UDC 544.77.022:725.731
DOI 10.36622/VSTU.2023.57.1.003

V. V. Volkov [1], O. A. Volokitina [2], O. B. Kukina [3]

USE OF THE THEORY OF AGGREGATIVE STABILITY OF DISPERSED MATERIALS IN DESIGNING ROAD LAYERS WITH SPECIFIED PROPERTIES

Voronezh State Technical University [1, 2, 3]
Russia, Voronezh

[1] *PhD in Physics and Mathematics, Assoc.Prof. of the Dept.of Building Mechanics, tel.: (473)271-52-30, e-mail: kotlac@yandex.ru*
[2] *PhD in Engineering, Assoc.Prof. of the Dept. of Highway and Bridge Design, tel.: (473)271-52-02, e-mail: dixi.o@mail.ru*
[3] *PhD in Engineering, Assoc.Prof. of the Dept.of Chemistry and Chemical Technology of Materials, tel.:(473)271-76-17, e-mail: lgkkn@rambler.ru*

Statement of the problem. The article considers the task of developing a modern theory for creating technologies for processing large-tonnage dispersed materials and technogenic products using non-firing technologies in the industry and road construction as structural layers of roads.
Results. We applied the Deryagin-Landau equation, supplemented by introducing the potential of induction and dispersion intermolecular interaction in diffuse layers of composite particles to predict the strength characteristics of building materials. We propose the diagnostics of raw materials, taking into account the energy of dehydration, which makes it possible to predict their binding properties and the ability to contact-condensation hardening. We obtained a regression dependence of the change in the strength of the resulting material on technological parameters and showed that the main contribution to the strength characteristics is made by pressing pressure, temperature and mixing time.
Conclusions. We proved the effectiveness of using a mathematical model for optimizing the process of obtaining lime-sand phosphogypsum material. We showed the prospects for the application of the proposed provisions of the theory of aggregative stability of dispersed mineral materials in the creation of new materials with desired properties that meet the requirements of modern construction of buildings, structures and pavement bases.

Keywords: dispersed mineral materials, theory of aggregative stability, building composites, specified properties, structural layers of roads.

Introduction. Rational nature management is one of the priority areas of scientific and technological progress throughout the world. The issue of preserving the environment to ensure a comfortable and high-quality living of mankind has been relevant for a lot of decades.

An increase in the intensity, speed and load-bearing capacity of road transport observed in recent years calls for more attention to the transport, operational and strength indicators of roads whose major structural element is pavement and structural layers. The absence of local stone materials in a number of regions of Russia makes the use of by-products and industrial waste in road construction more and more commonly, which considerably reduces the cost of construction and logistics costs.

The work of scholars from various countries is devoted to the rational use of natural, artificial, technogenic dispersed materials.

Nevertheless there is currently no unified systematic scientific approach to addressing the problem of utilizing mineral dispersed materials of various origins in industrial technology, in the production of building materials, in the construction of foundation bases, structural layers of pavements, etc. As technologies and enterprises are becoming increasingly modernized, an increase in production automation, the transition of factories to the modern level of digitalization, the use of diagnostics and testing of materials in order to predict their rational use in the chemical industry, construction of buildings, structures, road construction.

The objective of the work is to adapt the theory of aggregative stability to the basics of non-firing technology for obtaining building materials. In order to achieve this goal, the problems of estimating the disjoining pressure during the interaction between lyophilic (hydrophilic) particles in a dispersion medium were addressed including the analysis of the various contributions of the induction and dispersion intermolecular interactions in the process of solvation (hydration).

The design of aggregatively stable building materials using non-firing technologies with aggregative stability of colloids is one of the major problems of colloid chemistry — the physicochemistry of disperse systems and surface phenomena. In order to form the structure of composites with desired properties and determine the most appropriate operating conditions for the resulting materials, it is essential to understand the processes of interaction between dispersed mineral components through water films located on their surface.

The analysis of research materials is indicative of the relevance of the direction in the field of the use of dispersed building materials of natural and technogenic origin in the production of building composites [3]. It is based on the assessment of the structure of a double electric layer on the surface of nano- and micro-sized hydrophilic particles of mineral building materials, the representation of the mechanisms of interaction between such particles allows solving the problems of optimizing the compositions, technological parameters of non-firing building composites and the technology of their production during semi-dry molding (pressing).

A large proportion among the materials for the construction of pavement bases is made up of clay containing calcium and magnesium aluminosilicates. Clay is a highly dispersed material; it contains nano- and microsized particles with a complex structure of a double electric layer on the surface. For the construction of road foundations and foundations, aluminosilicates of natural and technogenic origin can be used. The studies [18, 16] present the fundamental possibility of using ash and slag waste from the fuel and energy complex in the production of ceramic bricks by semi-dry pressing based on low-melting aluminosilicate loams. In [16], the grinding of fused corundum was carried out, followed by the formation of a finely dispersed powder with a content of Al_2O_3 of 93—95% in the form of a low-quality material. The study [18] looked at the physical and chemical processes that occur during the production of ceramic materials using drilling waste and ash and slag waste from thermal power plants in the production of wall ceramics by semi-dry pressing. In [19], the authors found that accelerated carbonization transformed the initial hydration products and thus contributed to continuous hydration, as well as natural carbonization. The microscopic crystalline phases formed as a result of carbonization blended well with the hydrated matrix phases. The work [15] showed the possibility of obtaining a binder from the waste products of citric acid production — citrogypsum — by calcination in a semi-industrial plant. In the study [23], the authors assume that various types of industrial wastes, such as fly ash, bottom ash, and blast-furnace slag, can be used in geopolymer technology. In the study [22], a building material was developed from construction and demolition debris and lime production waste. [24] focused on the study of the use of recycled wood, paper, cardboard, metal, glass, mineral wool, gypsum, concrete and ceramics as raw materials for composite materials. [20] showed that binder composites, quicklime-sand-sand and lime-cement, were obtained by researchers in the form of new compositions, on the basis of which silicate products with improved strength properties and performance characteristics were obtained. In [4], the processes of structure formation in the presence of organic additives of acetone-formaldehyde resin and a water repellent were described. It was proved in [10] that on the basis of self-disintegrating slags of hydraulic and air cooling, polymineral clays whose main components are montmorillonite and hydromicaceous minerals, it is possible to obtain autoclaved silicate materials with improved physical and mechanical characteristics. The study [13] describes a simple environmentally sustainable approach to the synthesis of unfired building bricks with a compressive strength of 47 Mpa where the main ingredients were cement kiln dust, red clay brick waste and microsilica. The subject of the study [9] is the development of soft environmentally friendly and heat-treated adobe bricks. It also looks at the possibility of using waste generated as a result of the extraction and enrichment of raw materials in coal mines. It was established in [6] that argillite-like clays, argillites, and shales give strength to wall ceramic materials obtained by compression molding technology. The results of [12] show good compressibility of finely dispersed coal enrichment products and substantiate the high efficiency of using technogenic raw materials in the production of ceramic products. According to [1], clay has a satisfactory shaping abi-lity, medium plasticity, and is also characterized by low sensitivity to drying, low air and general shrinkage. It was shown in [11] that the main conditions for obtaining frost-resistant ceramic products of semi-dry pressing are fine grinding of raw materials by a dry method and the use of agglomeration processes (aggregation and granulation) in the technology for producing press powders, which allows solving the problem of uneven density of raw bricks. The results of [21] found that the use of finely dispersed recycled concrete aggregates and glass powder of various particle sizes in mortars reduces carbon dioxide emissions associated with concrete by up to 19%. In [5, 7], a complex of studies of phosphogypsum was carried out, which resulted in an assessment of the physical properties, environmental safety, as well as the development of a technology for its use in the design and construction of pavement structures.

The analysis of the above data shows that the problem of utilizing dispersed materials in construction can be solved by using them in the process of obtaining non-firing cementless artificial composites. At the same time, in the above studies the mechanisms of interaction between contacting particles are not presented, the structure of the double electric layer of colloidal particles is not considered, there is no thermodynamic assessment of the processes of structure formation, acid-base compatibility of components during molding under the action of press pressure, a systematic integrated approach to diagnosing the parameters of raw components in the process of obtaining artificial non-firing building composites with desired properties.

The scientific novelty of this work is in the use by the authors of the Deryagin-Landau equation to describe the mechanisms of interaction between contacting particles, investigation of the structure of the double electric layer of colloidal particles of hydrophilic dispersed building materials, energy assessment of the processes of structure formation in artificial building hydrophilic composites.

The working hypothesis of these studies is that a special place in the formation of the structure of building conglomerates belongs to water. Here and below, water should be considered as an aqueous solution containing ions and colloidal particles, micelles of materials forming a conglomerate. Water is an electrolyte that fundamentally changes the dielectric constant of the medium, predetermining the forces of interaction between particles [2].

While molding semi-dry mixes consisting of dispersed mineral building materials, wedging pressure arises between particles with hydrate shells — excessive pressure:

$$\pi(h) = p - p_0,\ \text{J/m}^2, \tag{1}$$

where π is the wedging pressure; p is the interfacial pressure; p_0 is the hydrostatic pressure in the environment; h is the thickness of the interfacial layer.

Moreover, these molding semi-dry mixes are thermodynamically unstable, and attractive forces prevail during pressing ($\pi < 0$). Wedging pressure is the work done when the thickness of the interfacial layer changes and per unit area of the overlapping surface of the surface layers provided that the thickness of the interfacial layer is half that of the layer surface (Fig. 1).

$$\pi(h) = \frac{\delta Ws}{dh},$$

where δW_s is the work done while changing the film thickness and per unit area of the surface of the overlapping of the surface layers provided $h < 2\delta$.

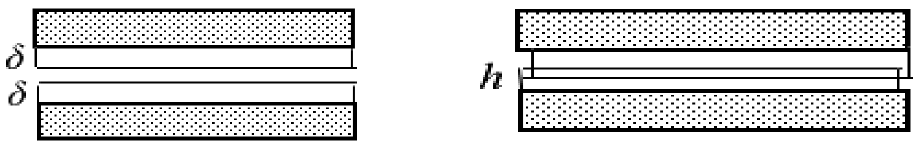


Fig. 1. Disjoining pressure in interfacial layers for h < 2δ (δ is the thickness of the surface layer)

From a thermodynamic point of view, the disjoining pressure is the partial derivative of the Gibbs energy with respect to the thickness of the interfacial layer:

$$\pi(h) = -\left(\frac{\partial G}{\partial h}\right)_{T,p,\mu}, \tag{2}$$

where π is the wedging pressure, J/m^2; h is the thickness of the interfacial layer, m; G is the Gibbs energy kJ/mol; T is the temperature, K; p is the pressure, Pa; μ is the chemical potential, J/mol (part of the free energy per 1 mole of any chemical element at constant pressure, temperature and mass of other substances).

The components of the disjoining pressure are molecular π_m, due to the van der Waals attraction forces acting between the nuclei of micelles, electrostatic π_e occurs when the double electric layer of micelles overlaps, due to electrostatic repulsive forces, adsorption π_{ads}, occurs when molecular adsorption layers overlap (adsorption of surface-active substances), structural π_{str}, is due to the existence of special liquid layers near the solid surface:

$$\pi = \pi_{м} + \pi_{э} + \pi_{адс} + \pi_{стр}, \tag{3}$$

where π_{ads} and π_{str} determine the adsorption-structural barrier, which ensures the stability of lyophilic systems that include dispersed mineral building materials.

In [22—27], the authors were able to to obtain a mathematical model of the change in disjoining pressure depending on the thickness of the water film for hydrophilic particles based on experimental data from the results of physical and mechanical tests of lime-sand phosphogypsum material. It has been established that when mixing the components of raw mixes, water is adsorbed in the form of films on hydrophilic particles. The maximum strength characteristics of the obtained materials are achieved at water film thicknesses of 10–7…10–9 m. At the temperature of 65…70 ºC and a change in the pH of water extracts from 2 to 8, water films change their thickness dramatically. At the same temperatures, the penetrating power of water sharply increases [2].

1. Materials and research methods. Phosphogypsum waste from the Uvarov Mineral Fertilizer Plant in the Tambov Region, quicklime produced by the city of Rossosh, Pridonkhimstroyizvest, sandy loam taken from a quarry located near the city of Uvarovo, and tap water were used. The molding of the samples (cylinders 5 × 5 cm in size) was performed using a PSU-125 hydraulic press. The ultimate compressive strength of cylinder specimens aged 1, 3, 7, 14, and 28 days was determined using an Instron 5982 universal electromechanical testing system. A JSM-6380LV scanning electron microscope was used to estimate the thickness of the solvate layers of interacting particles. In the course of the research, images of objects were obtained in the secondary electron emission mode at an accelerating voltage of 20 kV and an increase from ×350 to ×10000. The images show the microstructure of the cleavage surface of the samples. The micron marker present in the images makes it possible to estimate grain sizes, layer thicknesses, and scales of other features of interest.

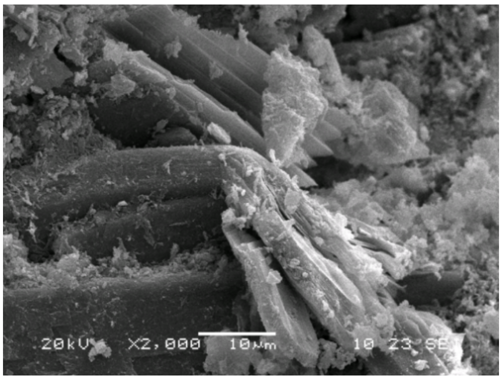
2. Research results. The results of the studies of the physical and mechanical characteristics of the non-firing lime-sand phosphogypsum material after 28 days of keeping the samples under normal humidity conditions of hardening are presented in Table.

Table

Physical and mechanical properties
of unburned lime-sand phosphogypsum material

Mass proportion of phosphogypsum, %	Compressive strength limit (28 days), MPa	Density, kg/m^3	Softening coefficient
60	4.76	1734	0.63

The results of microscopic examinations are shown in Fig. 2.



× 2000

Fig. 2. Crystallization contacts adjoining, intergrowth, germination between individual crystals of different phases lime-sand phosphogypsum material

3. Discussion of the results. The physical and mechanical characteristics of the unburned lime-sand phosphogypsum material indicate that with a mass fraction of phosphogypsum of 60%, the ultimate strength of the composite is 4—5 MPa, density 1700—1800 kg/m^3, softening factor 0.63. This is sufficient to use such material for the construction of pavement bases. As the thickness of water films decreases, there is an increase in the energy of dehydration, which indicates that in the number and strength of intercrystallization bonds. One of the causes of the solidity of the pressed lime-sand phosphogypsum material must be considered the high activity of the surface of phosphogypsum dihydrate, clay particles (Fig. 2). Pressing contributes to the creation of water film thicknesses corresponding to the data of the theory of structure formation of composites in dispersed media. In the general case, the energy of electrostatic repulsion between colloidal particles in a double electric layer is given by the formula:

$$U_{omm} = B \cdot e^{-kh}, \quad (4)$$

where U_{omm} is the energy of electrostatic repulsion, J; k is the reciprocal of the thickness of the diffuse part of the double layer, m^{-1}.

The repulsion energy of particles decreases exponentially with increasing distance h between them:

$$k = \frac{1}{\delta} = zF\sqrt{\frac{2C_0}{\varepsilon\varepsilon_0 RT}}, \quad (5)$$

where z is the charge of counterions; F is the Faraday number; C_0 is the concentration of counterions in the solution, mol/l; ε is the permittivity of the solution; ε_0 is the permittivity of the solvent;

$$U_{omm} = f(e^{-C_0, z}). \quad (6)$$

As the concentration and charge of the electrolyte in the solvate films of approaching particles increases, the repulsion energy decreases. This occurs under the action of external pressure in the process of pressing semi-dry mixtures. At the same time, the concentration of electrolyte ions increases with a decrease in the thickness of the solvate layers.

The attraction energy of approaching colloidal particles is given by the formulas:

$$dU_{прит} = \pi_{м} dh , \tag{7}$$

$$U_{прит} = -\frac{A^*}{12\pi dh^2} , \tag{8}$$

where A^* is the Hamaker's constant (considers the nature of particles and the dielectric constant of the medium, is calculated from quantum-statistical calculations, is expressed in units of energy and has a value of the order of 10—19 J).

In the case of the formation of a structure of a non-firing lime-sand phosphogypsum material at a pressing pressure of 5 MPa, the attractive forces prevail over the repulsive forces, the thickness of the solvate layers at the interfaces is 10—7...10—9 m, and the density of the material increases. Nanosized clay and lime particles remove the adsorption-structural stability barrier of this composite material resulting in the formation of crystalline intergrowths by the epitaxy mechanism.

Conclusions. Therefore the assessment of the energy state of the processes of overlapping of double electric layers made it possible to address the problem of estimating the disjoining pressure during the interaction between lyophilic (hydrophilic) particles of unburned lime-sand phosphogypsum material in a dispersion medium.

The efficiency of utilizing a mathematical model for optimizing the process of obtaining lime-sand phosphogypsum material is shown.

The prospects for the application of the suggested provisions of the theory of aggregative stability of dispersed mineral materials in designing new materials with desired properties that comply with the requirements of modern construction of buildings, structures and pavement bases are shown.

References

1. Andreeva A. V., Burenina O. N. Stroitel'nye materialy iz modifitsirovannogo glinistogo syr'ya dlya regionov kholodnogo klimata [Construction materials from modified clay raw materials for cold climate regions]. *Vestnik BGTU im. V. G. Shukhova*, 2011, no. 1, pp. 29—33.

2. Zolotukhin S. N. et al. Vliyanie tolshchiny vodnykh plenok na strukturu kompozitsionnogo stroitel'nogo materiala s ispol'zovaniem fosfogipsa [Influence of the thickness of water films on the structure of composite building material using phosphogypsum]. *Vestnik Voronezhskogo gosudarstvennogo tekhnicheskogo universiteta*, 2017, vol. 13, no. 4, pp. 138—143.

3. Zolotukhin S. N., Kukina O. B., Abramenko A. A., Volkov V. V., Eremin E. A., Volokitina O.A. *Bezobzhigovye stroitel'nye materialy iz otval'nogo fosfogipsa* [Bezobzhigovye building materials from dump phosphogypsum]. Saarbrucken, LAP LAMBERT publ., 2020. 160 p.

4. Ivashchenko Yu. G., Mukhambetkaliev K. K., Timokhin D. K. Effektivnye glinotsementnye kompozitsii, modifitsirovannye dobavkami [Effective clay-cement compositions modified with additives]. *Stroitel'stvo i arkhitektura*, 2014, no. 4, pp. 198—205.
5. Korotkovskii S. A., Talalai V. V., Kochetkov A. V., Yankovski L. V. [The use of phosphogypsum for the construction of a highway]. *Dolgovechnost' i nadezhnost' stroitel'-nykh materialov i konstruktsii v ekspluatatsionnoi srede: sb. statei po materialam I Mezhdunar. nauch.-tekhn. konf.* [Durability and reliability of construction materials and structures in the operational environment: collection of articles on the materials of the I International Scientific-Technical. conf.]. Moscow, MIFI publ., 2017, pp. 264—273.
6. Kotlyar V. D. et al. Osobennosti kamnevidnykh glinistykh porod vostochnogo Donbassa kak syr'ya dlya proizvodstva stenovoi keramiki [Features of stone-like clay rocks of the eastern Donbass as raw materials for the production of wall ceramics]. *Vestnik MGSU*, 2014, no. 10, pp. 95—105.
7. Kochetkov A. V., Yankovskii L. V. Primenenie fosfogipsa dlya stroitel'stva monolitnykh sloev dorozhnoi odezhdy [The use of phosphogypsum for the construction of monolithic layers of pavement]. *Zhurnal Permskogo natsional'nogo issledovatel'skogo politekhnicheskogo universiteta. Transport. Transportnye sooruzheniya. Ekologiya*, 2017, no. 4, pp. 91—102.
8. Kukina O. B., Abramenko A. A., Volkov V. V. Optimizatsiya sostavov bezobzhigovogo izvestkovo-peschanogo fosfogipsovogo materiala [Optimization of compositions of calcareous lime-sand phosphogypsum material]. *Nauchnyi zhurnal stroitel'stva i arkhitektury*, 2018, no. 3 (51), pp. 48—55.
9. Lam Tang Van et al. Ispol'zovanie otkhodov ugleobogashcheniya s tsel'yu polucheniya syr'ya dlya proizvodst-va stenovogo keramicheskogo kirpicha [The use of carbon enrichment waste in order to obtain raw materials for the production of ceramic wall bricks]. Vestnik BGTU im. V. G. Shukhova, 2017, no. 8, pp. 12—19.
10. Lupandina N. S., Voronina Yu. S., Starostina I. V. Glinoshlakovye materialy avtoklavnogo tverdeniya na osnove samorassypayu-shchikhsya staleplavil'nykh shlakov [Clay-slag materials of autoclave hardening based on self-dispersing steelmaking slags]. *Vestnik BGTU im. V. G. Shukhova*, 2012, no. 3, pp. 170—174.
11. Stolbushkin A. Yu., Ivanov A. I., Storozhenko G. I. Poluchenie morozostoikogo keramicheskogo kirpicha polusukhogo pressovaniya iz promyshlennykh otkhodov [Obtaining frost-resistant ceramic bricks of semidry pressing from industrial waste]. *Stroitel'nye materialy*, 2011, no. 12, pp. 4—7.
12. Yavruyan Kh. S., Gaishun E. S., Kotlyar A. V., Vlasova T. A. Keramicheskii kirpich kompressionnogo formovaniya na osnove tonkodispersnykh produktov ugleobogashcheniya [Ceramic compression molding brick based on finely dispersed carbon enrichment products]. *Vestnik nauki i obrazovaniya Severo-Zapada Rossii*, 2017, vol. 3, no. 4, pp. 44—50.
13. Abdel-Gawad Ala H. A. et al. The potential application of cement kiln dust-red clay brick waste-silica fume composites as unfired building bricks with outstanding properties and high ability to CO2-capture. *Journal of Building Engineering*, 2021, vol. 42, p. 27.
14. Acosta A. et al. Utilisation of IGCC slag and clay steriles in soft mud bricks (by pressing) for use in building bricks manufacturing. *Waste Management*, 2002, vol. 22, pp. 887—891.
15. Alfimova N. I. et al. Binders from gypsum-containing waste and products based on them. *IOP Conference Series: Materials Science and Engineering*. Belgorod: Institute of Physics Publishing, 2020. Vol. 945, p. 012057.
16. Getman A. A., Pavlova I. A., Farafontova E. P. The use of technogenic raw materials to produce a highalumina chamotte. *Materials Science and Engineering*, 2020, vol. 966, p. 012028.
17. Guryeva V. A., Doroshin A. The Press Powder Technological Parameters Optimization in wall Ceramics Production by the Semi-Dry Pressing Method. *Materials Science Forum*, 2019, vol. 974, pp. 419—423.
18. Guryeva V. A., Doroshin A. V., Dubineckij V. V. Features of the preparation of calcium-containing raw materials in the production of ceramic bricks. *Materials Science and Engineering*, 2020, vol. 775, p. 012111.
19. Engui Liu, Ghanim Kashwani, Liang Li Transformation of industrial solid wastes into carbon-infused in-frastructure materials. *Journal of Cleaner Production*, 2020, p. 120890.
20. Lesovik V. et al. Enhancing performances of clay masonry materials based on nanosize mine waste. *Con-struction and Building Materials*, 2020, p. 121333.
21. Letelier V. et al. Combined use of waste concrete and glass as a replacement for mortar raw materials. *Waste Management*, 2019, vol. 94, pp. 107—119.
22. Mymrin V. A. et al. Construction material from construction and demolition debris and lime production wastes. *Construction and Building Materials*, 2015, vol. 79, pp. 207—213.
23. Ong H. C. et al. Velopment of Ash-Based and Slag-Based Pressed Velopment of Ash-Based and Slag-Based Pressed Geopolymer. *Lecture Notes in Civil Engineering Volume*, 2021, vol. 129, pp. 51—72.
24. Sormunen P., Kärki T. Recycled construction and demolition waste as a possible source of materials for composite manufacturing. *Journal of Building Engineering*, 2019, p. 100742.

25. Zolotukhin S. N., Kukina O. B., Barabash D. E. Structure-forming role and properties of phosphogypsum in unburned technology of wall materials and rare-earth metals concentrate simultaneous production. *IOP Conference Series: Materials Science and Engineering*, 2019, p. 022028.
26. Zolotukhin S. N. et al. Waste-free phosphogypsum processing technology when extracting rare-earth metals. *Advances in Intelligent Systems and Computing*, 2019, vol. 983, pp. 339—351.
27. Zolotukhin S., Kukina O., Abramenko A. Partitions for high-rise construction using phosphogypsum. *E3S Web of Conferences*, 2018, p. 02043.