Modifying of ceramic mass by Kazakhstan bentonite for the purpose of improvement of structure and physicomechanical properties of front wall ceramics

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Abstract. Current trends of construction industry development by the criterion of rational environmental management demand scientific approach on further ensuring production of construction materials with available raw material resources, decrease in labor costs and energy resources. For the purpose of decrease in roasting temperature, sensitivity coefficient to drying, for reduction of energy consumption when drying, and also increases of strength indicators, the influence of bentonite clay for obtaining front wall ceramics was investigated. Rational structures of ceramic composition for the production of ceramic brick by the method of semidry pressing with the use of low-grade loam and bentonite clay were received as a result, influence of temperature of roasting on change of physicomechanical properties of the samples on the basis of the developed structures of composition was investigated, consistent patterns of structure and phase formation of ceramic composition depending on roasting temperature were determined. At the research of physicomechanical properties of the obtained samples, it was revealed that mix modifying by bentonite clay previously activated to nanodimensional particles, led to the increase in crack resistance of ceramic mass, durability increase for 30-40% at the general decrease in temperature of roasting for 100-150ºC.

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Introduction

Clays and clay minerals are very important in the processing industries, agriculture, mechanical engineering, construction, and geology, ecological and other appendices. They are widely used as the main raw materials at the production of various pottery for construction materials, such as brick and tile because of many specific properties before and after the roasting [1,2,3]

However, the source of raw materials of the majority of plants of the Republic of Kazakhstan is focused on the use of low-quality loesslike loams and loess which are strongly sanded and contain a large number of carbonates, not allowing receiving quality ceramic brick.

Because of poor quality and instability of chemical composition of loams when roasting the products, full courses of structurization processes are not provided even at high temperatures of roasting (t = 1000 … 1050 °C) that leads to the formation of imperfect crystal structure, responsible for providing operational properties of ceramic crock. [4].

The authors [5] suggest to add 5-10% on weight from various bentonites from Sardinia, Italy to the loams imported from foreign countries, such as Ukraine, Germany, France and Portugal (about 3 × 10 6 t year^{-1}) [6] and [7] which provide high durability. Rheological researches were conducted on the suspensions prepared for the check of bentonite presence effect in dependence of shift and time. The burned samples were estimated by the research of mechanical and esthetic properties, and also technological parameters, such as water absorption, linear shrinkage in comparison with standard industrial composition.

The choice of roasting temperature depends on mineralogical structure of raw materials, its temperature of melting and interval of agglomeration, chemical composition of raw materials, feature of the furnace used for the roasting of samples and excerpt lasting at maximum temperature of roasting. [8]] . The influence of roasting temperature is especially visible during the roasting of raw mix created from quite large number of additives [9] .

The authors of publication [10] showed that density and durability of ceramic samples changed depending on maximum temperature of roasting ranging from 1040 °C to 1080 °C. The changes of properties (density, chemical and mineralogical composition, durability) are considered in the work [11]. Glass additive was used as raw composition at the temperature from 600 °C to 1200 °C. Ceramics with durability up to 90 MPa and with density up to 2560 kg / m^3 was received as the researches result. [9]

Within the last two decades, a new direction of research and practical activities appeared in the materials science. It is connected with the emergence of new type of materials connected with formation of so-called nanostructures.

Manipulating nanoobjects by means of special technologies, it is possible to create essentially new materials and designs. Unusual properties of these materials are caused both by features of separate particles and their collective behavior. Nanomaterials can be used not only in pure form – addition of such particles to other materials in principle can also give specific properties to the materials which did not possess it earlier.

In particular, clay minerals, or smectites which are the main components of widespread bentonites, practically possess all properties of natural nanodimensional particles. They are used generally spontaneously at the production of construction materials, ceramics, plastic, and also as additives in metallurgy, foundry production and in the form of suspensions at hydroconstruction works and drilling. [12]

Today in West Kazakhstan there is one of the largest fields of Kazakhstan - Pogodayevskoe field with a stock of clays more than 6181 thousand m^3. These raw materials in our region are used only in the production of expanded clay at "Stroykombinat" enterprise. [13]]

This work is directed on the research of concrete fields of local clays collected from Westn Kazakhstan for the assessment of their use potential in the production of front ceramic brick.

Materials and Methods

Loam of Chagansky field and bentonite clay of Pogodayevskoe field of West Kazakhstan region was used as clay rock.

Samples of bentonite were taken from stacks of the material, being stored in a warehouse, by taking out method. The sample was taken by shovel at the depth not less than 30 cm from 20 points. 5-6 pieces weighing about 60 gr each were selected in each point.

Test of Chagansky field loam was selected from the field from 4 single samples. The mass of single sample was 6 kg. Single samples were mixed up for obtaining joint sample.

Definition of chemical and mineralogical composition of studied raw products was carried out by raster electronic microscope JSM-639LV of "JEOL" firm with a prefix of power dispersive analysis INSAEnergy of "OXFORD Instruments" firm (table 2, 3).

For the preparation of raw products for experimental works, laboratory jaw crusher and laboratory spherical mill of MShL-1P were used. At first raw products were separately split up in the crusher to lumpy fraction of 5-10 mm. Then loam of Chagansky field was ground in a spherical mill to the specific surface of 600-750 cm^2/g. Water suspension in the ratio 1:3 (bentonite-water) with a density of 1,3-1,4 g/cm^3 was previously prepared from average sample of Pogodayevskoe field bentonite clay in natural state. The density of suspension was defined by areometer. Propeller mixer was used for hashing of suspension МП-003.

Prepared raw products were mixed up on the studied ratios (table 1), thus humidity of furnace charge was 8-10%. Humidity of raw mix was controlled by MX-50 analyzer of humidity.

Compositions of composite ceramic mixes are given in table 1.

Table 1. Charge structure of the studied composition

No of composition	Maintenance of masses components, %	
	loam	bentonite suspension
1	90	10
2	85	15
3	80	20
4	75	25
5	70	30

Then raw mix was formed in the samples-cylinders by the sizes 50x50x50 by the method of moist pressing at hydraulic test press PGM-500MG4 with the automated system of data input. Pressure of pressing was 15-20 MPa.

The formed products were burned without preliminary drying in SNOL 12/12-V muffle furnace to 950-1000 °C. Speed of temperatures lifting is 1,5-2 °C a minute.

The research of raw materials and ceramic compositions microstructure were executed by raster electronic microscope JSM-6390LV and by JEM-6610LA device (JEOL firm, Japan). Analytical researches of element structures were conducted by x-ray X'Pert PRO diffractometer (Japan) which are given in figures 5, 6, 7.

The studied properties of ceramic masses are: sensitivity coefficient to drying, air shrinkage, raw durability as the criteria of drying and forming properties; fire shrinkage, durability at compression and bend - as indicator of products quality.

Sensitivity of clays to drying is defined by their crack resistance in this process. Emergence of cracks is connected with the tension resulting unequal size of shrinkage on section and on product surfaces. Cracks are formed when arising tension surpasses strength of material. With the increase in durability of clays, their crack resistance, as a rule, increases. Therefore the additive of high-plastic clay to the sanded one in some cases promotes improvement of dried-up products quality. [4]

Sensitivity coefficient to drying was determined by the express method offered by A.F. Chizhsky, is based on establishment of radiation duration of damp sample from clay by powerful thermal stream before emergence of cracks on its surface and differs in definition speed, and also quite simple equipment design. For the determination of Z_0 radiation duration, the set which schematic diagram is shown in drawing is recommended 1.

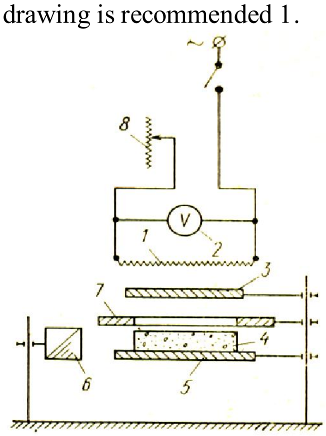


Figure 1 - The scheme for sensitivity definition of clays to drying by the express method

According to this scheme, on the basis of Zhangir khan WKATU laboratory, the set allowing to define sensitivity coefficient to drying was created. Laboratory autotransformer was used for power regulation of electric tile heating.

The set which schematic diagram is shown in fig. 25 is recommended for the determination of Z_0 radiation duration. The electrotile 1 with a power of 400 W can be the source of radiation. Sample 4 in the form of tile 55X55X16 mm from the mass of normal forming humidity is placed on a lifting table 5 by means of which certain distance between radiator and sample is determined. Comprehensive viewing of the sample is provided with mirror 6. The screen 7 with an opening of 55X55 mm serves for the prevention of sample lateral surface and lifting table heating. Constant voltage is supported on the voltmeter 2, regulating it by rheostat 8. The size of thermal stream is periodically controlled by heat meter (thermoprobe). The continuous screen 3 serves for the protection of tested sample and table from premature heating during the set preparation for the test (at a tile warming up, change of samples etc.).

By data of A.F. Chizhsky, at thermal flow of 7000 W/sq.m and distance between radiator and sample of 60 mm, Z_0 of highly sensitive clays is 35-50 s, medium sensitive 60-80 s, and insensitive 90-130 s.

Air shrinkage of clay masses was defined as follows:

$$Y_B = \left[\left(l_0 - l_1\right)/l_0\right]100, \quad (1)$$

where l_0— length of freshly formed sample; l_1— sample length after drying (EN 771-1:2003).

Fire shrinkage is expressed in percentage of the formed sample length:

$$У_{ог} = [(l_1 - l_2)/l_0]\ 100\%, \quad (2)$$

where l_0 — length of freshly formed sample; l_1— sample length after drying; l_2 — sample length after roasting (EN 771).

The strength at compression was defined by the way of samples series test (not less than 3) by hydraulic press PGM-500MG4. The press has automated system of calculation of the received values of durability on the formula:

$$R_{сж} = \frac{P}{F}; \text{ MPa or kgs/cm}^2 \quad (3)$$

where P - destroying force, H (kgs);
F - area of sample cross section, m^2, cm^2.

Frost resistance was defined with the use of heat and cold camera KTX with the microprocessor device. Chamber temperature is -17 ºC – when freezing and 22 ºC – when thawing. The device of heat and cold camera provides high precision of the set temperature maintenance and big degree of safety in operation.

Water absorption was defined on the burned samples according to EH 771-1:2003. On the average three samples were dried up up to the constant weight. Then they were stacked in the vessel filled with water with a temperature (20±5) so that water there was above the samples for 2-10 cm. Ceramic samples were maintained within 48+1 hours. The samples sated with water were taken out, rubbed off by dry fabric and weighed. Value of water absorption (W) was defined as follows:

$$W=[(m_1-m)/m]100;$$

where m_1 – mass of the sample sated with water, gr;

m – mass of the sample which has been dried up to the constant weight, gr.

Arithmetic average of water absorption definition results was taken for water absorption value definition accurate to 1%.

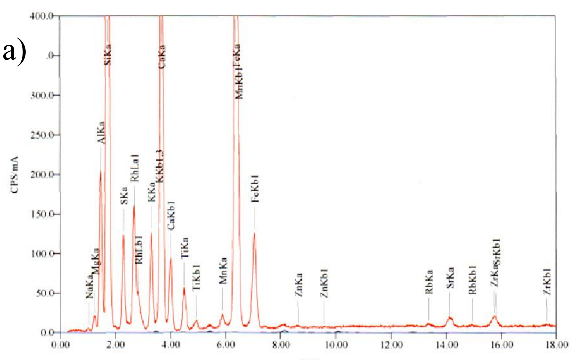
Research & results

Chemical compositions of loesslike loam of Chagansky field and bentonite clay of Pogodayevskoe field of West Kazakhstan region, the Republic of Kazakhstan are presented in table 2, 3.

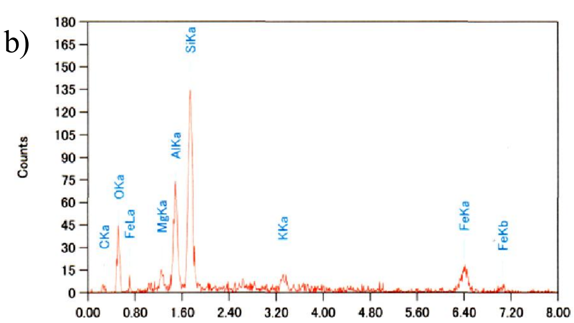
According to the maintenance of Al_2O_3, loam belongs to the group of sour raw materials, and fusible on fire resistance. According to the maintenance of

Raw materials name	Content of oxides, mass%												
	SiO_2	Al_2O_3	TiO_2	CaO	MgO	Fe_2O_3	P_2O_5	F	SO_3	CO_2	Na_2O	K_2O	Other chemical elements
Loam Chagansky	55,16	14,3	-	13,2	3,2	7,3	-	-	2,81	-	-	2,6	1,43

a)



b)



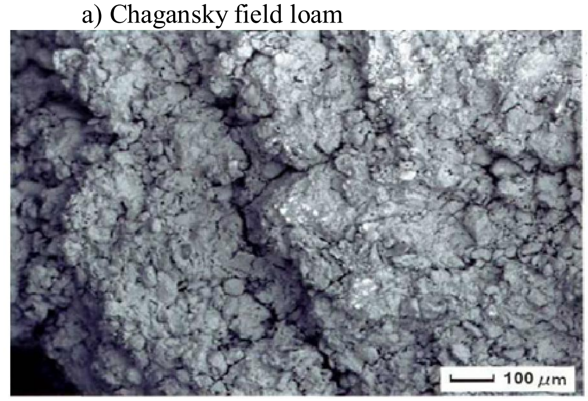
a) Chagansky field loam; b) bentonite clay of Pogodayevskoe field

Figure 2 – Roentgenogram

From crystal phases clay of Chagansky field also contains quartz d/n=4,23; 3,34; 1,974; 1,813; 1,538*10^{-10}m, feldspar d/n=3,18; 2,286*10^{-10} m, calcite d/n=3,02; 2,018; 1,912*10^{-10}m and hematite d/n=1,839; 1,686; 1,590*10^{-10}m.

Mineralogical composition of clay (figure 2) is presented generally by montmorillonite d/n=5,06; 4,46; 3,79; 3,06; 2,455; 2,28; 2,127; 1,977; 1,817; 1,675*10^{-10}m.

a) Chagansky field loam



Raw materials name	Content of oxides, mass%							
	SiO_2	Al_2O_3	CaO	MgO	Fe_2O_3	SO_3	Na_2O	Other chemical elements
Pogodayevskoe field	61,51	17,06	2,27	3,21	6,36	1,27	3,57	6,75

Analysis of chemical-mineralogical composition of loam showed that there are to 10-12% calcite minerals that negatively influences the quality of finished products. Calcite ($CaCO_3$) in ceramic mass when roasting forms free oxide of calcium (CaO). In the pottery having free oxide of calcium in structure, there are cracks which became result of chemical reactions at interaction with atmospheric moisture and emergence of internal tension because of intensive evaporation of gases and heat allocation. [Article NIA RK]

In mineralogical composition of bentonite clay there is no calcite and generally bentonite clay consists of montmorillonite minerals which promote enrichment of ceramic mass with useful minerals.

Mineralogical composition of bentonite consists generally of montmorillonite (85-95%) and belongs to layered silicates. At hydration of these minerals, molecule of water can enter intervals between elementary layers of crystal lattice and move them apart essential, increasing thus in volume, that explains their high swelling capacity. Besides, layered silicates possess high ability to ionic exchange, i.e. replacement of some ions on a surface and in a crystal lattice of particles on ions arriving from the outside. [7]

Results of the given-out parameters of processing on microstructure of ceramic composition loam-bentonite clay are given in table 4.

Table 4. Results of chemical element analysis of the sample from ceramic composition loam-bentonite on sites

Spectrum	B stat.	C	O	Na	Mg	Al	Si	S	K	Ca	Ti	Fe	Total
Spectrum 1	Yes	1.97	62.56		0.21	0.92	32.50		0.24	1.00		0.61	100.00
Spectrum 2	Yes	0.00	58.45	2.02	2.26	7.84	19.70	0.25	0.76	4.07	0.19	4.45	100.00
Spectrum 3	Yes	0.00	47.08	1.13	1.41	5.31	22.33	0.23	1.05	10.25	0.57	10.64	100.00
Total spectrum	Yes	0.00	53.65	1.23	1.50	6.74	23.78	0.18	1.68	6.46	0.35	4.43	100.00
Max.		1.97	62.56	2.02	2.26	7.84	32.50	0.25	1.68	10.25	0.57	10.64	
Min.		0.00	47.08	1.13	0.21	0.92	19.70	0.18	0.24	1.00	0.19	0.61	

Note - All results are in weight %

Sample number	Mineral code	Mineral name	Chemical formula	Contents, %
1	01-070-2517	Quartz low – theoretical *(quartz)*	$Si\,O_2$	28
2	00-041-1480	Albite, calcian, ordered *(albite, calcite)*	$(Na, Ca)\,Al\,(Si, Al)_3\,O_8$	7
3	01-074-2217	dialuminium oxide silicate *(di-aluminum silicate oxide)*	$Al_2\,Si\,O_5$	53
4	01-083-1614	albite, high, sodium tecto-alumotrisilicate *(albite, sodium alumino-trisilicate)*	$Na\,(Al\,Si_3\,O_8)$	12

Complex and comparative analysis of microstructure, change and distribution of main silicates forming chemical elements in ceramic masses show that introduction of bentonite suspension shows considerable general change of morphology of ceramic composition loam-bentonite microstructure. The confirmation is electronic pictures executed in various increases, both pure loam, and ceramic composition, modified by bentonite clay (figure 3).

Microstructure of ceramic crock modified by bentonite clay differs in homogeneity and more uniform and dense interposition of clay units owing to enveloping of larger particles of loesslike loam with fine dispersed particles of high-plastic bentonite clay.

Such interposition of two clay rocks differing on physicomechanical and chemical-mineralogical structures is provided with a choice of optimum ratios of making components, and also processing methods of raw materials preparation taking into account specific features of loesslike loam and bentonite clay properties.

It is confirmed by the improved indicators of ceramic samples properties at the stage of formation, drying and heat treatment (table 6).

Analysis of chemical-mineralogical structure change of the studied ceramic composition in the course of heat treatment shows the emergence of new aluminosilicate crystal phases and calcic compounds of difficult structure (table 5). Results of these data show that ceramic crock steadily contains minerals of anorthite, albite and crystal compounds including SiO_2. Comparison and comparative analysis of chemical elements distribution between pure loam and modified ceramic compositions demonstrate their

No of structure	Sensitivity coefficient	Raw characteristic			$T_{roasting}$	Characteristic of thermoprocessed samples			Frost resistance, F
		R, MPa	Air shrinkage, %	$R_{dried\text{-}up\ raw}$, MPa		Fire shrinkage, %	Strength at compression, $R_{compression}$, MPa	Water absorption, %	
1	70	1,42	3,7	4,12	950-1000 °C	1,82	8,41	26,4	20
2	90	1,65	4,2	4,72		2,43	8,87	24,26	30
3	110	1,78	4,4	5,24		2,75	10,26	22,81	50
4	114	1,98	4,8	6,12		3,24	14,45	21,7	40
5	130	2,14	5,2	6,81		3,82	15,67	21,4	45

in the content of bentonite clay in the form of suspension from 3,0 to 30%, due to reduction of loesslike loam content, gradually increases of ceramic mass sensitivity coefficient to drying.

The indicator increases from 70 sec. to 130 sec. that shows the transition of ceramic mass from insensitive category to medium sensitive. At the same time durability increases at the compression of recent molded sample from 1,42 MPa to 2,14 MPa that testifies to the increase of ceramic mass binding ability. Durability growth testifies to it at the compression of dried-up samples from 4,12 MPa to 6,81 MPa as well.

Analysis of physicomechanical properties change of thermoprocessed samples showed that durability increases at the compression from 8,41 MPa to 15,67 MPa with the increase of bentonite clay in the content that characterizes improvement of ceramic mass sinterability confirmed by the growth of fire shrinkage indicators from 1,82% to 3,82%. Thus decrease in indicators of water absorption is from 26,4% to 21,4%.

Frost resistance of the samples prepared by the offered way was 50 cycles.

The indicators of physicomechanical properties of the tested samples conform to EN 771-1:2003 requirements for front wall ceramics.

Disscussion, summary & conclusions

Distinctive feature of the offered way is that bentonite clay is entered into ceramic mass structure in the form of water suspension with the density of 1,35-1,40 g/cm ^{3}.

Bentonite clay naturally contains more than 20% of chemically connected water that creates a certain problem of natural lumpy clay translation into powdery state. Therefore to receive bentonite powders, its preliminary drying to air dry state is necessary, then it is necessary to subject it to crushing on pieces of 20-30 mm and only after that bentonite clay is capable to be exposed to mechanical crushing to powdery state. Besides, bentonite clay at the interaction with water strongly bulks up (to 16 times) and at its introduction into ceramic mass in a powdery state, forming and drying properties of raw worsen because of strong swelling. [5]

The way offered by us allows to exclude completely these shortcomings as bentonite possesses extremely important property and ability of self-dispersion at the interaction with water of the raised contents that is preparation of water suspension on its basis. The way of ceramic brick production, including clay loosening, crushing and stone partition at disintegrator rollers, clay grinding at runners, formation, drying and roasting, is known. [14]

Noted before feature of bentonite clay clay minerals, together with their high dispersion, and therefore extremely developed surface, cause very big adsorptive ability – ability to absorb actively various ions and substances from the outside. It should be noted that at the environment temperature increase at the surfaces of montmorillonite, the number of oxidation-reduction centers is formed and increases.

Thus, the process of usual lumpy bentonite clay transformation into reactionary-active small and ultra small particles is reached at the stage of bentonite suspension preparation.

At the realization of the offered way, already at the stage of joint hashing of loesslike loam-bentonite suspension, there are mutual migration of ions with the formation of structural elements that improves forming and drying properties of ceramic mass, and the number of oxidation-reduction centers increases by the stages of heat treatment with the formation of structural elements in which aluminum ions isomorphically replace silicon ions in tetrahedral coordination. These processes intensify the process of ceramic masses agglomeration, emergence new crystal and glass phases that provides rigid spatial carcass of products that explains high strength rates of finished goods. Besides, small and ultra small particles of bentonite clay shift formation temperature of glass and crystal phases in the area of low temperatures that allows to reduce temperature of products roasting for 100–150 °C. [15]

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