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Investigating Oil Sludges and Their Application as Energy Efficient and Modifying Component in Ceramic Pastes.

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ABSTRACT

The 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 sludges into organo-mineral conglomerate state have been developed. It has been established that addition of organo-mineral 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 organo-mineral 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 %. Basic possibility of application of oil sludges as energy efficient and modifying component in production of wall ceramics on the bases of loess loam has been verified.

Keywords: Organo-mineral conglomerate mixture, ceramic paste, oil sludge, loess loam, recovery, ceramic brick.

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INTRODUCTION

While global economy progresses and population increases, there is drastic growth of demand for energy resources. According to expert opinion, from early 1989-s up till now total global energy consumption increased by about 2 times.

In developed countries the strategy of energy efficiency promoted competitiveness of economy and industry, development of science, innovations, implementation of new technologies.

Researchers and experts of numerous countries on the basis of economic analysis concluded that it is more reasonable to invest into energy efficiency and to gain income due to savings of expenses for energy resources than to construct new facilities. In other words, energy efficiency is invisible fuel at competitive price [1].

This problem is solved by arrangement of conditions for implementation of legal, organizational, researching, industrial, technical and economical measures aimed at efficient and reasonable use of fuel resources and renewable energy.

Energy efficiency in industrial section means obtaining of the same economical result but at lower energy consumption, or obtaining of better results on the basis of the same or lower energy per unit product.

This means reduction of energy consumption and cost savings [2].

Hence, researchers of numerous countries continue their activity of innovative essence aiming at decrease in energy saving.

As a rule, such developments are commercially implemented, since industrial enterprises gain possibility not only to update their technologies but to be competitive on global markets in terms of costs and quality of final products [3-5].

In terms of improvement of energy efficiency in fabrication of construction materials oil sludges are of great concern.

Oil sludges are generated during oil and gas production, transportation and storage of petroleum products, as well as upon their processing. The amount of oil sludges increases continuously. Domestic and foreign petroleum companies are not interested in their utilization due to some objective and subjective reasons. One of the major motivations is intention to provide deep processing of oil sludges at site.

However, in order to solve this complicated problem petroleum companies need to establish new subdivisions and to erect additional processing facilities.

This approach obviously requires for additional time and financial expenses, including training of technical experts and managers specializing in innovative processing technologies of oil sludges.

Thus, nowadays most of petroleum companies consider oil sludges as wastes and attempt to transfer them to other companies specializing in recovery of oil sludges in storage ponds.

Widely applied methods of recovery of oil sludges include their combustion in specialized facilities, since oil sludges are highly inflammable and combustible materials [6].

Inflammability and combustibility of oil sludge is based on content of such valuable hydrocarbons as oil, paraffin, tars and asphaltenes.

Thus, oil sludge should be considered not as wastes but instead as valuable energy generating and additional modifying stock for other industries.

Due to insufficient amount of promising innovative developments, which can be readily applied to the use of oil sludges as energy generating and modifying stock for other industries the storage of oil sludges in ponds and their combustion result in complicated environmental issues up till now [7-8].

Numerous works of foreign researches are devoted to recovery of oil sludges [9-11]. Researchers from Vilnius Technical University (Lithuania) performed experimental study of the use of oil sludges in technology of ceramic materials. The obtained results demonstrated viability of application of oil sludges in ceramic industry in terms of modification of ceramic paste and improvement of physico-mechanical properties of final product.

The authors in [12] investigated into application of waste automobile oils (petroleum wastes) in the composition with quartz sand in order to produce light fillers as modifying additive to clay stock. Addition of 1 % of waste oil to the compositions with quartz sand made it possible to increase gas generation, swelling of clay stock, mechanical strength and to decrease swelling temperature of clay stock.

The use of oil sludge as raw stock for other industries is one of reasonable methods of its utilization, since it leads to certain environmental and economic effects.

One of important industries in economy of any country is fabrication of construction materials. This industry involves enterprises the technological route of which includes obligatory high-temperature annealing related with high consumption of energy resources (coal, gas, fuel oil and others).

Such technologies include fabrication of ceramic brick, keramzit, agloporite, lime, cement.

Currently the ceramic brick plants in Kazakhstan face the following difficulties:

- insufficient and limited amount of high quality clay raw stock for brick fabrication;
- due to insufficient reserves of high quality clay stock the manufactures of ceramic brick should use poor quality stuff in the form of loess loams;
- the final product on the basis of loess loams is characterized with low strength, water and frost resistance;
- the chemical and mineralogical composition of loess loams prevents fabrication of high quality brick even at high annealing temperatures (1050-1100°C);
- the ceramic brick technology with the use of loess loams is characterized with high energy consumption, since it requires for high annealing temperature in commercial kiln (1050-1100°C) and long time (up to 72 hours) in order to provide complete annealing of ceramic brick.

Hence, we investigated into oil sludge and possibility of its application in composition of ceramic paste and energy generating and modifying component in order to fabricate high quality ceramic brick.

This aim can be achieved by solution of the following tasks:

- study into properties and chemical-mineralogical composition of Chagan loess loam;
- study into chemical and rheological properties of oil sludge aiming at determination of hydrocarbon and other compounds which can improve molding, drying and annealing properties of loess loams;
- determination of combustion heat of oil sludge, promoting extraction additional energy due to combustion in the composition of ceramic paste on the basis of loess loams;
- development of technological solution for oil sludge conversion into conglomerate state suitable to addition into ceramic paste on the basis of loess loam;
- experimental studies of properties of ceramic paste and thermally processed samples based on loess loams in composition with oil sludge convert4ed into conglomerate state.

EXPERIMENTAL

The hydrocarbon composition of oil sludge was analyzed using an Agilent 5975C Series GC/MSD (US).

Fractional composition of the considered oil sludge was determined using an ARN-LAB-02 unit, sublimating 10 ml of tested sample and continuously monitoring the temperature readings and volumes of condensate.

Sulfur weight fraction in the considered oil sludge was determined using a Spektroskan-Maks GF2E (Russia) by measurement of sulfur fluorescent radiation intensity upon X-raying of the considered sample.

The combustion heat of the considered sample was determined using a C2000 calorimeter (IKA-Werke, Germany) upon complete combustion of pre-weighted sample in calorimetric bomb in compressed oxygen and measurement of heat amount evolved upon combustion including auxiliary substances.

The content of mechanical impurities was determined by filtration of the considered oil sludge with preliminary dissolution in petrol and filtration residue washing with solvent and subsequent drying and weighing.

The content of chlorides was determined by titration of aqueous extract by bivalent mercuric nitrate in the presence of diphenylcarbazide reagent.

The density of the considered oil sludge was determined by submerging of aerometer into the considered product and recording of aerometer readings at the temperature of determination with subsequent conversion of the results into the density at 20°C.

Crystalline phases, particle and crystal sizes, crystalline to amorphous phase quantitative ratio were determined using a JEM-2100 transmission microscope (JEOL, Japan).

The surface topography and microstructure of various samples, qualitative and quantitative analysis of sample composition in point area, plotting of profiles of element distribution along preselected line, mapping of element distribution from certain segment were obtained using a JSM-6390LV scanning electron microscope (JEOL, Japan).

Qualitative and quantitative phase analysis, determination of cell parameters and crystal orientation, analysis of polycrystalline structures, microstresses and textures, qualitative elemental analysis of inorganic matters, isotopic analysis were perforemed using a PANalytical XPert Pro MPD Diffractometer.

Upon experiments a sample of loess loam was crushed in a MShL 100x250 laboratory jaw crusher up to obtaining of 5-10 mm particle sizes, then milled in a MShL-1P laboratory ball mill up to obtaining of specific surface area enabling passing through 1 mm sieve.

RESULTS AND DISCUSSION

The study was performed with oil sludge supplied by TOO ZHAIKMUNAI petroleum company (Uralsk, Kazakhstan), the chromatography analysis was performed according to the procedure given elsewhere [13]. The obtained results are summarized in Table 1.

Table 1: Group composition of hydrocarbons on the basis of chromatography-mass spectrometry

Hydrocarbon groups in oil sludge	Quantitative content, wt %
Paraffin wax	46.38
Non-condensed cycloparaffins	27.71
Non-condensed cycloparaffins with 2 rings	8.45
Condensed cycloparaffins with 3 rings	6.92
Benzene	2.74
Naphtene-benzene	0.10
Di-naphtene-benzene	0.10
Naphthalene	3.66
Acenaphthene	2.96
Phenanthrene	0.98

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Rheological properties, that is, density, particle size distribution, sulfur weight fraction, combustion heat, content of mechanical impurities and chlorides in the considered oil sludge were studied according to the procedure given elsewhere [14]. The obtained results are summarized in Table 2.

Table 2: Rheological properties of oil sludge

Property	Values
Density, kg/m^3 at 20°C	836.4
Fractional composition, vol % 200°C 300°C 350°C	 11 39 54
Sulfur weight fraction, %	0.024
Heat of combustion, kJ/g	44.987
Mechanical impurities, %	0.027
Chloride salts, mg/dm^3	28.46

The obtained results demonstrate that oil sludges of TOO ZHAIKMUNAI petroleum company contain up to 97 % of hydrocarbons and are characterized with high combustion heat. The combustion heat of the considered oil sludge is 44.987 kJ/g, which is close to that of coal.

Chagan loess loam (West Kazakhstan Region) was used as clay component.

The Chagan loess loam is characterized with significant porosity up to 50 %. This is confirmed by the SEM analysis (Fig. 1). It can be obviously seen in the images that the particles of clay minerals are penetrated by pores of various shapes and sizes.

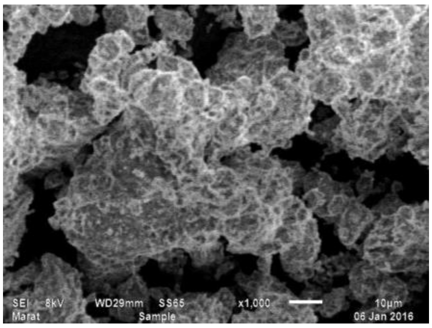

Figure 1: Porous microstructure of loess loam

Addition of oil sludge into the composition of ceramic paste should be thoroughly elaborated, since natural clay is referred to inorganic silicate substances, and oil sludge being hydrocarbon raw stock is referred to organic substances.

According to Koren'kova and Sheina [15] oil sludges are petroleum disperse systems of colloidal chemical origin. They are low concentrated slurries composed of mineral solids with absorbed high-molecular organic compounds on their surface. This is a disperse phase, and water-oil emulsion is a disperse medium. In such state the minor addition of oil sludge into ceramic paste leads to the following problems:

- the clay paste is immediately coagulated after contact with oil sludge, since porous macrostructure of loess loam facilitates absorption upon their contact;
- coagulation occurs only in the contact points which excludes possibility of uniform distribution of oil sludge in ceramic paste;
- it is impossible to add oil sludge into ceramic paste without development of innovative technological approaches to fabrication of ceramic brick;
- it is required to solve the problem of addition of oil sludge into ceramic pastes based on loess loams, providing uniform distribution of their finely-dispersed particles;

- it is required to provide technological conditions promoting energy efficiency of the technology and increase in quality of final products.

In order to achieve the preset aim it was required to convert oil sludge from slurry into loose condition, thus providing maximum similarity to looseness of loess loam. Herewith, it is necessary to achieve and maintain maximum concentration of oil sludge in the composition for their subsequent application in ceramic paste.

Loess loam was dried in drying oven at 100-120°C up to residual moisture content of 5-7 %.

Then the dried loess loam was crushed in a ball mill up to complete passing through 1 mm sieve.

The oil sludge upon natural storage in a tank is a slurry. The oil sludge in a tank is conventionally subdivided into three layer. The top layer of oil sludge slurry is composed of light fractions, and heavier fractions are in the middle and bottom layers.

In order to obtain uniform distribution of all three fractions of oil sludge in ceramic paste the composition was blended by intensive agitation in a propeller mixer.

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

Then, the oil sludge : loess loam mixture in volumetric ratio of 1:5 was prepared and thoroughly stirred in a laboratory stirrer. The obtained stuff is loose organo-mineral conglomerate with average density of 1000-1100 kg/m^3.

The macrostructure of the obtained organo-mineral conglomerate was studied using SEM (Fig. 2).

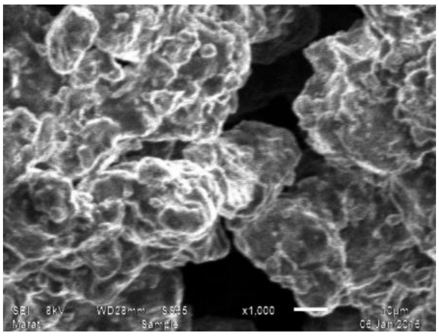

Figure 2: Microstructure of organo-mineral conglomerate mixture with oil sludge.

The SEM results (Fig. 2) reveal that finely dispersed slurry of oil sludge penetrates into pores of loess loam, forming steady contact links.

In such state in the structure of organo-mineral conglomerate based on finely crushed loess loam it is a disperse medium, and the oil sludge slurry is a disperse phase.

This procedure converts the oil sludge from slurry into loose organo-mineral conglomerate and provides good basis for subsequent procedures such as dosage and iniform distribution upon mixing with loess loam.

Another stage of the researches was preparation of ceramic pastes with addition of organo-mineral conglomerate based on oil sludge. On the basis of prepared components the raw stock composition was made by weighing. The obtained compositions are summarized in Table 3.

Table 3: Component compositions of ceramic paste

Composition #	Components, wt %	
	Loess loam	Conglomerate mixture with oil sludge
1	88.0	12.0
2	85.0	15.0
3	72.0	18.0

The considered compositions were used for preparation of ceramic paste with molding moisture content of 18--20 %. Then, cylindrical samples with the diameter and height of 50 mm were prepared by plastic molding. The prepared samples were dried in oven at 75-85°C up to residual moisture content of 7-8 %. The dried samples were annealed in a SNOL 80/12 electric oven in accordance with dedicated mode. Physico-mechanical properties of the cylindrical samples were analyzed before and after annealing. The obtained experimental results are summarized in Table 4.

Table 4: Physico-mechanical properties of the considered samples

#	Composition of ceramic paste, %	Physico-mechanical properties before calcination			Physico-mechanical properties after calcination, T=980°C±10°C			Time of calcination, hours
		Coefficient of sensitivity to drying	Average density, kg/m^3	Raw compression density, MPa	Compression strength, MPa	Average density, kg/m^3	Water absorption, %	
1	Loess loam 100 %	85	1355	2.4	8.7	1850	24	9
2	Loess loam 88 % Organo-mineral additive 12%	110	1240	2.9	9.3	1745	25.5	8.5
3	Loess loam 85 % Organo-mineral additive 15%	125	1180	3.4	10.2	1670	26.4	8.0
4	Loess loam 72 % Organo-mineral additive 18 %	145	1150	3.8	11.5	1510	26.8	7.5

CONCLUSIONS

The obtained experimental results demonstrated that while the content of conglomerate with oil sludge increases due to decrease in the content of loess loam the average density decreases from 1350 to 1150 kg/m^3. Herewith, the water absorption of thermally treated samples increases, evidencing increase in the porosity of the samples.

Low values of average density are observed for the compositions #3 and #4, they are in the range of 1150--1180 kg/m^3.

It should be mentioned that with increase in the content of organo-mineral conglomerate mixture with oil sludge the time interval before occurrence of the first cracks increases from 85 to 145 seconds, which evidences decrease in sensitivity of the ceramic composition to drying.

Thus, the addition of organo-mineral conglomerate mixture with oil sludge converts ceramic paste based on loess loam from high-sensitive to low sensitive substances.

Hence, the risk of occurrence of dry cracks in molded raw bricks decreases, and it is possible to carry out drying procedure in more intensive modes. In its turn, this facilitates keeping of quality of final products and decreases the drying time, thus reducing energy consumption at the drying stage.

The following conclusions can be made on the basis of the obtained experimental results:

- linear interrelation between decrease in average density and strength upon compression accompanied by increase in water absorption of thermally treated samples evidences burning-out of conglomerate mixture with oil sludge with generation of fine porous structure of ceramic bodies.

As shown by the analysis of variation of strength and average density of thermally treated samples at 980°C±10°C, despite decrease in the average density from 1850 kg/m^3 to 1510 kg/m^3 the compression strength increases from 8.7 MPa to 11.5 MPa, which obviously evidences increase in the annealing extent of ceramic paste.

In addition, the weight of final product decreased by 15-20 %, which improves labor conditions and increases labor performance during bricklaying. Moreover, the load on structure foundation decreases, thus reducing materials intensity for construction by about 10-12 %.

Combustion of oil sludge in organo-mineral conglomerate mixture makes it possible to increase the temperature in furnace and accelerates annealing of ceramic bodies. This is evidenced by decrease in annealing time from от 9 to 7.5 hours. This reduces energy consumption at annealing stage.

Therefore, we proved the possibility to use oil sludges as energy efficient and modifying component for production of wall ceramics on the basis of loess loams.

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