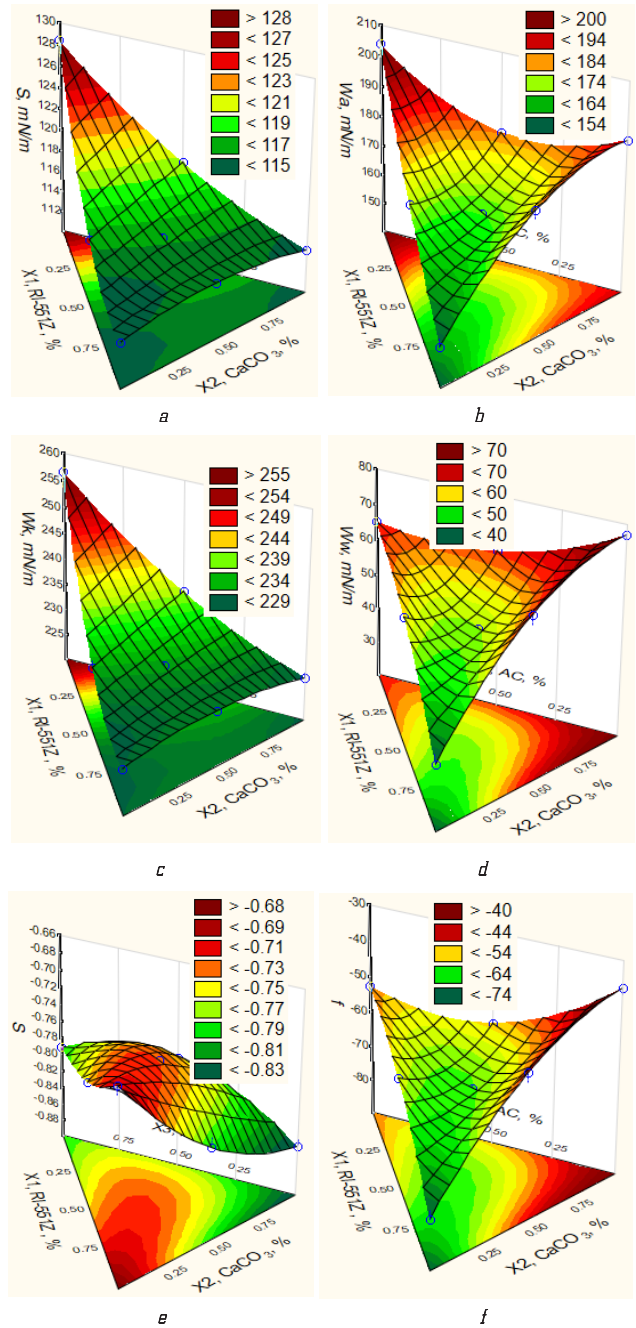

Fig. 2. Ternary surfaces of changes: *a* – surface tension; *b* – work of adhesion, *c* – work of cohesion; *d* – work of wetting; *e* –wetting coefficient; *f* – spreading coefficient of alkaline and aluminosilicate dispersion, depending on changes in concentrations of variable factors

7. SWOT analysis of research results

Strengths. The introduction of a complex organomineral additive containing from 2 to 2.3 % of polymer RI-551Z, microcalcite from 2.1 to 2.5 % and aluminate cement from 4.5 to 6.5 % contributes to the stabilization of the conventional viscosity, a decrease in surface tension, increase the work of adhesion and increase the wetting coefficient. It is also possible to regulate both the colloidal-chemical and technological properties of the geocement dispersion, as well as the degree of its filling with functional fillers. This will give a real opportunity in obtaining an even greater range of geocement-based materials.

Weaknesses. The negative effect of the object of research on its internal factors manifests itself in a constant rate of flow of the chemical reaction between the components of the dispersion and the mineral constituents of the organomineral additive. This leads to a change in the colloidal-chemical properties in time, namely, to thickening and hardening of the composition.

Opportunities. Opportunities for further research will be directed to the search for stabilizers and retarders of the rate flow of chemical reaction between the components of the geocement dispersion and the mineral constituents of the organomineral additive.

Threats. The time factor and external factors may have a negative effect on the research object: temperature drop, mechanical shake, which will be the subject of further research.

8. Conclusions

1. Optimization of the composition of the organomineral additive is carried out and the area of existence of the optimal concentrations of the constituent organomineral additives is determined, namely:

– along the X1 axis, 2–2.3 % of polymer RI-551Z;

– along the X2 axis, 2.1–2.5 % of microcalcite;

– along the X3 axis, 4.5–6.5 % of aluminate cement, which, when introduced into a geocement dispersion, allows $Na_2O \times Al_2O_3 \times 6SiO_2 \times 20H_2O$ to stabilize its colloidal-chemical properties. The introduction of geocement dispersion into the composition of an organomineral additive of optimal concentrations will allow in time to stabilize the values of its viscosity (not higher than 100 s), to reduce the wetting angle, to reduce the surface tension, to increase adhesion and to reduce the wetting coefficient.

2. As a result of the experiment, a relationship is established between the conditional viscosity and the wetting coefficient, between the wetting angle, adhesion, wetting and spreading work and between the density, surface tension and work of cohesion. It is determined that the changes in the values of the other output parameters are tied to the change:

– the conditional viscosity and their values are in the following limits:

ρ =1.571–1.766 g/cm^3, cosΘ=0.50–0.67;

– surface tension σ=114–128 mN/m;

– works of adhesion, wetting and cohesion, respectively, 184–204 mN/m;

– coefficients of wetting and spreading –0.77– (–)0.84, –37–(–) 55 mN/m (Fig. 1, *b*, *c* and Fig. 2, *a*–*f*).

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ИССЛЕДОВАНИЕ ВЛИЯНИЯ ОРГАНОМИНЕРАЛЬНЫХ ДОБАВОК НА КОЛЛОИДНО-ХИМИЧЕСКИЕ СВОЙСТВА ГЕОЦЕМЕНТНОЙ ДИСПЕРСИИ

Приведены результаты влияния органоминеральной добавки на коллоидно-химические свойства геоцементной дисперсии. Оптимизирован состав органоминеральной добавки и определены области допустимых концентраций, ее составляющих. Установлено, что изменения величин остальных выходных параметров привязаны к изменению условной вязкости. Количественное выражение выходных параметров находится в пределах: r = 1,571–1,766 г/см^3, cosQ = 0,50–0,67, поверхностное натяжение s = 114–128 мН/м, работы адгезии, смачивания и когезии, соответственно, 184–204 мН/м, коэффициенты смачивания и растекаемости –0,77–(–)0,84, –37–(–) 55 мН/м.

Ключевые слова: геоцементная дисперсия, оптимизация состава органоминеральной добавки, коллоидно-химические свойства.

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