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STUDY OF THE PROPERTIES OF OVERBURDENED ROCKS FROM COAL MINING: OVERBURDEN – AS A RAW MATERIAL IN THE PRODUCTION OF CERAMIC BRICKS

*Olga Vyshar[1], Andrey Stolboushkin[2], Galiya Rakhimova[1], Victor Stanevich[3] and Murat Rakhimov[1]

[1]Department of Construction Materials and Technologies, Karaganda Technical University, Kazakhstan; [2]Department of Engineering Constructions, Building Technologies and Materials, Siberian State Industrial University, Russia; [3]Faculty Engineering, Toraigyrov University, Kazakhstan

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ABSTRACT: With the development of industry, the amount of man-made waste is growing rapidly. It is necessary to develop energy-efficient resource-saving technologies for use in the processing of. Research in past years has proved the possibility of using coal industry wastes as burning and baking additives in the production of ceramic products. The purpose of this work is to review the properties and discuss the options for using overburden as the main raw material in the production of ceramic bricks. Overburden requires a small amount of organic and chemical additives to adjust its technological properties. This research is due to the need to solve environmental, economic and technological problems in the utilization of overburdened rocks from coal mining. The physical and mechanical properties as well as chemical and mineralogical compositions of overburden have been investigated. Due to the heterogeneity and non-wettability of raw materials in water, it is necessary to grind them. The structural-mechanical characteristics and molding properties using highly mineralized carbonaceous rocks as a solid additive and aluminum oxychloride as a liquid additive have been studied. The obtained products with the use of additives have higher indicators: compressive strength 33.6-48.2 MPa, frost resistance 50-100 cycles, and water absorption 7.0-10.8. Thus, the results of the research prove the possibility of producing high-quality ceramic bricks from overburden, which will help to significantly reduce the cost of production, compensate for the lack of high-grade clay raw materials, and improve the environmental situation. Nevertheless, further research and technological developments are needed.

Keywords: Overburden, Radiation Safety, Chemical Composition, Plastic Strength, Ceramic Brick.

1. INTRODUCTION

There is a continuous and rapid increase in the number of various waste products in the world [1,2]. A World Bank report states that by 2050, the total volume of waste will increase to 3.40 billion tons per year. More than 33% of waste is improperly disposed of, and only 13.5% of waste is recycled worldwide [3]. Therefore, it is necessary to constantly look for ways to solve this problem through proper disposal and recycling of waste generated. One of the ways of utilization of solid wastes in the fuel and energy complex is their use as construction materials or raw materials for their production [4].

Ceramic bricks are a durable and relatively inexpensive building material with high architectural and decorative properties [5]. The reserves of the high-quality clay used in factories for the production of ceramic products (bricks, tiles, pipes, tiles) are quickly depleted [6,7]. For sustainable development of the production of ceramic products, it is necessary to look for ways of technological transition to new types of raw materials - low plastic loams, low-melting clays with high impurities, as well as aluminosilicate man-made production wastes [8,9]. The experience of foreign studies, in particular from Western Europe, Japan, and other countries, shows that the production of ceramic bricks from man-made raw materials is cost-effective and environmentally appropriate [10]. Since there is no need to extract and process the conditioned clay raw materials when it is possible to replace them with production wastes suitable for these purposes [11,12]. Past studies have proved the possibility of using the waste coal industry as a burnout and baking additive in the production of ceramic bricks [13,14].

At the world's largest deposit Ekibastuz (Kazakhstan), during open-pit coal mining a huge amount of waste overburden is formed - overburden rock, which is removed to open dumps. During the operation, about 4 billion m^3 of overburdened rocks from coal mining have been accumulated. Dumps of overburdened rocks are located near the city limits, occupy large areas, and are up to 100 m high. They significantly worsen the environmental situation in the region and are a source of dust storms, self-combustion, and gas pollution. Overburden rocks are represented mainly by sedimentary rocks - clays, argillites, siltstones, loams, sands, gravels, conglomerates, and so on.

When studying the composition and properties of coal mines rocks, it was found that claystones and siltstones prevail in the main mass compared to other types of rocks. Argillites and siltstones are mainly represented by clay minerals - kaolinite, hydromica, and montmorillonite [15]. Claystones and siltstones are heterogeneous and non-dissolvable in water, so it is necessary to grind them further.

The aim of the work is a comprehensive study of overburden rocks of coal mining as the main raw materials. A small amount of organic and chemical additives is used to adjust the technological properties and improve the quality of the finished products. Determination of environmental safety, chemical, mineralogical composition and other properties of raw materials. Development of mass compositions and technology for the production of ceramic bricks.

The novelty of this work and the difference from previous works is the study of homogeneity, structural and mechanical characteristics, and molding properties of the masses of overburdened rocks. Selection of solid and liquid additives that improve the structural formation of products during shaping, drying, and firing. Development of the optimal fractional composition of the mass and resource-saving production technology. Determination of physical and mechanical parameters of the obtained ceramic products.

2. RESEARCH SIGNIFICANCE

Overburden rocks from coal mining are rarely used because the composition and technological properties are poorly studied, and there are no methods of evaluation and scientifically proven processing technology [16]. Now very advanced technology of using carbon wastes as additives in the production of ceramic products. [17,18]. The significance lies a comprehensive study of overburden is the main raw material. Selected additives, and developed mass compositions and technology for the production of ceramic bricks. Studies are of practical value for the coal industry and enterprises for the production of ceramics, which will reduce production costs and improve product quality.

3. RESEARCH MATERIALS AND METHODS

3.1. Raw Materials

The physical and mechanical properties were researched. The chemical and mineralogical composition of overburdened rocks have been studied. The similarity with the traditional clay raw materials used for the production of building ceramics is found.

Overburdened rocks have certain physical and mechanical properties that depend on the degree of their metamorphism. In their natural form are not soaked in water, which requires their mechanical grinding to break the cementation bonds of the clay components [18].

The quality of the finished ceramic products obtained from coal mining waste is greatly influenced by the physical and mechanical properties and chemical and mineralogical composition of the overburden [19]. Analyzing the data of chemical analysis of overburdened rocks of different lithological types, we can judge about the quantitative content of rock-forming oxides. The content of $Si0_2$ - silicon oxide is 56.7-61.3%. It is inbound and free states. Bound silica is part of the clay-forming minerals, free are represented by impurities of fine quartz, its content in the samples is 16-25%. In terms of free quartz content, coal mining wastes belong to the group of raw materials with average quartz content.

The clay-forming minerals and mica impurities include $A1_20_3$ - aluminum oxide. Its content for claystones is 17.6-18.4%, for siltstones 17.6-18.5%, decreasing for claystones depending on the horizon from +50 to +150 m. According to the content of aluminum in the calcined state, overburdened rocks belong to the group of semi-acidic raw materials. The content of iron oxides in the studied samples is 5.6-6.4% for claystones and 3.4-4.18% for siltstones. Iron compounds are represented by pyrite and siderite. Overburdened rocks in terms of iron oxide content belong to the group of raw materials with a high content of dyeing oxides.

Alkali earth metal oxides are part of clay minerals and carbonates. The total content of calcium and magnesium oxides is 1.67-2.3% for the claystones and 1.09-1.84% for the siltstones. The total content of sodium and potassium oxides in the lithological types of the different bedding horizons ranges from 2.96 to 3.36%. The alkaline oxides Na_2O, and K_2O are part of the clay-forming minerals and are also present in impurities in the form of water-soluble salts.

$S0_3$ - sulfur oxide content does not exceed 0.28%, this is typical for low-sulfur environmentally friendly raw materials and allows the use of overburden in the production of ceramic products without restrictions. In addition, overburdened rocks contain organic carbon. According to the mineralogy of the clay component, the overburdened rocks belong to the kaolinite-hydrosludite type. Among the non-clay minerals, the studied samples contain admixtures of quartz, feldspar, mixed-layer minerals, as well as organic matter. They can be used as a basic raw material for the production of ceramic building materials.

The main physical and mechanical properties of overburdened rocks are density, natural humidity, compressive strength, and porosity (Table 1). The analysis of the obtained data on density shows that it decreases from the horizon +50 to the horizon +150 m. The density of siltstones is 7-10% higher than that of claystone and amounts to 2.3-2.7 g/cm^3 for claystone. The natural moisture content of claystones varies from 4.9-5.8%, siltstones 4.3-4.7%, and increases from the +50 to +150 m horizon.

The compressive strength of overburdened rocks is 34.7-37.4 MPa for claystones, 45.0-48.9 MPa for siltstones. When the bedding horizon changes from +50 to +150 m, the compressive strength of siltstones and claystones decreases.

The porosity of claystones is 12.9-18.7%, siltstones 10.6-19.1%, and increases depending on the horizon.

Table 1 Physical and mechanical properties of overburdened rocks

Name raw materials	Density, g/cm^3	Natural humidity	Compressive strength, MPa	Porosity, %
Horizon +50				
claystone	2.4	5.1	37.4	12.9
siltstone	2.7	4.3	48.9	10.6
Horizon +100				
claystone	2.4	4.9	34.9	14.9
siltstone	2.5	4.3	47.0	15.9
Horizon +150				
claystone	2.2	5.8	34.7	18.7
siltstone	2.3	4.7	45.0	19.1

Thus, when changing overburden horizons from +50 m to +150 m indexes of physical and mechanical properties decrease, which is explained by a decrease in the degree of compaction of rocks when you change horizons their more significant weathering at the horizon +150 m.

According to the mineralogy of the clay component, the overburden rocks belong to the kaolinite-hydrosludite type of raw material. Of the nonclay minerals, the samples studied contain impurities of quartz, feldspar, mixed minerals, as well as organic matter. [20,21].

In general, according to the chemical and mineralogical composition, overburdened rocks belong to the group semi-acidic with low and medium content of coloring oxides of raw materials. The chemical composition of the mineral part of the samples is close to the chemical composition of clays used for the production of ceramic products.

3.2 Research Methods

Modern optical, spectrometric, thermogravimetric, X-ray-phase, and other types of research are used to study the overburdened rocks of the Ekibastuz coal basin [22,23]. Modern methods and equipment from accredited, certified laboratories were used. All indicators of raw materials, structural and mechanical characteristics and molding properties of masses of overburden, and physical and mechanical properties of the resulting ceramic products by the current regulatory documents determined.

4. RESULTS AND DISCUSSION

4.1 Checking the Safety and Suitability of the Raw Materials

According to the results of the research, the radioactive safety of rocks as raw materials for the production of building materials was assessed. Specific activities of natural radionuclides were determined (Table 2) [24,25].

Analysis of the data obtained showed that overburdened rocks, by the standards of radiation safety, can be used without restrictions for the production of all types of building materials.

Table 2 Results of determination of specific activities of natural radionuclides

Name raw materials	Name of natural radionuclides, Specific activity A, Bk/kg			Effective specific activity, A_{eff},Bk/kg
	radium, ^{226}Ra	thorium, ^{232}Th	potassium, ^{40}K	
Horizon +50				
claystone	37	52	618	158
siltstone	33	50	604	151
Horizon +100				
claystone	28	39	712	178
siltstone	24	36	703	165
Horizon +150				
claystone	29	36	685	182
siltstone	27	34	672	173

The content of rare-earth and noble elements in overburdened rocks is relatively low and of no value for their industrial extraction. The content of hazardous elements (lead, chromium, beryllium, arsenic, antimony, gallium, mercury, etc.) is below the maximum allowable concentrations and generally positions the rocks as relatively environmentally safe raw materials. The data obtained were compared with the contents to be quantified [26]. Determination of the amount and composition of water-soluble salts by the water-extraction method showed that the content of salts in overburdened rocks is small, characterized by a high degree of homogeneity, and does not require special technological measures for their decontamination.

Petrographic studies of the studied rocks showed that they are mainly composed of clay minerals: kaolinite and hydromica, clastic rocks in the form of quartz and feldspars, iron minerals, and carbonate inclusions. All samples contain organic matter in the form of elongated clumps of irregular shape, the number of inclusions of organic matter ranges from 5 to 10% for siltstones and from 10 to 12% for claystones. Thin strips of organic matter are observed in the rocks.

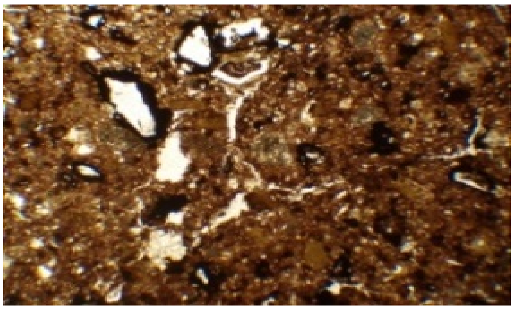
Claystone of the +50 m horizon has a pelitic structure. Grain size ranges from 0.01 to 0.001 mm, and the shape of the grains is isometric, less often irregularly elongated, which is characteristic of organic residues. The texture of claystone is thinly band due to changes in the concentration of carbonate grains and the orientation of elongated particles of organic matter.

In the rock (Fig. 1) observed calcite in the form of colorless grains of isometric shape ranging in size from 0.003 to 0.008 mm with high birefringence. There is a minor content of colorless volcanic glass with negative relief. Colorless quartz with low positive relief and birefringence is observed, as evidenced by gray interference colors. Argillite contains organic matter in the form of black concretions, the number of grains varies from layer to layer from 80 to 90%. The clay component is kaolinite, less frequently hydrosludite-kaolinite, with kaolinite grains up to 0.005 mm in size. The siltstone of the +50 m horizon has a siltstone structure with a grain size from 0.1 to 0.005 mm with a predominance of 0.07-0.01 mm grains. The rock contains quartz, strongly corroded with direct extinguishing. Feldspar grains are highly altered, many completely transformed into hydromica with preservation of the primary form of feldspar grains. Mica is represented by elongated, well-formed grains with direct extinguishing and bright greenish-blue interference colors. Observed organic matter in the form of elongated irregularly shaped shrinkage of a brownish color. The clayey part of the siltstone is represented by hydromica and kaolinite minerals.

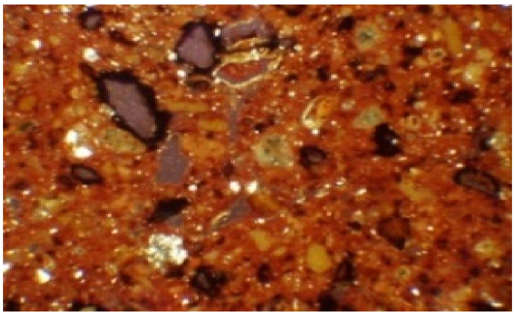
Claystone of the +100 k horizon has a pelitic structure with grain size less than 0.005 mm there are rare quartz grains of up to 0.05 mm. The thinly banded structure of the rock is due to the striped layer-by-layer brown coloration. Hydromica aggregates up to 0.005 mm in size with low positive relief red-yellow interference coloring are observed in the rock. The shape of hydromica grains is isometric, or weakly elongated. Siltstone structure +100 m siltstone, grain size from 0.1 to 0.01 mm. Grains with a size of 0.05-0.01 mm prevail. The texture is striated due to the orientation of organic matter. There are strongly altered feldspar and plagioclase grains, many grains are completely replaced by mica and sometimes even kaolinite with preservation of the primary grain shape. Micrograin carbonates are present in small quantities. Organic matter in the form of strongly elongated irregularly shaped concretions is marked (Fig. 1).

The claystone of the +150 m horizon has a pelitic structure with a grain size of 0.005-0.001 mm and a thin layered texture. Hydromica in the form of isometric grains up to 0.003 mm in size with low positive relief and red-yellow, less often greenish-blue interference colors are observed. Kaolinite is presented as isometric grains up to 0.005 mm in size, colorless with low positive relief. Observed organic matter as opaque, black, elongated particles up to 0.01 mm in size.

The siltstone of the +150 m horizon has a siltstone structure of 0.05 to 0.1 mm grains and a layered texture. Represented by strongly corroded quartz, feldspar grains, and plagioclases, which are heavily altered to complete replacement by hydromica. Individual particles and aggregates of hydromica are identical to feldspar. The organic matter of siltstone is brownish-brown in the form of strongly elongated formations. Clay component of the basal type kaolinite-hydrosludite.



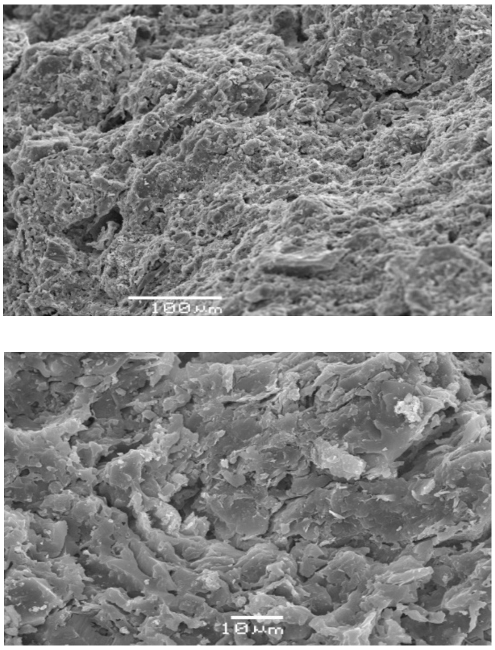
a



b

Fig. 1 Micrographs of the structure of burnt claystones. Slip, transmitted light: 25□: nicoly II (*a*); nicoly + (*b*)

Individual particles and aggregates of hydromica are identical to feldspar. The organic matter of siltstone is brownish-brown in the form of strongly elongated formations. Clay component of the basal type kaolinite-hydrosludite. Specific areas of the overburdened microstructure were studied scanning electron microscope (Fig.2).



a

b

Fig. 2 Microphotographs of the claystone structure of the +100 m horizon, magnification: 150□ (*a*); 750□ (*b*), scanning electron microscope

The microtexture of claystone is uniform, with individual larger aggregates of irregular shape, mostly 20-50 microns in size. In addition to X-ray diffractometry by international crystallographic and crystal-chemical databases on minerals and their structural analogs, infrared absorption spectra of overburdened claystones were studied (Fig. 3).

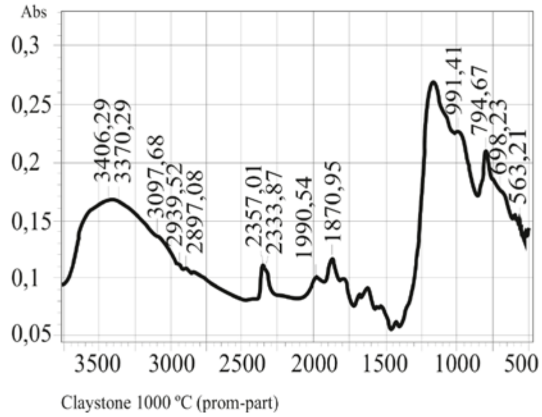


Fig. 3 Infrared absorption spectra of overburdened claystone at 1000 ºC

Absorption bands characteristic of quartz, feldspar, and hematite are fixed in the low- and medium-frequency regions of the spectrum. The 563, 1170 cm^{-1} peaks identify the pelite structure.

4.2 Technological Design of Compositions

One of the most important technological operations in the production of ceramic bricks is the preparation of masses and the formation of. At the stage of formation, a coagulation structure is formed, which has certain technological and structural-mechanical properties, which ultimately determine the quality of the finished products [27,28].

Overburden rocks in terms of their particle size distribution are medium-dispersed raw materials, as the medium-dispersed fraction in claystones is 45.5-53.2%, and in siltstones - 27.0-35.2%. Analyzing the granulometric composition of overburdened rocks based on "lithological type", it should be noted that in claystones the content of clay particles by 2-8% more than in siltstones, and in siltstones prevails sandy fraction, the content of which is 19-26% higher than in claystones.

The predominance of medium- and coarse-dispersed fractions in claystones allows them to be classified as dusty clays and siltstones as dusty loams, since the quantitative content of fractions from 5 to 1000 microns in them is 78.5-87.5%. The content of fine and medium-dispersed fractions decreases for claystones and siltstones with a change in horizons from +50 to +150 m, and the content of the coarse-dispersed fraction increase. This confirms the data of diffractometric and petrographic analyses on the reduction of clay minerals, and an increase in the content of terrigenous materials - quartz, siderite and feldspars when changing overburden horizons from the roof of the coal seam.

A series of samples-cylinders with a diameter of 50 mm and a height of 45-55 mm were molded, taking into account the granulometric composition of coal waste based on the previously selected optimal fractional composition of overburden rocks of different lithological types and horizons of occurrence. Optimal fractional compositions of ceramic charge materials are shown in Table 3.

Table 3 Optimal fractional composition of overburden rocks of coal mining

Name raw materials	Content of overburden, in % for fraction size, mm		
	0.5-0.25	0.25-0.125	0.125-0.0063
claystone	5-45	10-55	15-50
siltstone	10-55	15-50	15-45

The molded samples were kept in natural conditions for 24 hours and then dried in the desiccator for 24 hours at a maximum temperature of 90±5 ° C. After drying, samples were inspected and all changes in appearance were recorded, and air shrinkage and other characteristics were determined. Firing was performed in a laboratory furnace with automatic control of the firing mode at an average temperature rise rate of 1-3 degrees/min and holding at the maximum temperature of 1.5-2 hours according to the established mode at a maximum temperature of 1000 °C.

4.3 Use of Solid and Liquid Additives

Control the structure and properties of the masses in the process of their processing by introducing various additives, which contribute to changing the structural and mechanical properties of the masses in a wide range. The authors [29] found that a positive effect on the molding and structural-mechanical properties of the masses has organic impurities and plasticizing additives.

The selection of optimal fractional compositions of overburdened rocks has achieved some improvement in structural and mechanical properties. But the values of elasticity, true relaxation period, and elasticity have not reached the values characteristic of well-formed masses, although in the process of deformation slow elastic deformations predominated.

Given the absence of difficulties in mixing with overburden rocks, the effect of high-mineralized carbonaceous rocks and high-base technical aluminum oxychloride on the properties of molding masses and the quality of finished products was studied. Highly mineralized carbonaceous rocks were previously ground to a fraction of less than 0.125 mm and introduced into the mixtures in an amount of 4-12% (compositions 1-3, Table 4). The mechanism of the impact of carbonaceous rocks on the rheological properties of the masses is that the resin and bituminous substances in these rocks cause coagulation phenomena in the masses and provide a homogeneous and more plastic mass due to the ability of the clay components of siltstone-argillite rocks to emulsify the "tar-water" and "bitumen-water" systems as a result of wetting and sticking.

The introduction of carbonaceous rocks containing resin and bituminous substances has reduced the plastic viscosity of the mass by 6-13%, which improved the mobility of the mass, reduced the plastic strength by 18-23%, which led to a decrease in rigidity and had a favorable impact on the moldability of the mass. A characteristic feature of the impact of carbonaceous rocks on the structural-mechanical properties of the masses is their transition from the third to the first structural-mechanical type due to a significant reduction of plastic deformation and the predominance of slow elastic deformation (40.32-43.14%), which provides good formability of the masses.

The use of highly mineralized carbonaceous rocks allowed for to improve of the molding properties of the masses, reduced water absorption of products, as well as to increase their physical and mechanical properties (Table 5). As a liquid additive to improve the structural and mechanical properties of the mass of overburden rocks used highly basic technical aluminum oxychloride, which is obtained by dissolving in water sludge production of anhydrous aluminum chloride with subsequent neutralization by hydrochloric acid, formed in solution due to hydrolysis of the latter, the interaction with a metal oxidant. The composition of aluminum oxychloride corresponds to the formula $[Al(CH)_{3-X}Cl_X]_n$. Raw mixes of overburdened rocks crushed to fractions less than 0.5 mm were moistened with water mixed with aluminum oxychloride in an amount of 3-7% to the normal molding moisture (Table 4).

Table 4 Compositions of raw mixes containing solid and liquid additives

Components	Content of components in the mixture, masses, %					
	1	2	3	4	5	6
Overburden rock of coal mining	96	92	90	97	95	93
Coal-bearing rocks with tar and bituminous substances of 3-9%	4	8	12			
Highly basic technical aluminum oxychloride				3	5	7

When aluminum oxychloride introduced due to ionic exchange, clay particles of overburden rocks dispersed under the influence of OH-. Coagulation adhesion of particles occurs as a result of overcoming energy barriers created by sulfate shells, coagulation structures formed with predominant development of slow elastic deformations, which are characterized by fracture of particles without destruction of contacts between them.

When studying the effect of aluminum oxychloride on the structural and mechanical properties of overburden masses, it was found that the mechanism of the aluminum oxychloride effect is identical for all lithological rock types and does not depend on the depth of their occurrence.

Aluminum oxychloride helps to improve the molding and rheological properties of the masses by reducing their plastic viscosity by 40% and plastic strength by 20-25%. The plasticity indicators of these masses refer to the values typical for well-formed masses (Table 5).

Determination of physical and mechanical properties of the obtained products shows that the use of carbonaceous rocks as solid additives provides an increase in strength parameters of the products 2-2,5 times, a decrease in water absorption by 5-6%, and an increase in frost resistance by 20-25 cycles.

The effect of aluminum oxychloride on the physical and mechanical properties of the obtained products is manifested in the reduction of water absorption from 8.2 to 7.0%, increasing the compressive strength by 10-12%.

Table 5 Results of physical and mechanical tests

№	Component content by weight, %			Compressive strength, MPa	Water absorption, %	Frost resistance, cycle
	Overburden	carbonaceous rock	Aluminum oxychloride			
1	96	4		42.0	10.8	58
2	92	8		48.2	9,2	72
3	88	12		44.0	9.6	64
4	97		3	34.2	8.2	62
5	95		5	33.6	7.4	63
6	93		7	36.4	7.0	68

Products containing aluminum oxychloride can withstand more than 62 cycles of alternate freezing and thawing and have a good appearance.

5. CONCLUSION

As a result of studies overburden rocks of coal mines as raw materials for the production of ceramic bricks established:

- Rocks do not get soaked in water and exhibit the ability to plastic formation when cementation bonds are broken by grinding to a fraction of less than 0.5 mm.
- According to the standards of radiation safety can be used for the production of all types of building materials without restrictions, the number of potentially toxic elements in the waste does not exceed the maximum allowable concentrations, which characterizes them as environmentally safe raw materials.
- Physical and mechanical properties of coal mining wastes change in the direction of decreasing from the +50m horizon to the +150m horizon, which is explained by a decrease in the degree of compaction and more significant weathering.
- By chemical and mineralogical composition and content of water-soluble salts, coal mining wastes are close to clayey raw materials and belong to the group of semi-acidic with low content of dye oxides of raw materials.
- Structural and mechanical properties of rocks of different horizons are most optimal in limited areas of fractional compositions: 0.5-0.25 mm - 10-50%; 0.25-0.125 mm - 5-55%; 0.125-0.063 mm - 20-65%.
- Improving the structural and mechanical characteristics of masses from overburdened coal mining by using highly mineralized coal rocks and aluminum oxychloride as organic and plasticizing additives achieved.
- Products obtained with the use of additives have a compressive strength of 33.6-48.2 MPa, frost resistance of 50-100 cycles and water absorption of 7.0-10.8%.

Based on the above, we can conclude that waste coal mining is close to the traditional clay raw materials in the physical and mechanical properties, chemical and mineral composition. Appropriate technological preparation and the introduction of additives can be used for the production of ceramic bricks by a plastic method of molding.

In further research, it may be necessary to study other complex additives and use the technology of semi-dry forming ceramic bricks.

6. ACKNOWLEDGMENTS

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