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Use of overburden rocks from open-pit coal mines and waste coals of Western Siberia for ceramic brick production with a defect-free structure

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Abstract. The rational technology for the production of ceramic bricks with a defect-free structure from coal mining and processing wastes was developed. The results of comparison of physical and mechanical properties and the structure of ceramic bricks manufactured from overburden rocks and waste coal with traditional for semi-dry pressing mass preparation and according to the developed method are given. It was established that a homogeneous, defect-free brick texture obtained from overburden rocks of open-pit mines and waste coal improves the quality of ceramic wall materials produced by the method of compression molding by more than 1.5 times compared to the brick with a traditional mass preparation.

1. Introduction

Western Siberia along with the richest reserves of oil and energy resources has also a great mineral resource base. Intensive industrial development of the region in the XX century was accompanied by an uncontrolled development of natural resources which led to a constant growth and accumulation of industrial waste. The average utilization rate of wastes as secondary raw materials is estimated approximately at one third, which is 2.5 times lower than in the countries with the developed environmental policies [1]. As a result, Russia's industry suffers significant losses in raw materials and energy resources, the intensive accumulation of unused wastes in the environment continues – every year approximately 2 – 2.5 billion tonnes per year [2].

The problem of rational consumption of resources is especially acute in Kemerovo region, in the territory of which half of the country's wastes from mineral mining and processing are formed and stored. The main "suppliers" of wastes are the mining, energy and metallurgical industries. The annual volumes of coal mining and processing wastes stored on the territory of Russia equals to approximately 15 million tonnes [3]. As a rule, in case of open-cast mining one tonne of coal accounts for 4 tonnes of overburden the storage of which leads to significant environment pollution [4].

In the long term the solution of the problem is possible only through introduction of waste-free enrichment technologies and complex processing of accumulated wastes using their mineral part for manufacturing building materials. Overburden rocks, mining wastes, mining waste of various minerals are an inexhaustible source of raw materials for the production of building materials and products. The use of industrial wastes will cover up to 40% of construction industry's needs in raw materials and will reduce by 30% the costs for manufacturing of construction materials in comparison with their production from natural raw materials [5]. The overburden rocks at the open pit coal mines in West Siberian region are mainly represented by lean dusty loams, the wastes from processing plants – by



argillites and aleurolites of variable composition. These types of raw materials do not provide a high-quality (in terms of compressive strength and frost resistance) ceramic bricks when traditional production technology is applied. In this regard, the purpose of the present studies was to check the working hypothesis on the necessity to form a defect-free ceramic brick structure by using new approaches and methods for mass preparation of wastes from coal production and processing.

2. Methods and materials

The authors carried out research into the production of bricks from coal byproducts [6], and also developed a scheme for supplying brickworks with technological fuel due to secondary enrichment of waste coal [7].

The release of bricks from these types of raw materials in semi-dry pressing plants during the Soviet era showed a low quality of the products manufactured for strength and frost resistance [8]. This was facilitated by insufficient shredding of raw materials (up to 3 mm inclusive). The heterogeneity of the composition of press powders obtained from coarse-grained masses leads to the production of loose raw material of low strength. The method of formation of ceramic bricks with optimal structures by fine dry grinding, granulation, compression molding, drying and baking developed by the authors allows the structure defects to be avoided [9]. The rational granulometric composition of the press masses is the decisive prerequisite for good pressing and the compact packing of granules associated with it to a large extent determines the structure, physical and mechanical properties of ceramic bricks.

Implementation of the stated principles of rational mass preparation for the production of ceramics using the method of compression molding was performed on the example of overburden loamy rock and waste coal from Abashevskaya processing plant (Kemerovo region, Russia).

Novokuznetsk loam refers to the most common clay rocks on the territory of Siberia and Krasnoyarsk Krai. Raw material is low-dispersed, non-caking, highly sensitive to drying, by its mineral composition refers to polymineral rocks of hydromica-kaolinite-montmorillonite type.

The waste coal is close to clays and consists of a mineral part represented by hydromuscovite, quartz, plagioclase, montmorillonite, chlorite, siderite, calcite and a carbon residue containing free carbon in an amount of 6 to 8% after secondary enrichment. The raw material is coarse-dispersed with predominance of sand fraction, non-caking, insensitive to drying. The content of coloring oxides ($Fe_2O_3+TiO_2$) is 5.5-7 %. The chemical composition of waste coal is given in table 1. Drying and grinding of the raw material were carried out in a vortex mill-drier up to class of -300 μ m. After grinding, the plasticity of the loam was 12.5 (moderately plastic), the waste coal – 6.0 (low-plastic).

Table 1. Chemical composition of waste coal.

Raw material	Mass fraction of the components on the dried substance, %								
	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	CaO	MgO	K_2O	Na_2O	LOI*
Waste coal from Abashevskaya processing plant	57-64	14-17	0.5-1	5-6	3-5	1.5-2	2-2.5	1.5-2	6-8
Clay loam	61-64	14-15	0.8	4-5	4-4.5	2-2.5		3.5-4	5-6

*LOI – loss on ignition

3. Experimental research

In order to study the structure and properties of ceramic bricks from waste coal and coal enrichment using the developed method of mass preparation, the pilot baths of bricks were manufactured at Berdsky Brick Factory LLC. The composition of the batch for factory tests is given in table 2.

The technology of obtaining granulated press-powder for batch No. 1 consisted in the following: at the first stage the dry grinding of loam to a grade of -300 μ m was carried out, and then the loam was granulated in the intensive mixer to obtain granules fraction 1-3 mm with the simultaneous press

powder moistening up to the molding moisture 10.5-11.3 %. To obtain granulated press powder No. 2 waste coal with the fraction +13 mm was used.

Table 2. Batch composition for factory tests.

No. batch	Batch components	Content, wt. %
1	Waste coal	100
2	Waste coal	80
	Clay loam	20

The granulated press powder of batch No. 2 was obtained as follows: a separate dry grinding of loam and wastes to a class of $-300\ \mu\text{m}$ was performed, and then wastes were granulated in the intensive mixer to obtain granules fraction 1-3 mm with the simultaneous press powder moistening up to the molding moisture 12.5-13.0 % and subsequent powdering with dry grinded loam (20 wt.%).

Dry grinding of loam and wastes was carried out on the unit USP-S-04.55, granulation in the intensive mixer was carried out on the production base of Baskey Keramik LLC (Chelyabinsk). The finished press powders were packed in the bulk bags with polyethylene liners to preserve moisture and sent to Berdskiy Brick Factory LLC for compression molding of ceramic bricks.

Molding of bricks was carried out on presses SM-1085B at a pressure of 17-18 MPa. The moisture content of the granulated press powders was 9.9 % and 10.5 %, respectively. The pressed bricks in quantity of 800 pieces had a normal appearance without cracks of re-pressing. The drying and firing time in the tunnel furnace was 42 hours at a maximum temperature of 950-1000 °C. Defective bricks after firing was 1.5 % from the total amount, the fired bricks had a terracotta color, homogeneous uniformly-granular structure can be noted on the fracture. On the ceramic bricks firing cracks, swellings and curvatures were not observed. Bricks for physical and mechanical tests were selected from the fired products and the results are presented in table 3. The study of the structure and properties of ceramic bricks produced with the use of the developed technology was carried out in comparison with the samples of ceramic bricks produced from this raw material during the Soviet period (currently the plants are closed [9]).

Table 3. Physical and mechanical properties of ceramic bricks.

No. batch	Composition of batch, wt. %	Average density, kg/m^3	Compressive strength, MPa	Water absorption, %	Frost-resistance, cycle
1	Clay loam 100	1980	19.3	11.6	>50
1*	Clay loam 100	2010	11.2	14.5	35
2	Waste coal 80 Clay loam 100	1680	15.4	15.4	50
2*	Waste coal 100	1590	10.5	18.7	25

*Brick produced at the factories of the USSR

4. Examination of the brick structure

Figure 1 shows the macrostructure of ceramic bricks from loam prepared by the traditional method at Novokuznetsk brick factory in the 1970's (figure 1, a-c) and by the developed method with fine grinding and granulation (figure 1, d-f), fired at 1000 °C. The examination of the structure of ceramic compression-molded cork showed that the bricks prepared in the traditional way have a euporphytic texture (figure 1, a) [10, 11] with more intensely colored, against the background of the main mass, metamorphosed and decomposed fragments of the original minerals (figure 1, a). On their perimeter

there are macropores unfilled by the main amorphized mass and indicating the structural defects at the macrolevel occurring during firing (figure 1, a-c).

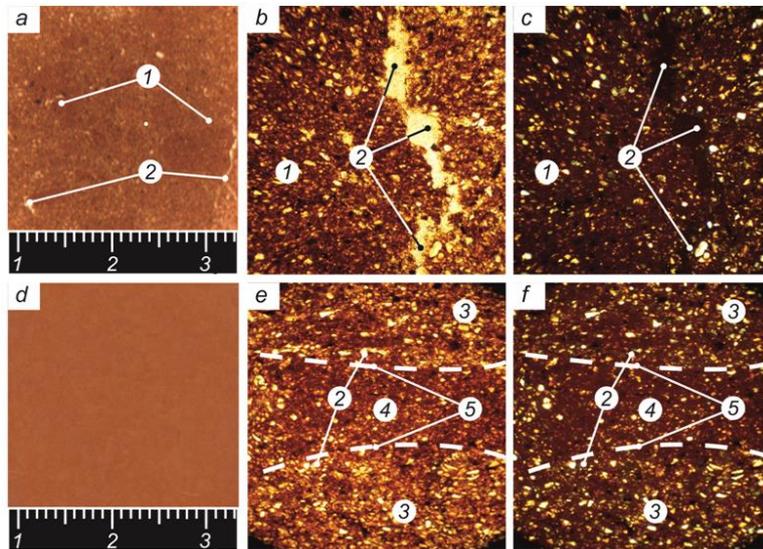


Figure 1. The macrostructure of ceramic brick made from loam prepared by the traditional method (a-c) and finely grinded granulated loam (d-f). Polished section, reflected light (a, d); the thin section, transmitted light, $\times 10$, nicoli II (b, e) and + (c, f): 1 – metamorphosed initial minerals; 2 – the pores; 3 – the granule body; 4 – the boundary layer of the granule; 5 – conditional boundary between the granules.

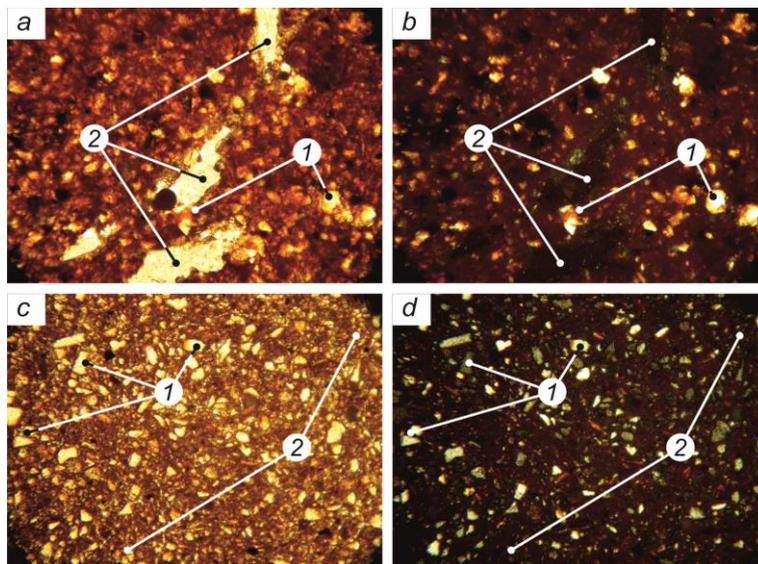


Figure 2. Microstructure of ceramic bricks made from loam prepared by the traditional method (a, b) finely grinded granulated loam (c, d). Polished section, transmitted light, magnification $\times 20$, nicoli II (a) and + (b); magnification $\times 20$, nicoli II (c) and + (d): 1 – fragments of relict minerals; 2 – pores.

In the brick from finely grinded granulated material the structure is afanite (cryptocrystalline) (figure 1, d). Fine-grained mineral phases are immersed into a full-crystalline bulk (figures 1, e, f, and figures 4.3, c, d). The bulk is colored with iron oxides, fine-grained quartz-feldspar, in which the idiomorphic quartz particles are uniformly distributed. Visually the appearance of the boundary layer between the granules on the transverse section of the brick from granulated batch is not expressed (figure 1, d).

A more intense color of the boundary layer is due to the concentration of colloidal clay fraction evolving at the boundary of the granules during pressing, in which the oxygen ions bind to the ions of aluminum, magnesium, iron and other metals located in the octahedral layer [12]. Similarly, the boundary layer is saturated with water-soluble minerals [13] which form low-melting eutectics during firing of ceramic materials. At the boundary of granules a eutectic melt is abundantly formed, in which the ions of alkali and alkaline-earth metals actively interact with iron, silicon and aluminum oxides [14, 16]. As a result during cooling ferri-ferrous dark brown (up to black) glass-crystalline formations appear. The amount and size of the pores are much less than in the samples prepared by the traditional method (figure 1-2) due to the formation of liquid-phase interlayers with a low viscosity filling the pores. The total area of pores along the section of the polished section

decreases by 50-70% (figure 2), the average pore size in the brick from loam prepared by the traditional method is 100-200 μm , in the brick of granulated loam is less than 5 μm . In crossed nicols the uniform distribution of mineral formations and relict minerals is observed (figure 2, c, d).

The decrease in the porosity in the brick from finely grinded granulated loam is explained, firstly, by the dense structure of the granulated material obtained on the mixer of intensive action at a rotational speed of the blades at 800-1000 rpm. Secondly, by rational packing of grains without air pressed inside during molding due to the established optimal granulometric composition and pressing parameters (pressing pressure and moisture of granulated powders). Thirdly, by the greater reactivity of finely dispersed mineral grains during sintering. As a result during firing a liquid phase is formed connecting relic and newly formed minerals.

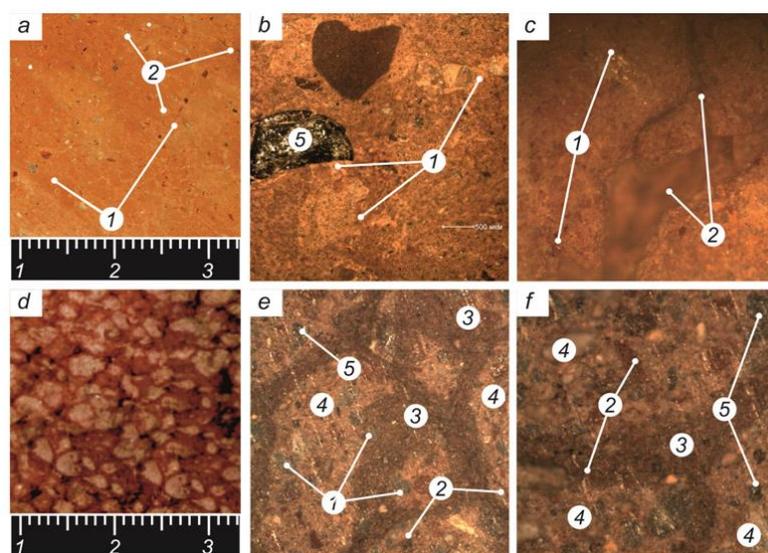


Figure 3. The macrostructure of ceramic brick made from waste coal prepared by the traditional method (a-c) and finely grinded granulated waste coal powdered by loam (d-f). Polished section, reflected light (a, d); the thin section, transmitted light, $\times 10$ (b, e) and $\times 20$ (c, f): 1 – metamorphosed initial minerals; 2 – pores; 3 – boundary layer of the granule from loam; 4 – the body of a granule from waste coal waste; 5 – unburned carbon.

pinkish-cream color (figure 3, d). The boundary layer between the granules is distinguished by a brick-red color and has a thickness of 80-200 μm . When studying polished sections under a microscope in the reflected light, a uniform distribution of large inclusions (on average 50-200 μm) of brown and gray color with an angular shape is observed. The examination results of thin sections in the transmitted plane-polarized light for parallel and crossed nicols are shown in figure 4.

On all photomicrographs of bricks made from finely grinded granulated waste coal, powered by loam, the characteristic separation of the boundary layer by the red color due to fine-dispersed hematite formations can be noted. At the microlevel the boundary layer between the granules also does not have clearly defined boundaries and passes into granules with the formation of a transition layer (figure 4, d-f).

The concentration of *carbon* in the brick from waste coal produced according to the developed method is significantly (more than three times) higher than that of a brick from waste coal prepared by the traditional method, which is caused by the secondary enrichment and fine grinding of industrial raw materials.

Figure 3 shows the macrostructure of ceramic bricks from waste coal prepared by the traditional method produced at Abashevskiy Brick Factory in the 1990's (figure 3, a-c) and by the developed method with fine grinding and granulated waste coal and subsequent powdering with loam (figure 3, d-f) fired at 1000 $^{\circ}\text{C}$.

The ceramic sample produced with the traditional mass preparation has macrocracks (figures 3, b, c) and a more porous texture at macro and micro levels (figures 3, 4). The macrostructure of ceramic bricks made from finely grinded granulated powdered with loam ... что... has a pronounced spatial framework (figure 3, d-f). The cells of the frame, formed from granulated waste coal are mostly 1-2 mm in size and filled with a fine-grained material of

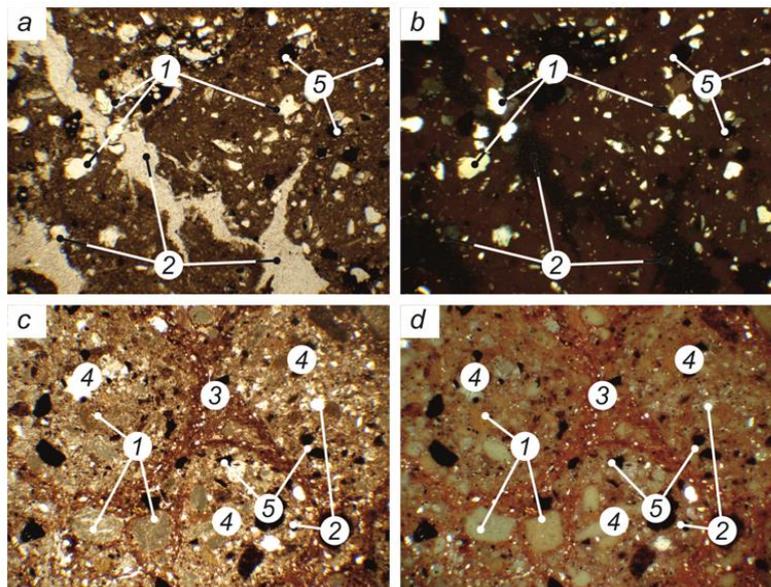


Figure 4. The microstructure of ceramic brick made from waste coal prepared by the traditional method (a, b) and finely grinded granulated waste coal powdered by loam (c, d). Polished section, transmitted light, magnification $\times 20$, nicols II (a) and + (b); magnification $\times 20$, nicols II (c) and + (d): 1 – metamorphosed initial minerals; 2 – pores; 3 – boundary layer of a granule from loam; 4 – the body of a granule from waste coal; 5 – unburned carbon.

5. Conclusion

Based on the study of the mechanical properties and structure of ceramic bricks made from coal mining and processing wastes prepared by the traditional method and using the developed method of mass preparation, it can be concluded that the surface energy of the grains and the surface area of their contact are the determining factors for the sintering of ceramic materials. The homogeneous, defect-free brick texture provides an improvement in the quality of compression-molded ceramic wall materials (compressive strength increases by 1.7 times from 11.2 to 19.3 MPa for bricks from overburden loamy rocks extracted at open-pit mines and by 1.5 times from 10.5 up to 15.4 MPa for bricks made of waste coal from Abashevskaya processing plant).

During firing in the boundary layer an intensive formation of pyroplastic phase takes place due to the predominance of the fusible component of the colloidal clay fraction migrating to the surface of granules during pressing, which leads to the formation of continuous surface contact sintering zones that increase the degree of interaction between the granules, which increases the compressive strength of ceramic bricks.

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