Rational preparation of waste coal mixture for production of bricks by the method of compression molding

A Yu Stolboushkin, A I Ivanov, M V Temlyantsev and O A Fomina
Siberian State Industrial University, 42 Kirova Street, Novokuznetsk, Russia, 654007

E-mail: ivanovaliv1989@gmail.com

Abstract. Rational preparation of the mixture containing technogenic raw material – waste coal for the production of wall ceramics is developed. It was established that the technology of high-quality ceramic bricks requires: grinding of raw materials to class 0.3 + 0 mm, its aggregation in the intensive mixers into granules 1-3 mm, compression molding of adobe to plastic deformation of granules, drying and firing.

1. Introduction
For the modern construction industry, having problems with sustainable supply of raw materials of good quality, one of the main directions of development is the utilization of technogenic wastes in the process of construction and technological production [1,2]. As a rule, the stripping ratio – 4 tonne of overburden per 1 tonne of coal causes a considerable pollution of the environment. Obsolete equipment and technology at the majority domestic coal preparation plants lead to a poor quality of coal preparation, as a result, the coal content in the waste ranges from 17 to 26%. This was the main reason for laying-off the brick factory, which more than 20 years used waste coal from the coal processing plant “Abashevkaya” in Novokuznetsk [3].

At the same time, there are examples of innovative technological solutions in our country, which allow us to consider the wastes generated during coal mining and processing not only as a source of raw materials, but also as a power base of enterprises manufacturing ceramic wall materials [4].

The authors carried out research into the problem of waste coal utilization for bricks manufacturing [5], and offered a scheme of provision brick factories with technological fuel obtained after secondary processing of coal (Figure 1).

Figure 1. Flow diagram of wall ceramics production from waste coal.

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The proposed technological idea consists in organization of the deep processing of coal right at the brick factory, where the produced fuel will be used for the main production. The “tailings” of stable composition with minimum carbon content formed in the process of enrichment should be used as the main raw material for the production of ceramic brick.

As part of the program on import substitution [6] for the industry of ceramic wall materials the development of domestic technology and equipment, focused on high quality products, with regard to the characteristics of technogenic ceramic raw material from waste coal, is of high importance.

The aim of this work was to develop rational methods for preparation of waste coal mixture for production of ceramic bricks of good quality by compression molding.

2. Rational preparation of raw material
Performance experience gained by the brick factories with the technology of semi-dry pressing showed low quality of products in terms of bending strength and frost resistance compared with the products of plastic molding. It was due to the insufficient grinding of raw materials (up to 3 mm). Heterogeneity of the press powder composition obtained from coarse-grained masses results in a low strength of adobe. The developed method of formation of optimal ceramic brick structures allows authors to avoid structural defects by fine dry grinding, granulation, compression molding, drying and firing [7].

Preparation of the activated fine grained press powders from raw material can be carried out by using modern mixers-granulators of intensive action. Due to the variation in the rotation speed of the blades the regulation of press-powder granulometry within wide limits is possible, and complex turbulent motion of the material ensures active mixing of the adobe components and uniform moisture distribution in the volume of the resulting granulate.

The rational size distribution is a decisive prerequisite for a good compaction and the compact packaging of granules largely determines the final average density of a semi-finished product and purposefully influences on its important properties such as strength, water absorption, frost resistance, etc. Research carried out with the use of traditional technology of semi-dry pressing technique showed that the adobe brick, containing 40-50% of fraction 3-1 mm, 10-20% of fraction 1-0.5 mm and 50-40% of fraction <0.5 mm, has the minimum porosity [8, 9]. We found [10] that the structure of adobe-brick from granulated fine masses with a predominant granule size 1-3 mm is significantly different from the adobe brick of semi dry pressing, produced by the traditional drying-grinding technology of mixture preparation, and gives results that are similar to the plastic molding.

3. Methods and materials
During planning and researching we developed the models of formation of adobe brick structures from granulated press powder (Figure 2). At the initial stage of compression molding of the material there is a reorganization and convergence of granules. Further, as the pressing powder increases, their plastic irreversible deformation with simultaneous air removal from the space between them takes place. It is found that at the optimal compression pressure during adobe compaction to plastic deformation of granules in the surface layer and at the interface between them the concentration of liquid phase occurs due to the squeezing of water from the central part of the granules body (Figure 2c).

The implementation of the described principles of rational mixture preparation for face wall ceramics of compression molding was carried out on the example of waste coal. We used waste coal with class + 13 mm from coal processing plant “Abashevskaya” in Kemerovo region, Russia after extraction of the carbon-containing part (about 7% of the concentrate with $Q=4500$ kcal/ kg). As a corrective additive lean loam raw materials (Novokuznetsk) was used.

Novokuznetsk loam is the most common type of clay rocks in Siberia. The raw material is low-disperse, non-caking, highly sensitive to drying, mineral composition refers to polymineral rocks of hydromica-kaolinite-montmorillonite type. Waste coal are similar to clays [11] and consist of mineral part represented by hydromuscovite, quartz, plagioclase, montmorillonite, chlorite, siderite, calcite, and carbon residue containing free carbon in the amount from 6 to 8% after the secondary processing.
According to the chemical composition (Table 1) both types of waste coal belong to the group of semi-acid raw material with a high content of coloring oxides.

Table 1. Chemical composition of waste coal (after processing) and corrective additives.

| Raw material | Mass fraction of components to the dried substance, % |
|--------------|-------------------------------------------------------|
|              | SiO$_2$ Al$_2$O$_3$ TiO$_2$ Fe$_2$O$_3$ CaO MgO K$_2$O Na$_2$O LOI |
| Waste coal   | 57-64 14-17 0.5-1 5-6 3-5 1.5-2 2-2.5 1.5-2 6-8 |
| Loam         | 61-64 14-15 0.8 4-5 4-4.5 2-2.5 3.5-4 5-6 |

Drying and milling of raw materials were carried out in a vortex mill-dryer USP-S-04.55M to class -0.3+0. Evaluation of a particle size distribution after grinding of the material was conducted on a laser granulometer Malvern Mastersizer 2000, the results are shown in Table 2.

Table 2. Particle size distribution of waste coal and corrective additives.

| Raw material | Fractions content in %, particle size in mm |
|--------------|---------------------------------------------|
|              | 0.315-0.1 0.1-0.05 0.05-0.01 0.01-0.005 0.005-0.001 <0.001 |
| Waste coal   | 0.02 1.16 25.11 19.80 21.41 32.50 |
| Loam         | 16.37 36.25 34.81 3.2 4.73 4.64 |

By particle size distribution waste coal refer to coarse dispersed raw material with a predominance of sand fraction; by ductility – low plastic (plasticity $\approx$ 6.0); by drying properties – insensitive to drying; by the degree of caking – non-caking material.

Granulation of fine powders was carried out in the intensive mixer to form granules of diameter 1-3 mm with simultaneous moisturisation to the molding moisture. In the process, we determined the optimal parameters for obtaining the granulated mixtures.

In order to improve the product sintering ensured by the formation of the ceramic matrix structure [12], the granulated waste coal was covered with a layer of fine loam. The granulation parameters, material composition and particle size distribution of ceramic mixtures are given in Table 3.
Table 3. Parameters of ceramic mixtures granulation.

| No. | Mixture composition | Content, W, % | W, V, s^-1 | Partial residuals in % on sieves, mm |
|-----|---------------------|---------------|-------------|-------------------------------------|
|     |                     |               | 5 | 2.5 | 1.2 | bottom |
| 1   | Loam               | 100           | 11.0 | 125.6 | 1.5 | 14.5 | 72.8 | 11.2 |
| 2   | Waste coal + Loam  | 80 + 20       | 16.4 | 157.1 | 2.5 | 40.3 | 33.7 | 23.5 |

Laboratory studies showed that the necessary amount of water for powder granulation is 10-17 % [13]. These figures are comparable in moisture values to the extrusion molding technology, therefore, when characterizing the method, generally called “semi-dry pressing”, for the developed method of brick production from granulated mixtures it will be more correct to use the term “compression molding”.

The optimization of ceramic mixtures compositions from granulated dispersed powders on the basis of waste coals was performed on samples of 50 mm in diameter and height 45-50 mm, the pressing pressure 12-15 MPa, with a two-sided load application. The samples were fired in the muffle furnace at a maximum temperature 1000-1050 °C. The macrostructure of the obtained ceramic wall materials and the test results of their physical and mechanical properties are presented in Figure 3 and Table 4.

**Figure 3.** The macrostructure of ceramic products from granulated mixtures of the following composition (%): loam – 100 (a); waste coal – 80 + loam – 20 (b)

Table 4. Physico-mechanical properties of ceramic samples.

| Mixture No. | Compressive strength, MPa | Average density, kg/m³ | Water absorption, % | Frost resistance, cycles | Strength-density ratio |
|-------------|---------------------------|-------------------------|---------------------|-------------------------|-----------------------|
| 1           | 19.3                      | 1670                    | 11.6                | >50                     | 11.5                  |
| 2           | 18.7                      | 1680                    | 18.5                | 50                      | 16.4                  |

4. Results and discussion

The study on phase composition and microstructure of ceramic samples from waste coal was carried by the complex of methods that includes X-ray phase analysis, petrography of thin sections, SEM, etc. The structure of the ceramic cork produced from waste coal is fine grained, in the granules and on their borders there are non-transparent segregations of brown and dark colour, clearly visible in the parallel and cross polarized light (Figure 4). The burn-out of residual carbon in the granules reacting with oxygen, results in the chemical reactions of reduction inside the material. As a result, during firing in the temperature range 900-950 °C the melt is formed on the boundary and in the central part of the granules.

In the boundary layer the macro pores of irregular form elongated along the granules are observed. The disperse phase of the composite material formed from the granules after firing contains conglomerates of fine cryptocrystalline particles sintered together and permeated by micropores. In the
body of granules (Figure 4) a significant number of quartz grains of angular shape can be noted. The boundary layer of composite matrix is a cryptocrystalline aggregate continuously transition from one granule into another. It consists mainly of evenly distributed in the in the glass phase of microscopic crystals and fragments of transparent yellowish-white minerals, and fine cryptocrystalline aggregates, predominantly of brownish-red color (Figure 6). On the border of granules there is an intensive formation of glass phase, contributing to the reaction flow and reinforcing binding effect of the matrix.

**Figure 4.** Micrographs of the ceramic samples matrix structure containing waste coal of secondary processing from coal processing plant “Abashevskaya”. Thin section, transmitted light, 25×, Nicols II (a); Nicols + (b): 1 – the boundary layer of granules; 2 – granule body; 3 – non-transparent segregations of brown and dark color; 4 – melted silica particles; 5 – pores.

Examination of the ceramic crock form granulated waste coal by the scanning electron microscope also revealed differences in the material structure over the surface of granules contact. At a greater magnification (Figure 5) the melted matrix microstructure containing closed, circular or oval pores (1-10 µm) compared with the disperse phase can be seen, which is consistent with the data of optical microscopy. Besides, in the boundary layer there are macropores of irregularly form, elongated along the granules perimeter.

**Figure 5.** SEM micrograph of dispersion medium areas (a, b) and disperse phase (c, d) of the ceramic matrix composite containing waste coal of secondary processing from “Abashevskaya” plant: 1 – boundary layer; 2 – body of a granule; 3, 4 – closed pores of round, oval and irregular shapes; 5 – melted grains of feldspar.
Thus, in case of use of fine dispersed waste coal as an aggregated filler and actively sintering clay component as a material forming the matrix, we obtain the matrix structure of a ceramic crock, which is a system of nuclei of non-caked material and the dense coked shell, providing high strength and performance characteristics of wall ceramics.

5. Conclusions
The studies conducted on the example of waste coal from coal processing plant “Abashevskaia” field proved the possibility of a complex use of waste coal as the main raw material (up to 80 wt.% in the mixture composition) for the production of ceramic bricks after their secondary processing and mixture preparation by the developed method. High physico-mechanical characteristics of the product are achieved due:

- prior fine grinding to class -300 µm of carbonized, coarse technogenic raw material to achieve high uniformity of the molding press-powder and eliminate the harmful effects of carbonate inclusions;
- aggregation of waste coal fine powders into granules to achieve their greater compaction rate and exclusion of air pressing during molding;
- rational particle size distribution of molding mixtures with a predominant granule size 1-3 mm, that allows a uniform structure of a capillary-porous body to be obtained.

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