Розглянуто перспективи утилізації відходів нафтогазовидобутку у виробництві будівельних матеріалів. Встановлена принципова можливість застосування досліджених зразків бурових шламів як основної сировини і мінеральної добавки у складах мас для отримання стінової кераміки з необхідними споживчими властивостями.

Досліджено основні технологічні параметри отримання стінової кераміки з використанням зразків відходів газовидобування. Розроблені склади керамічних мас з використанням легкоплавкого середньоспікливого суглинку і бурових шламів в кількості 20–80 мас. %. Проаналізовані властивості отриманих керамічних зразків з використанням глинистих та висококарбонатних бурових шламів. Виявлено, що збільшення вмісту бурових шламів в зразках від 20 % до 80 % призводить до зменшення густини, міцності та підвищенню водопоглинення зразків, що впливає на якість кераміки та можливості її практичного використання. Встановлені закономірності зміни властивостей зразків стінових матеріалів від кількості дослідженого бурового шламу.

Визначені оптимальні кількості зразків бурових шламів для виготовлення стінової кераміки з нормативними властивостями. Встановлено, що з використанням глинистого бурового шламу (20-80 %) в композиції з легкоплавким суглинком можна отримувати морозостійкі керамічні матеріали з водопоглинанням на рівні 12 % і маркою М 125–М 175. Добавка висококарбонатного шламу до легкоплавкого суглинку в кількості 20 % дає змогу отримати морозостійкі керамічні матеріали з маркою М 75, а в кількості 40 % – матеріали марки М 100

Ключові слова: буровий шлам, керамічні матеріали, кераміка, густина, водопоглинання, морозостійка стінова кераміка, суглинок, мінеральна добавка

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ESTABLISHING THE REGULARITIES IN FORMING THE PROPERTIES OF CERAMIC WALL MATERIALS CONTAINING WASTE FROM GAS EXTRACTION (DRILLING SLUDGE)

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1. Introduction

The disposal of gas extraction waste is one of the unresolved issues that this industry faces at present. The toxic components of drilling muds, heavy metals, and oil included in the composition of drilling solid waste are dangerous to the environment.

In the drilling process, drilling sludge (DS) is discharged to the sludge barn to be buried after drilling operations are over. Burying the waste from hydrocarbon wells drilling does not fully resolve the issue.

Given the leakage of anti-filtration screens at the bottom of the barn, there are leaks of liquid waste that pollute the soil and aquifers. This adversely affects the environment due to the content of pollutants in the drilling waste, which can be very toxic. Therefore, it is necessary to implement modern environmental monitoring and drilling waste management systems [1].

For example:

– the minimization of the negative impact on the environment, reducing the volume of wastes;

– the implementation and improvement of the environmental management system in accordance with the requirements of the international standard ISO 14001:2015; accident prevention on equipment;

– the improvement of the environmental culture and awareness of employees about their role in addressing environmental issues.

Drilling each gas-extracting well produces, on average, about 0.4 m^3 of DS per a linear meter of the operational well. The depth of oil gas wells drilled, for example, in Ukraine, is, on average, from 2,500 meters to 6,000 meters. The volume analysis shows that the drilling of one well at a depth of 5,000 m can yield approximately 1,775 m^3 of drilling sludge [2].

One of the promising ways to dispose of drilling sludge is to use them in raw ceramic technologies, for example for making bricks. Typically, such wastes are used as technological additives, such as a clay substitute or as a supplement to natural clays.

The technological scheme for the disposal of drilling sludge in a combination with a phosphogypsum binder demonstrates [3] that 900 ons of sludge can produce 3,188.77 m^3 of gypsum concrete, that is, 229,592 gypsum concrete blocks. This indicates the prospects and economic feasibility of the use of drilling wastes in construction technologies.

Studies into determining the optimal amount of drilling sludge in the composition of ceramic products with norm-compliant characteristics are relevant at present. This is because identifying the optimal amount of drilling sludge could solve the task of the disposal of drilling solid waste and would lead to saving clay raw materials in the manufacture of ceramic wall products. Given this, an important issue is to study various drilling sludges for their use as raw materials in the technology of rough building ceramics. That would make it possible to increase the volumes of the disposal of drilling sludge, formed in the amount of about 900 tons at each well [2, 3]. The application of sludge in multi-tonnage ceramic technologies could effectively solve environmental problems in the oil and gas extraction regions, which adds relevance to the comprehensive environmental and technological studies into the possibility of disposal of a given type of industrial waste.

2. Literature review and problem statement

The use of oil and gas extraction waste in the production of building materials is a widespread international practice.

Article [4], for example, argues that drilling sludge can be used, after neutralization or hardening, in the manufacture of such building materials as bricks, ceramite, and other construction articles. The unresolved issue is the optimal content of sludge, which would not worsen the quality of the resulting products.

Paper [5] proposed the recycling of drilled rocks and sludge into slags to produce clinker materials. It was established that the production process based on waste recycling is environmentally-friendly and does not affect the quality of types of cement obtained. However, the properties of the resulting clinker materials were insufficiently studied.

It was found in [6] that in Egypt drilling sludge is successfully used as an additive in the manufacture of environmentally-friendly, rather effective construction bricks made of red clay. The results of the laboratory experiment show that the water absorption, apparent density, hue and compressive strength of the annealed briquettes correspond to the Egyptian standard «The Egyptian Code for Building Units» (ECP) No. 204-2005 for clay masonry blocks used for loading and bearing walls during construction. An important issue is a long-term process of preparing the drilling sludge. Sludge briquettes are left for two weeks in a dry place at room temperature, which significantly slows down the process of brick making.

Study [7] examined the potential of using drilling sludge in concrete as a partial replacement of cement. The novelty of the cited study is not only the production of new and cost-effective material from drilling sludge but also mitigating its negative impact on the environment. To achieve this objective, the authors performed a laboratory study aimed at quantifying the compression strength of concrete samples and determining the chemical composition of drilling sludge. The results showed that replacing 5 % of cement with dried drilling sludge reduces compression strength by 10 %. However, the overall strength of concrete samples decreases by 20 % when replacing 10, 15 % and 20 % of cement with drilling sludge. In addition, the authors investigated the influence of fly ash and silicon dioxide on the characteristics of the concrete samples containing drilling sludge. It was concluded that the addition of these additives significantly affects the compression strength of concrete samples containing 20 % of drilling sludge.

The solid drilling waste from a gas field in south-western China, considered in article [8], was collected and stored. Once drilling was completed, the drilling sludge was dehydrated and hardened with a cementing agent in a mud pool. It was established that the content of heavy metals and radioactive elements of the drilling sludge complies with the national standards for nonmetallic building material or wall material. Upon accumulation of certain quantities of hardened sludge, they were transferred to a brick plant to be further annealed to make bricks. The experiments demonstrated that 1 m^3 of drilling solid waste and 1 m^3 of shale clay could be annealed to make 1,000~1,200 bricks. Depending on the strength of annealed bricks, they were used for the household construction by local residents. However, the brick plant faced certain issues related to high operating costs, secondary air pollution during operation, and bad sales of the bricks.

In study [9], the solid waste of drilling, contaminated with oil, was stabilized by mixing. The authors mixed drilling sludge, ash, lime, and cement. The mixture was used as the base material for road construction. Such stabilization produced the physically, mechanically, and chemically stabilized new mixtures. The best result in all the tests was demonstrated by the soil polluted with oil, stabilized by 20 % of lime, 10 % of ash, and 5 % of cement. This mixture can be effectively and safely used as an underground material.

Work [10] reports the analysis of various drilling sludge recycling methods. The most common of them are thermal desorption, extraction with a solvent, bioremediation, burning, solidification/stabilization, and the drying of sludge. The main idea of the cited work is that there is no single universal method that can be applied anywhere. A method should be chosen according to the field application requirements. The authors suggested choosing a combination of methods according to the location of a drilling site and the properties of specific drilling sludge, which is inconvenient. The work did not study any particular efficient combined method for drilling sludge recycling.

Paper [11] proposed biological composting as the most effective method for heavily contaminated drilling sludge. The efficiency of composting was shown in comparison with other methods for the bioremediation of organic wastes. However, the disadvantage is that the mineral component is not subject to use.

The authors of [12] describe an innovative method of drilling waste recycling, namely the processing of wastes and their use as mineral additives of industrial nature for cement suspensions as a filler of the annular space at horizontal directional drilling. However, the addition of the dried and milled drilling sludge to cement suspensions, to fill the annular space, is possible only in very small quantities because sludge significantly reduces mobility and increases the curing time of the plugging solution. This is unacceptable and can lead to poor-quality cementing and, consequently, emergencies in the well's operation.

Article [13] considered the possibility of recycling drilling waste by using it as a filler for the manufacture of a polymeric composite material. The authors obtained the polymeric composites of high-density polyethylene, which were filled with the wastes of drilling with the content of waste up to 30 %. The result of the cited study is the established patterns of change in the impact viscosity, destruction stress at bending, as well as water absorption, depending on the solid phase content of used drilling mud in the secondary polymer. It was determined that there is an optimal content of the dispersed drilling waste in the composition of polymeric composites, which was 20 % by weight. Increasing or decreasing the share of waste within the composite formulation led to a decrease in the operational characteristics of the examined samples. Similar data were obtained by the authors of paper [14] when recycling the solid phase of finely-dispersed waste from foundry production but the optimal content of waste in the composite was at the level of 45–60 %. This indicates that the amount of waste as a filler of the polymer or ceramic mixtures depends not only on the waste dispersity but also on the chemical composition, properties, and other factors. As a rule, it is impossible to consider all the variety of factors that can affect the result. Therefore, the only advisable technique is to experiment with actual waste samples. Our analysis of publications [13, 14] testifies to the need to find the optimal amount of waste when it fills the examined samples.

Continuous drilling sludge treatment was demonstrated using a microwave chamber based on an electronic field applicator [15]. The microwave chamber is described as a device in which the electromagnetic energy interacts with the technological material. The microwaves are reflected from the metal walls of the chamber and interact with microwaves from the source of pollution, which would result in the destruction of pollution. The removal of the polluting oil depends on the applied power of the microwave chamber and the time inside the microwave cavity. The residual level of oil can be reduced to 1 % and lower. Under conditions of constant processing, it can be further reduced to 0.1 %. The energy consumption is very low, only 70–100 kWh per ton. It is shown that the 1 % discharge threshold can be achieved in sludge with a substantially variable oil and water content. Sludge consumption at the level of 500 kg/h is possible when using a microwave source of 30 kW. The cited paper did not investigate the possibility of treating sludge of alternating liquid content.

The authors of study [16] suggest using the drilling sludge pre-treated by low-temperature thermal desorption as a sand substitute for making sand-concrete blocks with improved properties. The sand-concrete samples with the ratio of sand to cement 6:1 were prepared. The results show that replacing up to 50 % by weight of sand with the drilling sludge contaminated by oil makes it possible to receive sandy concrete with reduced water absorption and reduced sorption, increased density, and reduced thermal conductivity. In addition, the compressive strength of the samples containing drilling sludge was compared to reference samples. However, the study does not address the estimation of durability and the properties of leaching of concrete sand blocks containing purified drilling sludge.

Thus, manufacturing construction articles using solid waste from drilling is the practice widespread all over the world. At the same time, the introduction of drilling sludge to the composition of ceramic mixtures as an anthropogenic raw material requires the study of properties of the obtained wall ceramic products. Despite the large body of research into the disposal of drilling sludge, the issue that remains unresolved is the possibility of recycling a particular sample of the drilling sludge from a particular well.

The issue of using drilling sludge as a raw material for building ceramics is associated with the considerable variability of the chemical and mineral composition of drilling sludge, as well as the composition of the organic residues of drilling muds included in it. Such variability in the sludge composition, predetermined by the depth of drilling and the lithology of drilled rocks, makes it difficult to obtain ceramic materials with reproducible properties even when recycling the waste from one drilling station. Therefore, experimental research into the quality of the obtained samples of the ceramic masses and their optimal composition is necessary for each type of drilling sludge from a particular well.

3. The aim and objectives of the study

The aim of this study is to find the optimal amount of drilling sludge in the composition of ceramic wall materials with norm-compliant properties.

To accomplish the aim, the following tasks have been set:

– to investigate the regularities of change in the basic technological parameters of wall ceramics depending on the amount of waste in the composition of ceramic masses;

– to determine the optimal amount of drilling waste to obtain the norm-compliant samples of wall ceramics with satisfactory operational characteristics.

4. Procedure to study the compositions of ceramic masses containing drilling sludge

The base used for ceramic masses was the loam of good technological quality, which refers to the typical brick-tiled raw materials, used by the plant that manufactures ceramic bricks in the city of Aktobe (Kazakhstan).

The waste used to fill the ceramic masses involved the samples of drilling sludge – the waste from gas well drilling in Poltava oblast; their properties, as well as the physical-chemical and mineralogical composition, are described in detail in [17].

When compiling the masses, we considered that sludge No. 1 (DS1) could replace clay raw materials, so it would be used in large quantities (from 20 % to 80 %). To ensure the possibility of comparing the annealing properties of the masses, sludge No. 2 (DS2) was used in the same quantity. The charge composition of ceramic masses is given in Table 1.

Table 1

The charge composition of ceramic masses

Mass code	The content of components in the charge, % by weight		
	Loam	Sludge 1	Sludge 2
1_2	80	20	–
1_4	60	40	–
1_6	40	60	–
1_8	20	80	–
2_2	80	–	20
2_4	60	–	40
2_6	40	–	60
2_8	20	–	80

The samples were obtained by the method of plastic molding under the normal moisture content of the masses; after drying, they were annealed at a temperature of 950 °C. Annealing the samples at a higher temperature, for example, 1,100 °C, is inappropriate according to previous studies whose results are reported in [17].

The annealing products from the masses are shown in Fig. 1, 2.

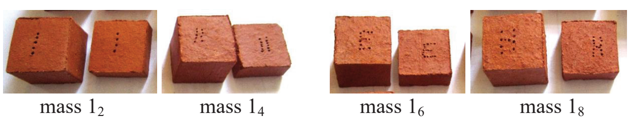

Fig. 1. Samples based on the mass containing sludge No. 1

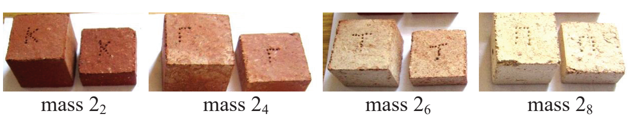

Fig. 2. Samples based on the mass containing sludge No. 2

The samples containing sludge No. 1, as expected, demonstrate a terracotta color and clear geometric dimensions. Sludge No. 2, when it is added in the amount of 40 % (mass 2_4) changes the color of the materials from terracotta light to light beige, due to the illuminated effect of carbonates that are in large quantities present in sludge. The samples containing sludge No. 2 can also retain their shape but their edges are susceptible to chipping, which degrades the physical appearance.

For the annealed ceramic samples, we determined the following characteristics based on standard procedures:

– mean density – the ratio of the mass of a sample with pores to its volume;

– water absorption – by methods of saturation and boiling;

– the limit of compressive strength – by measuring the destroying compressing load per unit area of the sample;

– a coefficient of structure (the predicted characteristic of frost resistance) – the ratio of water absorption by a sample, determined by a saturation method, to water absorption, determined by a boiling method.

We treated experimental data, constructed charts, and derived regression equations using the software «Statistica».

5. Studying the properties of ceramic samples using the samples of drilling sludge

5. 1. Regularities of change in the basic technological parameters of wall ceramics depending on the amount of waste in the composition of ceramic masses

The following properties were defined for the obtained samples: density, water absorption by the methods of saturation and boiling in water, tensile strength at compression. The results of studying the specified properties depending on the content of sludge in the mass are shown in Fig. 3–6.

The influence of the amount of sludge on the mean density of ceramic samples is illustrated in Fig. 3.

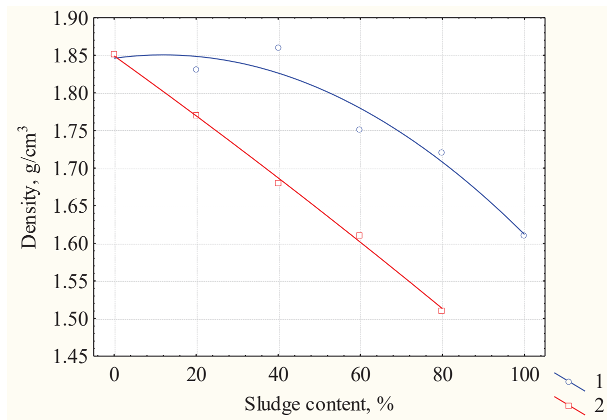

Fig. 3. The impact of sludge amount on the mean density of ceramic samples: 1 – the sample of drilling sludge DS1; 2 – the sample of drilling sludge DS2

The result of the statistical treatment of experimental data is the derived equations of approximation curves, which make it possible to calculate a value of the mean density ρ (g/cm^3) depending on the amount of waste in the ceramic samples x (% by weight):

$$\rho_{DS1}=1.8461+0.0007\cdot x-0.00003\cdot x^2; \quad (1)$$

$$\rho_{DS2}=1.852-0.0042\cdot x. \quad (2)$$

The impact of sludge amount on the water absorption by ceramic samples is shown in Fig. 4.

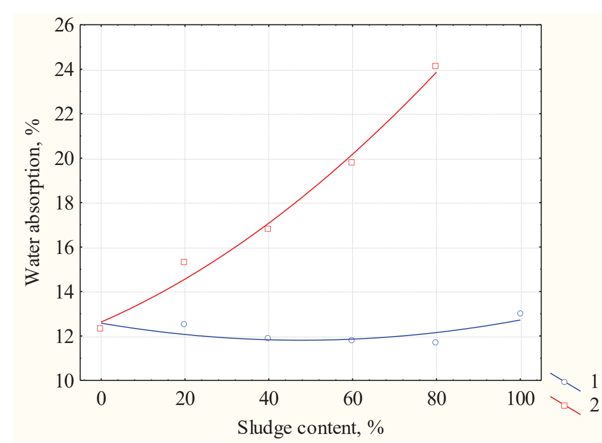

Fig. 4. The effect of sludge amount on the water absorption by ceramic samples: 1 – the sample of drilling sludge DS1; 2 – the sample of drilling sludge DS2

The equations of approximation curves were obtained as a result of the statistical treatment of experimental data, which make it possible to calculate a value of water absorption (%) depending on the amount of waste in ceramic samples x (% by weight):

$$\text{Water absorption}_{DS1}=12.575-0.0321\cdot x+0,0003\cdot x^2; \quad (3)$$

$$\text{Water absorption}_{DS2}=12.6257+0.0819\cdot x+0.0007\cdot x^2. \quad (4)$$

The influence of the amount of sludge on the mechanical strength of ceramic samples is shown in Fig. 5.

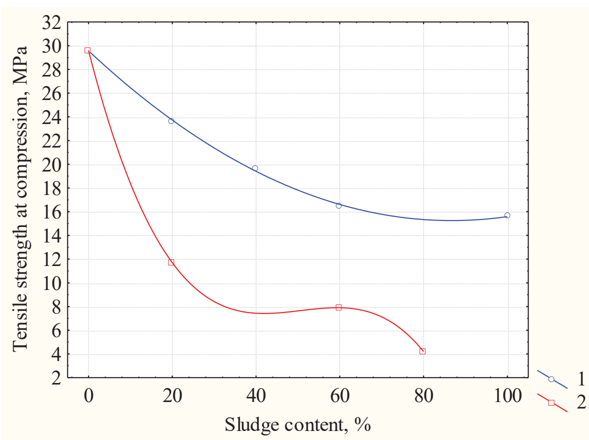

Fig. 5. The influence of the amount of sludge on the mechanical strength of ceramic samples: 1 — the sample of drilling sludge DS1; 2 — the sample of drilling sludge DS2

The result of the statistical treatment of experimental data is the constructed equations of approximation curves, which make it possible to calculate a value of mechanical strength R (MPa) depending on the amount of waste in the ceramic samples (% by weight):

$$R_{DS1}=29.5711-0.3303\cdot x+0.0019\cdot x^2; \quad (5)$$

$$R_{DS2}=29.6-1.3842\cdot x+0.0282\cdot x^2-0.0002\cdot x^3. \quad (6)$$

The influence of the sludge amount on the coefficient of the structure of the ceramic samples is demonstrated in Fig. 6.

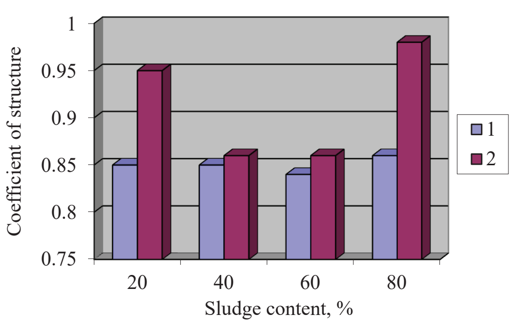

Fig. 6. The impact of sludge amount on the coefficient of the structure of the ceramic samples: 1 — the sample of drilling sludge DS1; 2 — the sample of drilling sludge DS2 1

The revealed regularities (1) to (6) allow us to predict the changes in the samples of wall ceramics depending on the amount of the filler in the form of a given type of waste.

5. 2. The optimal amount of drilling waste in order to obtain the norm-compliant samples of wall ceramics with satisfactory operational characteristics

Our analysis of the results of studying the ceramic samples indicates in general that the use of drilling waste leads to the worsening of the performance of finished ceramic articles relative to pure loam. An increase in the amount of waste in the range from 20 to 100 % leads to a gradual reduction of the density of ceramic articles, the increase in their water absorption and the decrease in the strength of the products.

Fig. 3 shows that both kinds of sludge, at increasing their content in the masses, reduce the mean density of ceramic materials but the degree of such an impact is different for various sludge. Thus, sludge No. 1 at its minimum content (20 %) and maximum content (80 %) reduces the density by 6 % (from 1.83 g/cm^3 to 1.72 g/cm^3). At the similar content in the mass, sludge No. 2 reduces the density by 15 %. The chart in Fig. 3 demonstrates that the lowest density at a value of 1.51 g/cm^3 characterizes the samples containing sludge number 2 in the amount of 80 %. The absence in this chart, as well as in other charts, of a data point concerning the density of pure sludge No. 2 (100 %) is explained by that the sample was unsuitable for measurements as it crushed.

The trends in the changes in the samples' water absorption depending on the amount of sludge in the masses are consistent with the change in the mean density: the greater the mean density, the less the water absorption by the materials (Fig. 4). The level of water absorption by the materials with sludge No. 1 is low (~12 %) and corresponds to water absorption that is acceptable even for face ceramics. On the contrary, even 20 % of sludge No. 2 in the mass leads to a noticeable increase in the water absorption by samples (from 12.3 % to 15.3 %), and so on. The high water absorption by the samples containing sludge No. 2 (60–80 %) is characteristic of the carbonate-containing raw materials, which is logical, taking into consideration the mineral composition of this sludge described in [17].

The mechanical strength of ceramic materials decreases when adding 20 % of sludge, in comparison with pure loam (Fig. 5). In this case, sludge No. 1 does not reduce the strength as much as sludge No. 2. This is due to the close mineral composition of sludge No. 1 to clay raw materials. The overall level of mechanical properties, when adding sludge No. 2, is relatively low; at its content in the mass of 60–80 %, it is generally unacceptable for wall ceramics. When using sludge as the main raw material (more than 60 %), such a function can be performed only by sludge No. 1, using which can enable obtaining wall ceramics of grade M 125–M 175 according to DSTU B V. 2.7-61:2008 (EN 771-1:2003, NEQ). Sludge No. 2 can only be used as an additive (not more than 40 % in the mass) when achieving the grade of the materials within M 75–M 100.

By analyzing the values of the structural coefficient of the samples (Fig. 6), one can note that, based on this indicator, all the materials are considered frost resistant. At first glance, this is illogical, particularly for the samples containing sludge No. 2, given the high water absorption, low density, and, consequently, open porosity of the corresponding samples. The value of this property for the samples containing sludge No. 2 varies within 27–37 %. At the same time, it is known that in terms of frost resistance the safe pores are the large ones, so, most likely, the resulting ceramic samples contain such safe pores. This assumption is evidenced by that the maximum value of the structural coefficient (Fig. 6) is demonstrated by samples containing sludge No. 2, which, due to its mineralogy and inability to sinter, renders the annealed materials greater porosity.

6. Discussion of results of studying the properties of ceramic materials containing the waste from gas extraction

The result of studying the ceramic materials made by using the waste of drilling is the established possibility to obtain materials with the norm-compliant properties. It was experimentally found that by using the clay-like drilling sludge DS1 in the amount of 20 % to 80 % in composition with fusible loam it is possible to obtain the frost-resistant ceramic materials, which, in terms of strength, correspond to grade M 125–M 175. The addition of the high-carbonate sludge DS2 to fusible loam in the amount of 20 % and 40 % makes it possible to obtain the frost-resistant ceramic materials that correspond to grades M 75 and M 100, respectively.

Thus, we have established the possibility to recycle waste in the technology of manufacturing ceramic articles. This trend is promising from a practical point of view but requires further research on the development of drilling waste recycling technology.

The disadvantage of the current study is the use of only two types of drilling sludge from specific drill wells, which does not reveal all the possibilities of using the drilling waste. Given that the waste from a specific well differs in its chemical composition from similar waste from another well, the established dependences and recommendations cannot be used without additional research. However, the methodology and results of studying the ceramic samples, reported in this paper, as well as the procedure for examining the samples of waste, described in earlier work [17], can be used for sludge from other industries or other wells. This is the advantage of our research.

7. Conclusions

1. We have developed the formulations of the raw material compositions using the fusible medium-sintered loam and drilling sludge in the amount of 20–80 % by weight. The main physical and technical properties of the ceramic materials obtained at the annealing temperature of 950 °C have been investigated: they determine that the properties of ceramics comply with applicable standards. We have defined the functions of drilling sludge in the composition of ceramic masses for the manufacture of wall ceramics. It has been established that the drilling sludge with a high content of clay-like substances can perform the function of the primary raw material and mineral additive while the sludge with a high content of psammo-siltstone-pelitic material – the function of disreader and a chromophore additive. We have determined the regularities of change in the characteristics depending on the amount of waste in the composition of a ceramic mass.

2. Sludge No. 1 can be used in the compositions of the masses as an additive and the main raw material for obtaining the frost-resistant ceramic materials at water absorption of ~12 %, and, in terms of mechanical strength, of grade M 125–M 175. Sludge No. 2 can only be used as an additive. The content of this sludge in the mass in the amount of 20 % makes it possible to receive a frost-resistant ceramic material at water absorption of ~15.3 %, and of grade M 75. The same sludge in the amount of 40 % makes it possible to obtain materials at water absorption of 16.8 % and of grade M 100 with an additional function of illuminating the red-burned clay-like raw materials.

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