Production technology of ceramic roof tiles based on raw materials of Ural region

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Abstract. The technology of production of ceramic roof tiles on the basis of low-melting red-burning clay, namely on the basis of clay from the Sadovoye deposit of the Sverdlovsk region, is proposed. The samples of ceramic roof tiles, molded by plastic and semi-dry way, were obtained with the properties that meet the requirements of Russian standards and correspond to the quality of the Röben ceramic roof tiles. The water absorption of such products is 6.2–6.9%, the apparent density is 2.19–2.31 g/cm³, the products are waterproof for more than 4 hours, and withstand frost resistance for more than 150 cycles. The production technology involves the use of high-speed firing way in the temperature range of 1150–1200°C. The production technology provides for grinding the clay to a grain size of less than 0.3 mm. Molding of products can be done both in a plastic or semi-dry way. The use of high-speed firing is allowed. There is no destruction of products made on the basis of Sadovoye deposit clay.

1. Introduction
Ceramic roof tile has significant advantages over other types of roofing not only in terms of architectural expression, durability, but also in other physical and mechanical characteristics. Ceramic roof tile is considered as the environmentally friendly roofing material, contributing to the creation of a favorable microclimate in the house. The raw material for the ceramic roof tiles production is a natural material – clay. Ceramic roof tile belongs to the class of non-combustible materials. It should be viewed as durable, strong, fire-resistant roofing material, characterized by great architectural merit [1-8].

Currently, the modern building materials market of the Russian Federation includes ceramic roof tiles from various manufacturers, such as: Terreal (plants Tuiles Lambert, TBF and Guiraud Frères – the oldest French brands in the production of ceramic tiles and bricks), Röben (Robin or Rebon) [9] in Sroda – Slinska (Poland), CREATON is one of the largest European manufacturers of ceramic tiles (Germany), Ludowici – USA, Tejas Cobert Company (Spain, Portugal), KORAMIC – Austrian concern “Wienerberger”. The cost of such roof tiles is 20–76 $ per 1 m².

At present, the production of ceramic roof tiles in the CIS countries is about 15 million units per year. The demand for a modern building complex of the Russian Federation in ceramic roof tiles is currently not met by domestic manufacturers. There are papers concerning the production of roof tiles on the basis of waste [10-11]. In [12-14] the ways to improve the properties of ceramic roof tiles due
to various technological parameters are proposed. Clays of the Ural region have been studied in many papers [15-18] to produce ceramic bricks, expanded clay, refractories and glass.

The production technology of ceramic roof tiles is similar to the production of ceramic tiles. The cost of such tiles, produced in the Urals, ranges from 5–13 $ per m². The cost of ceramic roof tiles should be the same. There is still a large issue regarding the production of ceramic roof tiles which are not inferior in their operational qualities to world analogues. The aim of this work is the developing of the production of ceramic roof tiles based on local regional raw materials.

2. Experimental Procedure

There is a national standard in Russia [19]. This standard applies to ceramic roof tiles used for roofing, and establishes the technical requirements, acceptance rules and test methods for roof tile. As a reference for comparing the properties of the obtained tiles in the laboratory based on local raw materials, Röben's tiles of two types were used. The properties of clay raw materials are determined in accordance with [20].

To determine the mineral composition of clays, differential thermal (DTA) and X-ray phase analysis (XRF) were used. Thermogravimetric studies were performed on a STA 449 F3 Jupiter differential scanning calorimeter (Netzsch-Geratebau GmbH) according to DIN 51004: Determination of melting temperatures of crystalline materials using differential thermal analysis in air environment, in the temperature range of 20–1000°C. According to the mass loss in DTA, a predictive analysis of the mass loss kinetics of materials was performed using the Kinetic3 program (Netzsch-Geratebau GmbH). X-ray phase analysis was performed on a MiniFrlcx 600 rotary anode diffractometer (CuKα radiation, λ = 1.541862 Å, shooting interval – 3–90°, scanning step – 0.02°) “Rigaku – Carl Zeiss” (Japan) with control programs and MiniFlex guidance data acquisition and PDXL Basic data processing package. The diffraction peaks were identified using the JSPDS databank. To determine the chemical analysis, we used emission spectral analysis with inductively coupled plasma on the Optima 4300 DV (PerkinElmer, USA).

3. Results and Discussion

The properties of ceramic roof tiles produced by Röben (table 1) were studied in order to compare their properties with the obtained samples based on clay from the Sadovoe deposit. The water absorption of “Röben” roof tile is 6–8.5%, the apparent density is 2.05–2.17 g/cm³. Ceramic roof tile “PIEMONT havanna braun” (Środa Śląska, Poland) is covered with brown engobe. It is used in the roofing of a sloped roof with a minimum slope of 22°. The red roof tile “LIMBURG rot” (Brüggen, Germany) is also covered with a terracotta-colored engobe. It is used in the roofing of a sloped roof with a minimum slope of 30°.

| Table 1. Properties of „Röben“ ceramic roof tile. |
|-----------------------------------------------|
| Roof tile type                  | PIEMONT havanna braun | LIMBURG rot |
| Water absorption (%)           | 6.6                   | 8.4           |
| Open porosity (%)              | 14.3                  | 17.2          |
| Apparent density (g/sm³)       | 2.17                  | 2.05          |

To produce ceramic tiles in the Sverdlovsk region it was proposed to investigate the properties of Sadovoe deposit clay. The chemical composition of clay is given in table 2. According to the content of Al₂O₃ in the calcined state, the clay of the Sadovoe deposit belongs to semi-sour raw materials (14.98%) with a high content of coloring oxides (7.68%).

According to the dispersed analysis (table 3), the clay of the Sadovoe deposit is coarsely dispersed, some large inclusions in the clay reach sizes greater than 10 mm.
Table 2. The chemical composition of Sadovoye deposit clay.

| Matter type   | SiO₂   | Al₂O₃ | Fe₂O₃ | CaO | MgO | TiO₂ | K₂O | Na₂O | LOI  | ∑     |
|---------------|--------|-------|-------|-----|-----|------|-----|-----|------|-------|
| Calcined matter | 69.32  | 14.98 | 7.21  | 0.50| 0.68| 0.47 | 2.25| 4.10 | –    | 99.51 |
| Dry matter    | 66.78  | 14.43 | 6.95  | 0.49| 0.65| 0.45 | 2.16| 3.95 | 3.65 | 99.51 |

Table 3. Dispersed analysis of Sadovoye deposit clay.

| Particle size (mm) | > 0.06 | 0.06–0.01 | 0.01–0.005 | 0.005–0.001 | < 0.001 | The content of the fraction (wt.%) |
|--------------------|--------|-----------|------------|-------------|---------|---------------------------------|
|                    | 51.89  | 24.84     | 11.64      | 3.87        | 7.76    |                                 |

According to the quantity, size and type of coarse-grained inclusions, the clay raw material from the Sadovoye deposit belongs to the group of raw materials with a high content of coarse-grained inclusions (40.4 wt.%). The predominant grain size in clay is more than 5 mm (57.8 wt.%), which corresponds to the group of raw materials with large inclusions.

According to DTA and XRF, the clay under consideration belongs to kaolin-montmorillonite one. By plasticity, this clay refers to low-plastic raw material with a plasticity number of 6. The Sadovoe deposit clay is insensitive with a sensitivity coefficient of drying of 0.20. No cracks and defects were found on the clay samples after drying.

The properties of the samples molded by the plastic method at a relative humidity of 24% and calcined in the temperature range of 1000–1200°C are given in table 4. The samples that annealed in the temperature range of 1000–1150°C have overestimated water absorption – 25.9–17.9% in comparison with the roof tile produced by Röben – 8 and 6% (table 1). However, with the firing temperature increase to 1200°C, the water absorption indicators decrease – to 17.9 and 6.9%. The apparent density increases from 1.61 to 2.22 g/cm³. The density of the obtained roof tile samples that are correspond to the water absorption of roof tiles PIEMONT havanna braun (Röben), is slightly higher than 2.22 g/cm³ compare the water absorption of roof tiles of Röben (table 1) – 2.17 g/cm³.

Table 4. The properties of plastic molding samples.

| Firing temperature (°C) | 1000 | 1050 | 1100 | 1150 | 1200 |
|-------------------------|------|------|------|------|------|
| Complete shrinkage (%)  | 6.7  | 7.1  | 7.2  | 7.5  | 8.0  |
| Water absorption (%)    | 22.0 | 20.3 | 19.0 | 10.1 | 6.9  |
| Apparent density (g/sm³)| 1.61 | 1.71 | 1.72 | 2.07 | 2.22 |

The high degree of leaning of Sadovoye deposit clay raw materials (40.4%) suggests that this clay is good to obtain roof tile using semi-dry molding method. According to preliminary research it was established that the moisture content in semi-dry molding should be 6–10%. It was determined the pressing pressure at a relative humidity of 10%. The clay was pre-crushed to a grain size of less than 0.3 mm. Then it was established the firing temperature of roof tiles based on Sadovoye deposit clay.

To determine the pressing pressure, the Berezhnoy–Pokrovsky equation was used:

\[ P_t = a - b \log P \]  

(1)

where \( P_t \) – true porosity; \( P \) – pressing pressure, kg/sm².

The dependence of the true porosity \( (P_t) \) of samples on the pressing pressure \( (P) \) in the \( P_t–\log P \) coordinates is shown in figure 1. The true porosity decreases with an increase in the pressing pressure up to 225 kg/cm². At the same time, the critical pressing pressure is not achieved, where the loosening of the structure is observed. Thus, the recommended pressure for molding samples is 100–225 kg/cm².
Figure 1. The dependence of true porosity on the pressing pressure.

Thus, the samples were molded at a moisture content of 10% and a pressing pressure of 175–225 kg/cm². Samples were fired in the temperature range of 1000–1200°C. Properties of samples are presented in table 5.

Table 5. The properties of clay samples molded by semi-dry method.

| Pressing pressure (kg/sm²) | Firing temperature (°С) | Complete shrinkage (%) | Water absorption (%) | Apparent density (g/sm³) |
|---------------------------|-------------------------|------------------------|----------------------|-------------------------|
| 225                       | 1000                    | 5.5                    | 19.3                 | 1.83                    |
|                           | 1100                    | 6.0                    | 15.2                 | 1.95                    |
|                           | 1150                    | 6.1                    | 11.9                 | 2.03                    |
|                           | 1200                    | 6.6                    | 2.0                  | 2.32                    |
|                           | 1000                    | 5.4                    | 20.4                 | 1.80                    |
| 200                       | 1100                    | 5.9                    | 15.8                 | 1.90                    |
|                           | 1150                    | 6.0                    | 12.3                 | 2.00                    |
|                           | 1200                    | 6.4                    | 2.1                  | 2.31                    |
|                           | 1000                    | 5.3                    | 21.0                 | 1.76                    |
| 175                       | 1100                    | 5.8                    | 16.2                 | 1.93                    |
|                           | 1150                    | 5.9                    | 12.5                 | 1.99                    |
|                           | 1200                    | 6.2                    | 2.2                  | 2.30                    |

As to table 5, the water absorption of less than 8.5% is achieved by sample firing at a temperature of 1200°C, that are molded at a pressing pressure of 175–225 kg/cm², while the apparent density is 2.30–2.31 g/cm³. Shrinkage of samples in this case is 6.2–6.6%.

The use of high-speed firing in the roof tile production is an important issue in connection with the possible increase in productivity by reducing the firing time of products from 36 hours to several hours or even minutes.

Hence the samples were molded at a molding moisture of 10%, and a pressing pressure of 225 kg/cm2. Samples of roof tile were fired in the temperature range of 1160–1210 °C. The properties of samples are presented in table 6.

Table 6. The properties of samples fired at high-speed method.

| Firing temperature (°C) | 1160 | 1170 | 1180 | 1190 | 1200 | 1210 |
|-------------------------|------|------|------|------|------|------|
| Open porosity (%)       | 28.6 | 28.3 | 25.2 | 25.2 | 19.2 | 14.5 |
| Water absorption (%)    | 14.6 | 14.5 | 12.6 | 12.6 | 8.9  | 6.6  |
| Apparent density (g/sm³) | 2.00 | 1.90 | 2.00 | 2.00 | 2.17 | 2.19 |

Data in table 6 show that when using high-speed firing way, the properties of the samples deteriorate slightly. Therefore, it is possible to obtain roof tile samples corresponding to the properties
of Röben production, when fired at a temperature of 1210°C. Given this, the water absorption is 6.6% and an apparent density is 2.19 g/cm³. The samples fired at a temperature of 1210°C are waterproof for more than 4 hours and withstand more than 150 freeze-thaw cycles.

4. Conclusion
Consequently, the possibility of obtaining of ceramic roof tiles based on Sadovoye deposit clay was established. The sample properties are similar to those made by Röben production. At the same time the samples of tiles have been molded both by plastic and semi-dry ways. The possibility of using high-speed burning ways when producing ceramic roof tiles based on Sadovoye deposit clay was established.

When using the plastic molding method, samples were obtained with a water absorption of 6.9% and an apparent density of 2.22 kg/cm³; the firing temperature was 1200°C.

When using the semi-dry method of molding, the following parameters were used: humidity 10%, pressing pressure 175–225 kg/cm², while samples of ceramic roof tiles have a water absorption of 6.2–6.6% and apparent density of 2.30–2.31 g/cm³. In addition, with the use of high-speed burning way for obtaining ceramic roof tiles based on Sadovoye deposit clay, the samples were obtained with a water absorption of 6.6% and an apparent density of 2.19 kg/cm³ during firing at a temperature of 1210°C.

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