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THE INFLUENCE OF ANNEALING TEMPERATURE ON VARIATION OF PHYSICOMECHANICAL PROPERTIES OF WALL CERAMIC TILES BASED ON LOESS LOAM MODIFIED BY OIL SLUDGE

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ABSTRACT

This work is aimed at investigation into oil sludges and their application in the composition of ceramic pastes as energy-efficient and modifying component for production of ceramic bricks. The article discusses the results of experimental study of oil sludges. Innovative engineering approaches to conversion of oil sludge into organomineral conglomerate state have been developed. It has been established that addition of organomineral conglomerate mixture with oil sludge converts ceramic paste based on loess loam from high-sensitive matters into low-sensitive ones. Combustion of oil sludge in the composition of organomineral conglomerate mixture makes it possible to increase temperature in furnace and accelerates baking of ceramic bodies. This is evidenced by reduction in baking time of samples from 9 to 7.5 hours. This process provides decrease in energy consumption at the stage of article baking. Moreover, the weight of final product decreases by 15-20 %, which simplifies labor procedures and increases labor efficiency upon brick laying. Herewith, the load on building foundation decreases, thus reducing material consumption upon construction by nearly 10-12%.

Keywords: annealing temperatures; ceramics; oil sludge.

1. INTRODUCTION

Demand in energy resources is continuously increasing. According to expert appraisal from early 1980-s up till now, the global energy consumption increased by about two times.

Strategy of many countries aimed at improvement of energy efficiency promoted increase in competitiveness of economy and industry, development of science, innovations, implementation of new technologies.

Researchers and experts of many countries on the basis of economic predictions have concluded that it is more efficient to invest into energy efficiency and to gain profits by savings with respect to energy resources than to create new power facilities. Hence, energy efficiency is an "invisible fuel" at reasonable cost [1].

This aim is supported by provision of legal, organizational, scientific, engineering and economic measures aimed at efficient and rational use of fuel and power resources and renewable energy sources.

Energy saving in industrial sector means achievement of the same economic result but at lower power consumption or higher results at the same or lower power consumption per unit product.

It means decrease in power consumption together with cash saving [1].

Thus, researchers continue intensive studies aiming at decrease in power consumption for production process.

As a rule, such projects are implemented successfully on commercial scale, since companies obtain possibility not only to retrofit technology but to compete in global markets with regard to cost and quality of ready products [2-4].

Oil sludge is attractive for production of construction materials in terms of improvement of energy efficiency.

Oil sludge is generated during oil and gas production, transportation of oil and petroleum products, storage in reservoirs and during their processing. Amounts of oil sludge are continuously increasing. National and foreign oil producing companies are not interested in its usage due to numerous external and internal reasons. Implementation of deep conversion of oil sludge is one of these reasons.

However, in order to solve this problem, the companies should arrange new departments and construct required refinancing facilities.

This approach requires for supplemental time consumption and financial expenses including training of experts and managers to support advanced refining of oil sludge.

Therefore, nowadays oil sludge is considered as waste by numerous oil producing companies which transfer this product to other companies specializing in disposal of oil sludge at landfill sites.

Combustion of oil sludge using specialized facilities is used quite often since oil sludge is a highly flammable material [5].

Oil sludge contains valuable hydrocarbons, such as petroleum, paraffin, resins, and asphaltenes.

Hence, oil sludge should be considered not as waste but as valuable energy containing and additional modifying agent for other industries [6].

Since advanced research regarding application of oil sludge as energy containing and modifying agent is not sufficient, storage and combustion of oil sludge are accompanied by severe environmental problems [7, 8].

2. LITERATURE REVIEW

Disposal of oil sludge is considered by foreign researchers [9-11]. The researchers of Vilnius Technical University (Lithuania) carried out R&D studies on application of oil sludge in ceramic materials. The obtained results proved to be successful for application of oil sludge in ceramic production in terms of modification of ceramic pastes and improvement of physicomechanical properties of final products.

The researchers [12] studied application of spent automobile oils (petroleum product wastes) in combination with quartz sand for production of light weight fillers as modifying agent of clay feedstock. Addition of 1% of spent oils in combination with quartz sand increased gas generation, bloating of clay feedstock, improved mechanical strength and decreased bloating temperature.

Application of oil sludge as feedstock for other industries is an advantageous method since it provides obvious environmental and economic result [13].

Production of construction materials is an important field in any national economy. Companies producing construction materials apply high temperature annealing involving high consumption of power sources (coal, gas, diesel fuel, etc.).

This procedure is used in production of ceramic brick, expanded clay aggregate, aggloporite, lime, cement [2-4].

The plants producing ceramic brick in Kazakhstan encounter the following difficulties:

- unavailability and constrained resources of high-quality clayey feedstock for brick production;
- as a consequence of unavailability of high-quality feedstock, producers of ceramic brick work with low quality feedstock: loess loam;
- final product based on loess loam is characterized by low strength, water and frost resistance;
- chemical and mineralogical composition of loess does not allow to produce high quality ceramic bricks even at high annealing temperatures (1,050–1,100°C);
- production of ceramic bricks on the basis of loess loam is characterized by high power consumptions because it requires high annealing temperature in commercial furnace (1,050-1,100°C) and long time (up to 72 h) in order to provide complete annealing of ceramic brick.

Thus, the aim of this work was to study oil sludge and possibility of its application in ceramic pastes as energy containing and modifying agent for production of high-quality ceramic bricks.

The following tasks should be performed in order to achieve the specified target:

- analysis of properties and chemical mineralogical composition of loess loam of Chagan deposit;
- analysis of chemical and rheological properties of oil sludge regarding quantitative detection of hydrocarbons and other compounds capable to improve molding, drying, and annealing properties of loess loam;
- determination of heat of combustion of oil sludge promoting release of additional energy due to combustion in ceramic paste based on loess loam;
- development of engineering approaches to conversion of oil sludge into conglomerate state for convenient addition into ceramic paste based on loess loam;
- investigation into properties of ceramic pastes and thermally treated samples based on loess loam in combination with oil sludge converted into conglomerate state.

3. METHODS

Hydrocarbon composition of oil sludge was analyzed using an Agilent 7890A/5975C gas chromatograph (US).

Fractional composition of the considered oil sludge was determined using an ARN-LAB-02 instrument by distillation of 100 ml of tested sample with continuous recording of temperature and amounts of condensate.

Weight content of sulfur in the considered oil sludge was determined using a Spektroskan-Maks GF2E instrument (Russia) by measurement of intensity of fluorescent radiation of sulfur induced by X-ray radiation.

Heat of combustion of tested sample was determined using a C2000 calorimeter (IKA-Werke, Germany) upon complete combustion of pre-weighted paste in calorimetric bomb in compressed oxygen environment.

Content of mechanical impurities was determined by filtration of tested oil sludge with preliminary dissolution in petrol and washing of sediment on filter by solvent with subsequent drying and weighing.

a) Content of chlorides was determined by titration of water extract with bivalent mercuric nitrate in the presence of diphenylcarbazide.
b) Density of the considered oil sludge was determined using hydrometer submersed into tested product and converting the instrument readings to density at 20°C.

Crystalline phases, particle size distribution, quantitative ratio of crystalline to amorphous phases were determined using a JEM-2100 transmission microscope (JEOL, Japan).

Topography and surface microstructure, qualitative and quantitative analysis of sample in point area, plotting elemental distribution along selected line, elemental distribution map from a selected area were obtained using a JSM-6390LV scanning electron microscope (LEOL, Japan)

Qualitative and quantitative phase analysis, determination of cell parameters and crystal orientation, analysis of polycrystalline structures, stresses and textures, quantitative elemental analysis of inorganic substances, isotope analysis were obtained using an XPert PRO X-ray diffractometer (PANalytical).

Laboratory tests were performed with loess loam crushed in a MShL 100×250 jaw crusher to particle sizes of 5-10 mm with subsequent milling in a MShL-1P ball mill to complete passing sieve with 1 mm mesh.

4. RESULTS AND DISCUSSIONS

The subject of research was oil sludge of Zhaiyk Munai oil producing company (Uralsk, Kazakhstan). Chromatography was performed according to the procedure described in [14]. The experimental results are illustrated in Figure-1.

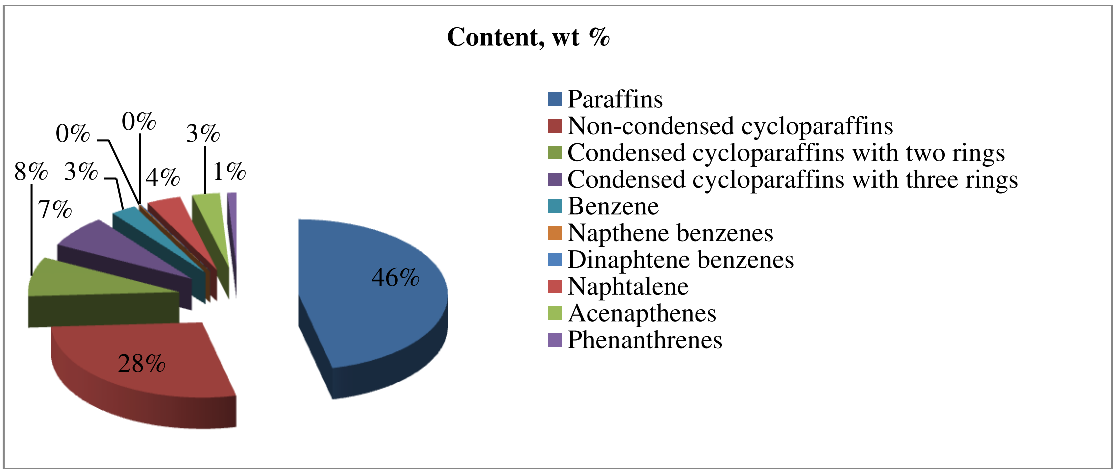

Figure-1. Hydrocarbon content determined by chromatography-mass spectrometry.

Rheological properties, density, particle size distribution, sulfur mass fraction, heat of combustion, content of mechanical impurities and chlorides of the considered oil sludge were determined using appropriate procedures [15]. These properties of oil sludge are summarized in Table-1.

Table-1. Properties of oil sludge.

Property	Value
Density, kg/m^3 at 20°C	836.4
Fractional composition, vol %	
200°C	11
300°C	39
350°C	54
Mass fraction of sulfur, %	0.024
Heat of combustion, kJ/g	44.987
Content of mechanical impurities, %	0.027
Content of chlorides, mg/dm^3	28.46

The oil sludge of Zhaiyk Munai oil producing company contains up to 97% of hydrocarbons and is characterized by significant heat of combustion. The heat of combustion of the considered oil sludge is 44.987 kJ/g equaling to that of coal.

Loess loam of Chagan deposit (West Kazakhstan) was used as clayey constituent, its chemical composition is illustrated in Figure-2.

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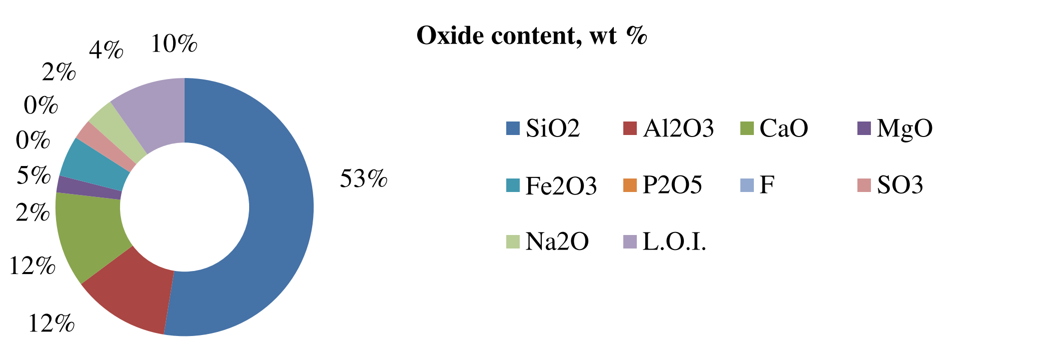

Figure-2. Chemical composition of loess loam.

Addition of oil sludge to ceramic paste should be substantiated since clayey feedstock is an inorganic silicate system, and the oil sludge being hydrocarbon feedstock is an organic system.

According to Korenkova and Shein [16], oil sludge is a petroleum disperse system of colloidal origin. It is a low concentrated suspension comprised of solid mineral particles with adsorbed high molecular organic compounds. It is the dispersed phase, and water-petroleum oil emulsion is the dispersion medium. In such state, minor additions of oil sludge to ceramic paste result in following consequences:

- clayey paste is instantly clogged immediately after contact with oil sludge since porous macrostructure of loess loam is impregnated upon their contact;
- clogging occurs only in points of contact, which prevents uniform distribution of oil sludge in ceramic paste;
- it is impossible to add oil sludge into ceramic paste without new approaches to production of ceramic brick;
- it is required to solve the problem of addition of oil sludge into ceramic paste based on loess loam providing uniform distribution of fine particles;
- it is required to provide production conditions promoting energy efficiency and improved quality of final products.

The specified target could be achieved by conversion of oil sludge from suspension to loose state, thus achieving loose state of loess loam. Herewith, it is necessary to achieve and to retain maximum concentration of oil sludge in the composition for further use in ceramic pastes.

With this aim, loess loam was dried in drying cabinet at 100-120°C to residual moisture content of 5-7%.

Then the dried loess loam was milled in a ball mill to complete passing 1 mm sieve meshes.

During storage in a reservoir, oil sludge is a suspension, and in this state, it is conditionally subdivided into three layers. The top layer of oil sludge is comprised of light fraction and heavier fractions are in the middle and bottom layers.

In order to provide uniform distribution of oil sludge in all three fractions in ceramic paste, they were averaged by intensive agitation in a propeller stirrer.

After averaging, the density of oil sludge was 820-850 kg/m^3.

Then the oil sludge: loess loam mixture was prepared by volumetric batching at the ratio of 1:5 and thoroughly mixed in laboratory mixer. The obtained composition is a loose organomineral conglomerate with average density of 1,000-1,100 kg/m^3.

In this state, the organomineral conglomerate based on fine loess loam is the dispersion medium, and oil sludge suspension is the dispersed phase.

This procedure converts oil sludge from suspension to loose organomineral conglomerate and provides background for subsequent process stages such as metering and uniform distribution upon mixing with loess loam.

The next stage of the experiments was preparation of ceramic pastes with addition of organomineral conglomerate based on oil sludge. The prepared components were used for production of raw composition by weighing and dosing. Actual considered compositions are summarized in Table-2.

Table-2. Ceramic compositions.

No.	Components, wt %	
	Loess loam	Conglomerate with oil sludge
1	88.0	12.0
2	85.0	15.0
3	72.0	18.0

Ceramic paste with initial moisture content of 18--20% was prepared of the considered samples. Then, sample cylinders with the diameter and the height of 50 mm were prepared by plastic molding. These samples were dried in drying oven at 75-85°C to residual moisture content of 7-8%. The dried samples were annealed in SNOL 80/12 electric furnace according to specialized procedure. Physicomechanical properties of the cylinders were determined before and after annealing. The experimental results are summarized in Table-3.

Table-3. Physicomechanical properties of considered samples.

No.	Ceramics compositions, wt %	Physicomechanical properties before annealing			Physicomechanical properties after annealing, T=980°C±10°C			Annealing duration, h
		Coefficient of sensitivity to drying	Average density, kg/m^3	Raw compression strength, MPa	Compression strength, MPa	Average density, kg/m^3	Water absorption, %	
1	Loess loam 100 %	85	1,355	2.4	8.7	1,850	24	9
2	Loess loam 88 % Organomineral additive 12%	110	1,240	2.9	9.3	1,745	25.5	8.5
3	Loess loam 85 % Organomineral additive 15%	125	1,180	3.4	10.2	1,670	26.4	8.0
4	Loess loam 72 % Organomineral additive 18 %	145	1,150	3.8	11.5	1,510	26.8	7.5

It can be seen that with the increase in the content of conglomerate with oil sludge due to decrease in the content of loess loam, the average density decreases from 1,350 to 1,150 kg/m³. Herewith, water absorption of thermally treated samples increases, evidencing increase in porosity of the samples.

Compositions 3 and 4 are characterized by low average density in the range of 1,150–1,180 kg/m^3.

It should be mentioned that with the increase in the content of organomineral conglomerate with oil sludge, the time of occurrence of initial cracks increases from 85 to 145 s, which evidences decreased sensitivity to ceramic drying.

Therefore, addition of organomineral conglomerate with oil sludge converts ceramic paste based on loess from high sensitive to low sensitive materials.

This decreases the risk of occurrence of drying cracks in raw bricks, drying can be performed more intensively. As a consequence, the quality of ready products is retained; the drying time is reduced together with power consumption at the drying stage.

5. CONCLUSIONS

The following conclusion can be derived on the basis of the performed experimental studies: linear decrease of average density and compression strength with the increase in water absorption of thermally treated samples evidence burning-out of conglomerate with oil sludge accompanied by formation of fine porous structure of ceramic tile.

As evidenced by variation of strength and average density of treated samples at 980°C±10°C, despite decrease in average density from 1,850 to 1,510 kg/m^3, the compression strength increases from 8.7 MPa to 11.5 MPa, which confirms increase in sintering capability of ceramic paste.

In addition, the weight of final product decreases by 15-20%, thus facilitating brick laying efficiency. Herewith, load on basement of structures decreases, promoting total decrease in material consumption for construction by nearly 10–12%.

Oil sludge burning in organomineral conglomerate increases temperature in furnace and accelerates sintering of ceramic tile. This is confirmed by decrease in sintering duration from 9 to 7.5 h with respective decrease in power consumption during product sintering.

Therefore, the authors proved the possibility to apply oil sludge as energy efficient and modifying agent for manufacturing of wall ceramic tiles based on loess loam.

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