Lightening of ceramic bricks based on red-burning clay

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Abstract. To determine the possibility of using a mineral pigment based on the rutile form of titanium dioxide in the production of volume-colored ceramic brick in order to obtain lighter brick tones the studies are conducted. The previously studied low-melting clay of the Tyumen deposit was used as a basis. It was established that significant lightening of the brick does not occur even with the introduction of 8% of the specified pigment, although some lightening is appeared. In addition, with the pigment introduction, an increase in water absorption of bricks is observed, which leads to a decrease in the sample strength. Increasing the firing temperature to 950°C does not lead to an improvement in the quality characteristics of the samples, and even vice versa makes them worse. Thus, the use of a white pigment under investigation does not give a significant effect of lightening of ceramic red-burning clay.

1. Introduction

One of the directions of development of the facial ceramics production is the expansion of the range of output goods due to the variety of colors. This, in turn, determines the relevance of research aimed at obtaining facial ceramic products of less common tones. An important issue is the lightening of brick clays in order to obtain a lighter base, which could be colored in any color. When using red-burning clays, it is almost impossible to change the color of the products to lighter ones [1-3].

There are several ways to obtain colored ceramic bricks. The main method is the volume coloring of ceramic masses with various chromophore additives (chemical and mineral). It should be noted that this method, compared with, for example, with glazing or engobing, has an advantage: the brick is more durable, and the method itself does not require the technological treatments that are not typical for brick technology. In addition, the use of engobing or glazing bricks, as a rule, leads to the separation of the vitreous glaze or engobic coating from the base of the brick directly in the building structure, especially in regions with large temperature drops. At the same time, the volume coloring of the product, especially in the case of obtaining saturated color tones, requires an increased consumption of coloring additives, which increases its cost. Reducing the cost of the product in this case is possible due to the use in the masses of by-products containing sufficient quantity of color-forming oxides necessary to give the material the desired color. Such materials include iron-containing dust from the electric steelmaking production of metallurgical enterprise [4-13], materials based on dicalcium silicate (nepheline sludge) [14-15]. Nevertheless, when using waste, it is only possible to obtain a darker color than the fired base itself.

Currently, there is a large variety of ceramic pigments for volume coloring of ceramic bricks. Pigments are highly dispersed substances, insoluble (as opposed to dyes) in water, organic solvents, film-forming agents and other media that have a certain set of optical, mechanical, sorption properties.
To produce ceramic products, because of high firing temperatures (up to 1000°C), inorganic pigments are usually used. The choice of pigments is determined by the properties of pigment and material, the nature of their interaction, as well as the processing parameters and service conditions of products.

Basically, in the production of wall ceramics, low-melting raw materials are used, which after firing is red. The color of ceramic bricks based on low-melting red-burning clay is predetermined by the amount of iron oxides, the phase composition and the crystal-chemical state of iron ions in its structure. The presence of iron oxide in the form of an independent hematite phase $\alpha$-$\text{Fe}_2\text{O}_3$, having the lowest reflection coefficient of 6.5%, causes an intense red or red-brown color. The distribution of iron in the glass phase or in aluminosilicate phases with the formation of solid solutions of substitution $\text{Al}^{3+} \leftrightarrow \text{Fe}^{3+}$ increases reflection coefficient and contributes to a certain degree of neutralization of color and lightening of brick.

When fired in oxidizing conditions, the brick color is red or red-brown, due to hematite $\alpha$-$\text{Fe}_2\text{O}_3$ of red color. During reductive firing, the color of the brick is dark brown or black, which is predetermined by the formation of ferrospinel phases containing $\text{Fe}^{2+}$ and $\text{Fe}^{3+}$ ions, mainly magnetite $\text{Fe}^{3+}\text{Fe}^{2+}\text{O}_4(\text{Fe}_3\text{O}_4)$ of black color in combination with fayalite and hematite [16-19].

The lightening of ceramic stone using raw materials with a high content of iron oxide occurs when a significant amount of calcium carbonates is introduced into the mixture, which ensures the formation of anorthite reducing the coloring intensity of ceramic stone. However, there is instability of color, a significant decrease in strength, and sometimes the lack of lightening. In the technology of ceramic bricks, low-temperature (up to 1000°C) firing in an oxidizing environment is most common, which determines its physical and technical properties and one of the most important indicators – color. In addition, titanium dioxide is also used to lighten ceramics, which is one of the most important inorganic materials, the unique properties of which determine technical progress in many sectors of the world economy.

To obtain a brick of light beige or pink color, it is necessary to utilize the clay raw materials with an $\text{Fe}_2\text{O}_3$ content of not more than 2% and an $\text{Fe}_2\text{O}_3$ content of more than 4% – for a red or red-brown color with the required use of oxidative firing.

Thus, the lightening of ceramic bricks occurs because of the formation of a phase composition, which as a result of various oxidation-reduction conditions leads to the formation of high colored iron-containing phases, including black ones, of magnetite structure of iron-containing solid solutions [20].

2. Experimental procedure

To obtain colored samples of facial bricks, a previously studied brown clay sample from the Tyumen deposit was used as a base [21]. Mineral pigment white titanium dioxide with a $\text{TiO}_2$ content of 93% was used as dyes. It is a rutile pigment based on titanium dioxide with surface treatment with aluminum and silicon compounds.

The quality assessment of the obtained brick samples was determined in accordance with [22]. Frost resistance was determined indirectly by the method described in [23] as quotient $\text{B}_c/\text{B}_h$, where $\text{B}_c$ is a water absorption of samples determined according to [24], $\text{B}_h$ – water absorption determined after boiling for 4 hours with further cooling to room temperature. Samples with a quotient less than or equal to 0.85 are frost-resistant.

To obtain the volume-colored samples the plastic ceramic mass with the pigment in the amount of 2, 4, 6, 8% was prepared. The pigment was introduced into the mass after diluting it in water, which is necessary for wetting the mass to a molding moisture of 19%. Samples-cylinders of size 36×36 mm were formed. Further, samples were air dried and annealed at 850, 900, 950°C. After firing, the sintering degree, ultimate compressive strength, as well as indirect frost resistance of the obtained samples were determined.

3. Results and discussion

The test results are shown in table 1 and in figures 1–4.
Table 1. Post-firing properties of the samples.

| Parameter                  | Firing temperature (°C) | 0   | 2   | 4   | 6   | 8   |
|----------------------------|-------------------------|-----|-----|-----|-----|-----|
| Frost resistance coefficient| 850                     | 0.91| 0.95| 0.94| 0.92| 0.93|
|                            | 900                     | 0.89| 0.95| 0.96| 0.94| 0.95|
|                            | 950                     | 0.88| 0.94| 0.95| 0.95| 0.95|
| Apparent density (g·cm⁻³)  | 850                     | 2.00| 2.00| 2.00| 2.00| 2.00|
|                            | 900                     | 2.01| 1.97| 1.95| 1.93| 1.92|
|                            | 950                     | 2.04| 2.03| 2.01| 1.96| 1.92|
| Water absorption (%)       | 850                     | 12.0| 12.7| 13.2| 13.2| 13.2|
|                            | 900                     | 11.6| 12.8| 13.3| 14.2| 14.4|
|                            | 950                     | 10.3| 12.6| 13.2| 14.7| 15.7|
| Compressive strength (MPa)| 850                     | 16.5| 14.3| 8.1 | 7.0 | 6.8 |
|                            | 900                     | 13.2| 13.1| 8.4 | 7.8 | 7.7 |
|                            | 950                     | 18.4| 11.9| 8.3 | 6.5 | 5.0 |

Table 1 shows that the pigment introduction leads to water absorption increasing and the strength decreasing of the samples. In addition, the frost resistance coefficient increases, which indicates a decrease in frost resistance of the samples. With an increase in the pigment concentration at the firing temperature of 850°C, the apparent density of the samples remains unchanged, although there is a decrease in the strength. A subsequent increase in the firing temperature does not increase the samples strength, but on the contrary, the structure is loosened with an increase in the pigment concentration and with an increase in the firing temperature.
Figure 3. A water absorption of volume-colored ceramic brick samples.

Figure 4. A compressive strength of volume-colored ceramic brick samples.

Figure 5 shows the dependence of sample color on the pigment content at different firing temperatures: 850°C (a), 900°C (b) and 950°C (c).

Figure 5. The effect of pigment content on the color of brick samples.

Lightening of brick samples is more intense with increasing pigment content. The firing temperature in the range of 850–950°C has almost no effect on the sample color.

4. Conclusion

Thus, it was found that the use of pigment based on the rutile form of TiO₂ in clay masses to produce clarified bricks does not give a positive result. Lightening occurs lightly even at a pigment concentration of 8% and at the same time the technical characteristics of the brick are reduced. An increase in the firing temperature up to 950°C also does not lead to an improvement in brick properties, on the contrary, the strength and frost resistance are decreased.
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