Quality Improvement of semi-dry pressing ceramic bricks from low-quality raw materials by the directional additives

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Abstract. The advantages and disadvantages of ceramic bricks production by the method of semi-dry pressing, using traditional technology, are considered. Urgent problem of improving the available clay rocks’ properties, depleted in clay fractions and containing large amounts of dust particles is appeared in many regions due to the shortage of high-quality clay raw materials. The used materials were clay rocks of the Novosibirsk region. Research methods were introduction of directional additives to the composition of raw mixes that affected the compressive strength, water absorption, brightness and color of the burnt shard. The research results showed the need for an individual approach to the raw materials of each field. Types of additives, contributed to reducing water absorption, eliminating crack formation, increasing strength, expanding the color range of products, were identified.

1. Introduction
Increasing attention is paid to semi-dry brick pressing technology recently. Stony inclusions are removed from clay raw materials and subjected to grinding in disintegrators with the traditional technology of semi-dry pressing. Since this technology works stably, the breed must be pre-dried at a moisture content of raw materials of not more than 10 wt. %. The dried rock is not uniform in moisture, so the press powder is sorted into two fractions, which are then mixed in the required proportion [1-3].

Gurov N.G. [4] justifies the building advisability of semi-dry pressing brick factories especially in Siberia. One of the main reasons for this recommendation is the lack of high-quality raw materials’ charges for plastic molding technology. A feature of the proposed technology is the introduction to the technological scheme of the grinding and drying plant (GDP) developed by Baskey LLC (Novosibirsk). Raw materials’ sensitivity to drying decreases and the harmful effects of carbonates are eliminated after processing the raw materials in GDP. The shell-core agglomerates are formed due to the tribo-charging of particles with different signs (quartz is positively; clayey, feldspar, glandular minerals are negatively) [5]. After GDP, the powder is granulated in granulator-mixer [6], the raw material is pressed, and the robot-buncher forms bags for charging into the oven. Uniflox company, as a representative of the Dutch company Ankerpoort NV in Russia, provides mineral additives for bulk dyeing and functional purposes [7]. The use of concentrated imported pigments is limited by their high cost. Therefore, both in Russia and abroad, studies are being conducted, aimed at exploring the possibility of replacing them with industrial waste containing coloring pigments [8-13]. A significant
part of the sedimentary rocks of the West Siberian lowland and steppe regions of the Krasnoyarsk Territory is overwhelmingly represented by loams with a small amount of clayey and significant - dusty particles, which leads to low technological properties of raw materials: high sensitivity to drying and crack formation during drying, difficult sintering etc as a result [14-17].

The goal of the work. Assessment of the possibility of semi-dry pressed products obtaining, including facial ones, from dusty clay rocks with a low content of clay fractions by introducing directional additives.

2. Research methods
The ceramic samples technology of semi-dry pressing was as follows. The raw material was dried and passed through a 1 mm sieve. The part of the raw material sample was in a ball mill for 2 hours. Then non-plastic components were introduced into charges and the mixture was thoroughly mixed. The resulting compositions were moistened to the specified experimental conditions. Humidity of the press components is 12 wt. %, chalk content is 25-35% by dry weight, pressing pressure is 15-25 MPa and firing temperature is 950-1050 °C. These factors are determined by the method of rational planning. Each factor changes at three levels [18]. The press powder was rubbed through a sieve with a hole diameter of 1.25 mm, simulating the granulation process. At the pressure, specified by the experimental conditions, cylinder samples with a diameter of 40 mm and a mass of 80 g were pressed. The pressure was applied in two stages. The obtained samples were dried according to the generally accepted method: for several days under a damp material, and then in an oven with a temperature of 100 - 105 ° C. Firing was carried out in a laboratory electric oven for 8 hours with exposure at a maximum temperature of 1 hour.

The burnt samples were determined by: average density, compressive strength, water absorption, brightness, color. Water absorption was determined by saturating the samples for 48 hours, the degree of the samples whiteness on a gloss meter by comparison with the standard.

3. Raw materials
Clay rocks of the Novosibirsk region were used in the work, which technological pre-burning properties are shown in Table 1.

| Clay deposit          | Granulometric composition,% by volume | Plasticity number | Air shrinkage, wt. % |
|-----------------------|--------------------------------------|-------------------|----------------------|
|                       | 50 - 1 mm - 1 mm | 5 - 50 μm | < 5 μm |                        |
| Novonikolskoe         | 17,2 | 68,1 | 14,7  | 14 | 6,9                        |
| Kleschikinsky         | 18,0 | 75,2 | 6,8   | 13 | 6,5                        |
| Berdskoye             | 11,5 | 75,5 | 13,0  | 15 | -                          |
| Maslyaninsky          | 1,8  | 72,7 | 12,3  | 13 | -                          |

Analysis of the data given in table 1 showed that clay raw materials are of low quality, therefore, it is necessary to select the composition of raw mixtures carefully, including effective additives of directed action. The following directional additives work were used in the: mineral substances — fluxes with a high content of RO and R₂O (albitofir, diopside, diabase); fluxes of silicate composition (ground window glass, nepheline-syenite); mineral bleaching additives (limestone, chalk); mineral chromophore additives (ocher, redoxide); micro-reinforcing mineral additives (wollastonite) and organic plasticizers (C-3, Sika-20HE). The albitofir and diabase, used in this work, were wastes of crushed stone production of OJSC “Stone quarry” (Gorny settlement, NK), deposited in cyclones, wollastonite is the “Vesely” Mine, Altai Republic. Diopside was used in the Burutuiskoye mine, which was a product of the following mineral composition (% by weight): diopside (CaO·MgO·2SiO₂) – 85, quartz (SiO₂) - 15 after enrichment [19]. Fine-ground nepheline-syenite and window glass were
introduced as smooth additives. Ne-felin-syenite (Kemerovo region, “Sokol” quarry) was a natural alkaline aluminosilicate, consisting mainly of nepheline and alkaline feldspar. Window glass was a system of $\text{Na}_2\text{O}$–$\text{CaO}$–$\text{SiO}_2$–$\text{MgO}$–$\text{Al}_2\text{O}_3$. Limestone and chalk were introduced as whiteness additives. C-3 and Sika-20HE were used as plasticizers. The basis of Sika-20HE was an aqueous solution of modified polycarboxylates with pH=4.5±1.0. Super plasticizer C-3 contained 80-85% polymethylene naphthalenesulfates, 7-10% sodium sulfate and up to 10% moisture. In the ceramic industry, these plasticizers are not used.

4. Experimental results
The experiments were carried out in the following directions:
1. The effect of directional additives on the strength and water absorption of a crock;
2. The effect of directional additives on the color of a crock.

4.1. Study of the directional additives effect on the strength and water absorption of a crock
Clay rocks, studied in the work, are difficult to burn, providing a porous shard with high water absorption. Rocks with a high content of fusible oxides RO and R$_2$O are expected to have a positive effect on the process of brick charges burning. At the drying stage, these additives can act as cleansers, preventing crack formation. Since the introduction of non-plastic components into charges on the base of silty loams degraded molding properties, so C-3 and Sika-20HE plasticizers additives were widely used both in the manufacture of artificial cement-based conglomerates and in some experiments. Table 2 shows the properties of the burnt samples, molded by semi-dry pressing.

| The charge composition | Sample Properties |
|------------------------|-------------------|
| Clay deposit           | ultimate compressive strength, MPa | average density, kg / m$^3$ | water absorption, wt. % | Note |
|                        |                              |                           |                           |      |
| Maslyaninsky           | 31.4                        | 1860                      | 16.1                      | hairline cracks |
|                        | 30.3                        | 1890                      | 15.8                      |      |
|                        | 30                          | 1840                      | 16.5                      | cracks |
|                        | 35.7                        | 1920                      | 15.6                      |      |
|                        | 34.5                        | 1880                      | 15.6                      | efflorescence |
| Kleschikhinsky         | 34.4                        | 1870                      | 18.3                      | control |
|                        | 36.2                        | 1910                      | 17.9                      |      |
|                        | 40                          | 1930                      | 17.6                      |      |
|                        | 39                          | 1970                      | 17                        | efflorescence |
| Berdskoye              | 42.7                        | 1890                      | 13.9                      |      |
|                        | 40.3                        | 1960                      | 12.2                      | $T_{\text{burn}}=1000 ^\circ \text{C}$ |
|                        | 42.2                        | 1920                      | 12.8                      | $T_{\text{burn}}=1050 ^\circ \text{C}$ |
|                        | 36.6                        | 1920                      | 13.4                      | efflorescence |
| Novonikolskoe          | 30.6                        | 1900                      | 14                        |      |
|                        | 38.9                        | 1910                      | 13.5                      |      |
|                        | 42.1                        | 1930                      | 14                        | efflorescence |
|                        | 40.5                        | 1950                      | 13.6                      |      |

The results’ analysis showed that diopside has the greatest effect on the compressive strength in table 2. The crock strength with the introduction of diabase and albitofire is often close. The strength
increases up to 13% with the use of Maslyaninsky raw materials and up to 27% of those from Novonikolsky in a comparison with non-additive compounds. It should also be noted that the introduction of diopside and diabase in some cases can eliminate crack formation (Maslyaninsky raw materials). The most difficult ones in burning are Maslyaninsky and Kleshikhinsky clay rocks, forming a crock with water absorption of 16.1 - 18.3 wt. %. The introduction of the considered additives did not significantly reduce it for charges based on Maslyaninsky loam. Although the value of water absorption of samples, based on Kleshchinsky clay rock, decreased by 1.3 times, it remained very significant 17.6 - 17.9 wt. %

Efflorescences formed on the burnt samples' surface with C-3 introduction, so its use should be excluded. The reason for this is the presence of water-soluble sodium sulfate in C-3 composition [20] with the ability of either adding crystallization water to form sodium hydrosulfate \((\text{Na}_2\text{SO}_4\cdot10\text{H}_2\text{O})\) or giving it away. The formation of crystalline hydrate in the pores and capillaries of the brick is accompanied by a significant increase in its volume, contributing to its removal to the surface in the form of efflorescences. Such effect does not only spoil the brickwork appearance, but also reduces the strength of the material [21]. Unlike C-3, Sika-20HE hyperplasticizer does not contain water soluble mineral impurities and does not cause salt formation [17].

The use of the Berdsk deposit’s loam with the introduction of 10 wt. % diopside allowed reducing water absorption from 13.9 to 12.2%. With the addition of only 0.5% Sika-20HE, strength indicators were obtained, as for the non-additive composition and water absorption of 12.8 wt. %, however, the firing was carried out at a temperature of 1050 °C. Consequently, 10% of imported diopside could be replaced with 0.5% Sika-20HE plasticizing additive.

The results’ analysis, presented in table 3, confirms the need for an individual approach to the clay raw materials of each studied deposit and a careful selection of the charge composition. The occurrence of cracks in some cases, even with semi-dry pressing in laboratory conditions, indicates the need to pay serious attention to the decrease in the sensitivity of raw materials to drying.

**Table 3. Properties of the burnt samples with fluxes, bleaching and chromophore additives.**

| The charge composition,% by dry weight | Sample Properties | The crock color |
|--------------------------------------|------------------|----------------|
| loam | limestone | ground glass | nepheline syenite | other | redoxide | average density, kg/m³ | ultimate compressive strength, MPa | water absorption, % by weight | degree of whiteness, % | crock color |
| 100 | - | - | - | - | - | 1842 | 24.2 | 13.1 | 46.0 | brick |
| 95 | 5 | - | - | - | - | 1824 | 18.1 | 16.1 | 48.5 | light brick |
| 90 | 10 | - | - | - | - | 1823 | 18.3 | 16.3 | 53.0 | apricot |
| 85 | 15 | - | - | - | - | 1851 | 18.3 | 16.6 | 57.0 | light apricot |
| 75 | 10 | 5 | 10 | - | - | 1845 | 26.6 | 12.5 | 54.0 | light apricot |
| 90 | - | - | - | 10 | - | 1852 | 31.0 | 13.7 | 44.0 | red brown |
| 85 | - | 5 | - | - | 10 | 1846 | 30.8 | 11.4 | 34.0 | cherry |

Note: compositions 1 - 4 and 6 are pressed under a pressure of 20 MPa and burnt at a temperature of 1000 °C, compositions 5 and 7, respectively, at 25 MPa and 1050 °C.

**4.2. Research of the directional additives effect on the crock color**

The use of non-sintering raw materials for the production of ceramic facing bricks requires the joint introduction into charges of either melting additives, or also additives to change the crock color (bleaching and chromophore). Below experiments’ results are on the raw materials of the Baryshevsky field. The moisture content of the press powder was 10 wt. %, pressing pressure 20 -was 25 MPa, firing temperature 1000 was 1050 °C. The obtained samples were determined by the degree of
whiteness. The greatest clarification was obtained with the introduction of limestone into the charges (whiteness of 48.5 - 57%). Moreover, the increase limestone in the range of 5 to 15 wt. % mass in the content does not affect the strength and water absorption, which remains high. Only a reduction in limestone proportion to 10 wt. % and an increase in flux to 15 wt. %, increasing the pressing pressure from 20 to 25 MPa and the firing temperature from 1000 to 1050 °C allowed reducing water absorption to 12.5 wt. % and increase strength compared with non-additive composition by 10% (table. 3). With the introduction of 10 wt. % redoxide together with 5 wt. % of ground glass: the crock color is cherry, the water absorption decreased by 15%, and the strength increased by 27% compared with the control composition.

The authors also performed studies that showed the possibility of obtaining a facial cream-colored brick based on clay of the Kleschikhinsky deposit, containing only 6.8% by volume of clay particles and more than 75% of dusty fractions. Chalk served as a clarifying additive and wollastonite, a needle-like particle shape and it also served as a micro-reinforcing component. The experimental results in accordance with the planning matrix are shown in table 4. As a result of mathematical research processing, the optimum loam was obtained: chalk ratio was 3:1 and a very high strength of the samples (49.2 MPa) and low water absorption (10.5%) were obtained together with the wollastonite content of 10 % more than 100%. An increase in the chalk content above the optimum led to a sharp drop in strength and an increase in water absorption (Table 5). After firing, the material usually contains crystalline minerals such as quartz, anorthite, mullite, wollastonite, etc. No doubt, it is advisable not to introduce finely ground wollastonite, but to initiate its production during the raw mixes firing of a certain composition. A similar work was performed and presented in [22].

| The charge composition,% by dry weight | Properties of burnt samples |
|--------------------------------------|----------------------------|
| chalk content,% by dry weight        | moisture content of the press powder, wt. % | pressing pressure, MPa | average density, kg/m³ | ultimate compressive strength, MPa | water absorption, % |
| 25                                   | 12                          | 1000 15 | 1950          | 33,3                  | 13,2               |
| 25                                   | 8                           | 1050 20 | 1900          | 28,9                  | 14,3               |
| 25                                   | 10                          | 950 25 | 1980          | 40,8                  | 12,9               |
| 30                                   | 8                           | 950 15 | 1850          | 21,7                  | 16,3               |
| 30                                   | 10                          | 1000 20 | 1800         | 32,2                  | 14,6               |
| 30                                   | 12                          | 1050 25 | 1720         | 12,8                  | 20,2               |
| 35                                   | 10                          | 1050 15 | 1690         | 9,4                   | 21,6               |
| 35                                   | 12                          | 950 20 | 1650         | 10,6                  | 22,2               |
| 35                                   | 8                           | 1000 25 | 1880         | 32,8                  | 16,3               |

It was found that, crystals of anorthite CaO·Al₂O₃·SiO₂ with a lamellar structure and wollastonite were formed when the mass of calcium carbonate was up to 20%. Wollastonite crystals mainly grew with an increase in calcium carbonate content of over 20%. They reinforced the crystal crock, giving it high physical and mechanical properties. According to Yezer's V.A. and Krolevsky D.V. it must be borne in mind that with the introduction of carbonate rocks, water absorption of products often increases, which leads to rapid contamination of a building facade. In this regard, it is necessary that the water absorption of the front brick does not exceed 12-14 wt. % It is desirable to be 7-10 wt. %

Table 5. Properties of burnt samples under the optimal conditions.
### Table: Properties of burnt samples

| Pressing powder | Humidity, % | Firing temperature, °C | Average density kg/m³ | Compressive strength, MPa | Water absorption, % |
|-----------------|-------------|------------------------|------------------------|---------------------------|---------------------|
| loam chalk      | 75 25       | 20 12                  | 1000                   | 2000                      | 49.2                |
|                 | 70 30       |                        |                        | 1880                      | 37.1                |

5. **Conclusion**

Summarizing the research results, the following ways to improve the quality of ceramic bricks, based on low-quality raw materials, can be formulated: each deposit requires an integrated and individual approach; it is necessary to use advanced technological solutions at individual stages of raw materials processing into finished products: reduce the sensitivity of raw materials to drying and eliminate the harmful effects of carbonates, mechanic - thermal activation in GDP is recommended; reduce water absorption, it is most effective to introduce smoother additives into the mass, especially in combination with organic surfactants that do not contain water-soluble mineral impurities; elimination of cracking allows the introduction into the fluxing additives charges with a high content of RO and R₂O; difficult firing of clayey rocks necessitates the use of silicate composition supplements; use of bleaching additives (chalk, limestone) is recommended to white the crock; combination of whitening additives with wollastonite significantly increases strength; use of chromophore additives (ocher, redoxide) allows expanding the color range of the crock.

Thus, thanks to the use of new technological solutions and directional regulation of the raw mixes properties by introducing effective corrective directional additives, it is possible to improve significantly the quality of semi-dry pressed ceramic bricks based on low-quality clay raw materials.

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