# PROTECTIVE COATINGS FOR CONSTRUCTION CERAMICS AS FACTOR OF INCREASING ENERGY EFFICIENCY AND OPERATIONAL PROPERTIES OF PRODUCTS

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**Abstract.** The production of building materials is important for any sector of economy, as they are widely used to create residential, commercial and industrial structures. This is especially true for products such as ceramic bricks, as their environmental friendliness and energy efficiency make them virtually indispensable. However, the higher the energy efficiency of ceramic bricks, the lower their strength and durability. This is due to the porous structure of the material – water gets into the open pores, which causes gradual destruction as a result of freezing and thawing and changes in volume. To reduce the destruction of the outer layer, decorative and protective engobe coatings have been developed that help seal the outer surface of products and prevent moisture from seeping inside. The compositions, rheological and technological as well as physical and mechanical properties of engobe coatings for ceramic bricks with a firing temperature of 950 ºC are presented. It is established that the presence of engobe on the surface of the brick, with its rather porous internal structure, allows to reduce water absorption from 14.8 to 3.2% and increase the frost resistance of products from 15 to 65 cycles. The coating opens up the possibility of significantly extending the service life of the brick.

**Keywords:** ceramic brick, protective coating, frost resistance, thermal conductivity, strength.

## Introduction

Ceramic bricks are one of the most popular materials in private and industrial construction. The use of bricks allows for a good indoor climate in countries with both hot and cold microclimates [1; 2]. Ceramic bricks are durable and fire-resistant [3], which makes them stand out among other building materials and opens up wide prospects for their use in energy-efficient construction [4-6].

Buildings of the future with high energy efficiency can play a crucial role in addressing the challenges of sustainable energy [7]. It should be noted that heat loss through walls accounts for about 50% of the total energy consumption of buildings, making energy saving a critical issue in the construction sector. Therefore, minimising energy consumption in buildings through passive methods, such as thermal insulation, is becoming increasingly popular [8]. Since the European standard EN 823 requires that the thermal conductivity of walls should be 0.4-0.7 $W \cdot (m^2 \cdot K)^{-1}$ [4], ceramic bricks are among the best options.

The most common ceramic brick is made on the basis of low-melting clays and loams, contains burning additives and is fired at relatively low temperatures (950-1000 ºC). It has a rather porous structure with water absorption of 12-16% and higher [9-11]. Waste from the agricultural sector, such as rice husks [12], sugar cane ash [13], oil refining waste [14], and others, play an important role in the production of lightweight porous bricks as burning additives. Such bricks have high energy efficiency in building structures due to their low thermal conductivity (thermal conductivity coefficient is 0.18-0.25 $W \cdot (m^2 \cdot K)^{-1}$), but their durability is unsatisfactory.

This is explained by several factors. The most significant is related to the high porosity of such bricks [15]. The moisture in the atmosphere in the form of precipitation gets into the pores of the brick and freezes and thaws when the temperature changes. This changes the volume of water in the pores, which destroys the ceramic tile due to the occurrence of microstresses [16]. In addition, the access of water into the brick through the pores and capillaries leads to the appearance of efflorescence - soluble salts that also destroy the structure of the brick [17; 18]. In addition, saturation of the brick with water leads to a decrease in its thermal insulation properties, which is extremely irrational in the current conditions of energy shortage.

Therefore, it is important to increase the density of the outer layer of the brick while maintaining its porous structure inside. The most appropriate is the use of an engobation operation, which consists in applying a protective coating to the front surface of the brick [19; 20]. The coating can be applied to a raw or dried semi-finished product, after which the brick is fired once according to a specified mode.

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No additional energy consumption is required for firing the engobed products, but a significant increase in their operational, aesthetic and decorative properties can be achieved.

The role of engobe on the brick surface is that during firing it sintered to significantly lower water absorption values than the ceramic base. This is facilitated by a specially selected coating composition, which contains an increased number of melts and ensures intensive liquid-phase sintering of the material [21]. However, the main problem with developing engobe coatings for energy-efficient bricks fired at low temperatures is the limited availability of low-melting compounds and the difficulty of coordinating shrinkage processes between the coating and the ceramic base.

Therefore, in this paper, it will be relevant to consider the principles of developing protective coatings for low-temperature fired building ceramics, the use of which will increase the energy efficiency and performance properties of products.

## Materials and methods

A typical ceramic mass based on clay materials from the Sursko-Pokrovsky deposit (Ukraine), which is used to make ordinary bricks, was chosen for the study, wt%: fusible clay – 40, loam – 50. Waste from the coal preparation of the Zakhidnodonbasskoye deposit developed by DTEK Pavlohradvuhillya was added as a burning additive (10 wt%). The chemical composition of the ceramic mass components is shown in Table 1.

Table 1

**Chemical composition of ceramic mass components, wt%**

| Name of raw materials | $SiO_2$ | $Al_2O_3$ | $Fe_2O_3$ | $TiO_2$ | CaO | MgO | $Na_2O$ | $K_2O$ | L.d.c. |
|---|---|---|---|---|---|---|---|---|---|
| Fusible clay | 52.25 | 20.32 | 7.07 | 1.10 | 5.65 | 0.85 | 1.69 | 2.06 | 9.00 |
| Loam | 68.80 | 13.25 | 2.65 | 1.09 | 3.56 | 1.04 | 0.66 | 1.95 | 7.00 |
| Burning additive | 47.82 | 19.51 | 12.95 | 0.75 | 3.68 | 0.11 | 0.69 | 1.06 | 18.43 |

The ceramic raw material mixture was crushed until it passed through a 1600 mesh·cm$^{-2}$ sieve, filled with 22% water, and carefully averaged. The plastic mass was used to form specimens in the form of bricks measuring 50x30x10 mm. The samples were coated with experimental engobe coatings, dried and fired. The firing temperature was 950 ºC.

The following raw materials were selected for the preparation of the engobe coatings, wt.%: refractory clay of the Druzhkovka deposit (Ukraine) 35-65, container glass 10-25, quartz sand of the Avdiivka deposit (Ukraine) 10-25. The chemical composition of the engobe coating components is given in Table 2.

Table 2

**Chemical composition of engobe components, wt.%**

| Name of raw materials | $SiO_2$ | $Al_2O_3$ | $Fe_2O_3$ | $TiO_2$ | CaO | MgO | $Na_2O$ | $K_2O$ | L.d.c. |
|---|---|---|---|---|---|---|---|---|---|
| Refractory clay | 51.06 | 34.54 | 0.72 | 0.10 | 0.50 | 0.48 | 0.53 | 2.00 | 10,17 |
| Container glass | 73.33 | 3.47 | 0.62 | – | 2.88 | 3.70 | 16.10 | – | 0.10 |
| Quartz sand | 97.60 | 2.10 | 0.10 | 0.20 | – | – | – | – | 0.20 |

The engobe coatings were prepared by fine wet grinding in the form of slurries until they passed through a sieve of 8000 mesh·cm$^{-2}$. The moisture content of the slips was 43-44%. To determine the properties of the engobes, samples of 30x30x5 mm were prepared. To determine the coordination with the ceramic substrate, the engobe slips were applied to ceramic bricks, dried at 70 ºC in a drying oven for a day and fired in a muffle furnace at 950 ºC.

The flowability of the engobe slips was determined by measuring the time for the suspension to flow out of a 100 ml Ford cup through a 4 mm diameter hole after standing for 30 min. Shrinkage was determined by the change in linear dimensions of the sample in the freshly moulded state and after firing. Water absorption and porosity of brick and engobe samples were determined by the weight gain after vacuuming in water for 1 hour. The mechanical compressive strength of the samples was determined by measuring the destructive load per unit surface area. Frost resistance was determined by the number of cycles of alternate freezing to –15 ºC and thawing at + 15 ºC until signs of surface destruction of the samples appeared.

The temperature coefficient of linear expansion was determined using a Netzsch DIL 402 Expedis Classic dilatometer. The thermal conductivity coefficient of the ceramic samples was measured using the Netzsch HFM 436 Lambda instrument. Q-1500 D was used to study the thermal transformations in the burning additive. The study of the microstructure of the samples was carried out by the scanning electron microscopy on REM 106.

## Results and discussion

Ceramic masses based on low-melting clays and loams are widely used in brick production. They make it possible to obtain a strong stone-like body at relatively low temperatures, but the properties of the final product strongly depend on the composition of the ceramic mass (presence of porous additives, sintering components) and technological factors (grinding size of the mass components, moulding pressure, firing temperature) [4; 9]. In particular, the following properties were obtained for the experimental ceramic mass and fired samples after firing at 950 ºC (Table 3).

Table 3

**Main properties of the ceramic samples**

| Property name | Indicator |
|---|---|
| Total shrinkage, % | 9.1 |
| Water absorption, % | 14.8 |
| Open porosity, % | 29.1 |
| Mechanical compressive strength, MPa | 13.8 |
| Thermal conductivity coefficient, $W \cdot (m \cdot K)^{-1}$ | 0.28 |
| Temperature coefficient of linear expansion, $ºC^{-1}$ | $58 \cdot 10^{-6}$ |
| Frost resistance, freeze-thaw cycles | 15 |

The high water absorption (14.8%) and porosity (29.1%) of the fired ceramics are achieved by the presence of a burning additive in the mass of the additive – coal preparation waste. The nature of thermal transformations of this additive is shown in Fig. 1. The differential thermal analysis (DTA) curve records the amount of heat absorbed or released in a substance when it is heated, and the change in the mass of the substance is shown on the thermogravimetric curve (TG).

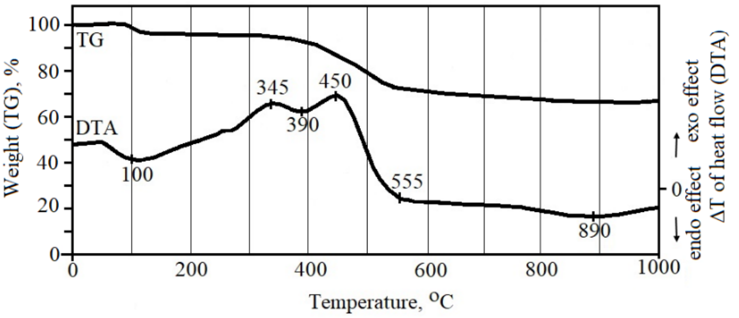


Fig. 1. **Results of differential thermal analysis of coal preparation waste**

There are two exothermic effects on the DTA curve with maxima at 345 and 460 ºC. These effects indicate the burning out of the organic component of the additive, which creates an additional thermal effect and saves fuel consumption for firing. In addition, the burning of organic matter with a decrease in the mass recorded on the TG curve creates additional voids in the structure of the ceramic tile, increasing its porosity and thermal insulation properties.

The mechanical compressive strength of the brick samples is 13.8 MPa (Table 3), which is a fairly high value for porous thermal insulation bricks [14; 21]. At the same time, the frost resistance indicators (15 cycles) at such a high porosity remain very low, which limits the use of these products in the construction of external structures.

To obtain a dense outer layer of ceramic samples, we prepared engobe coatings of the following composition (Table 4). The main properties of the engobe coatings are shown in Fig. 2.

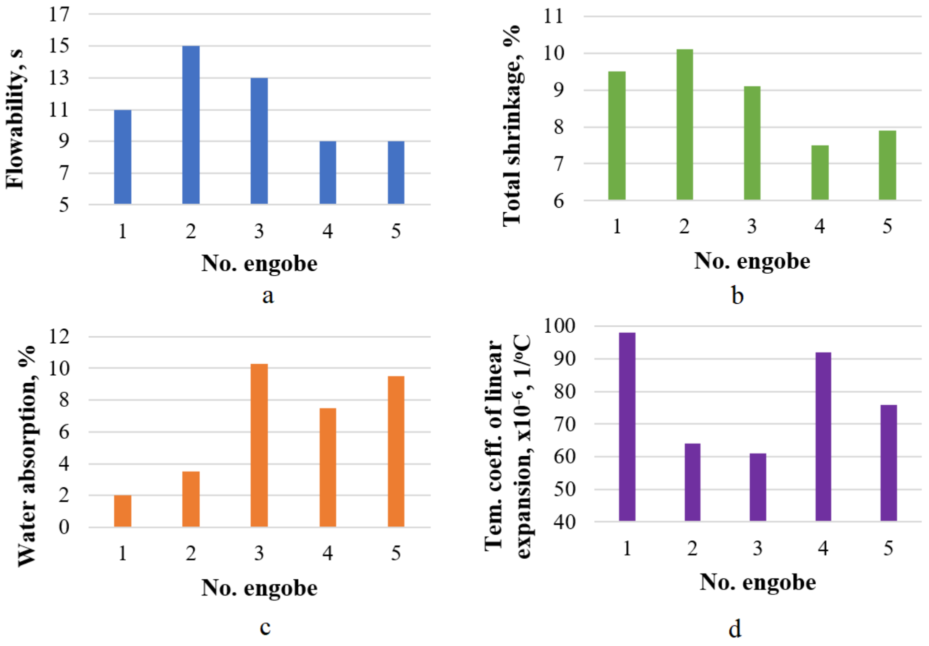


Fig. 2. **Main properties of the experimental engobe coatings:** a – flowability of the slips; b – total shrinkage; c – water absorption; d – temperature coefficient of linear expansion

The figure above shows that the flowability of engobe slips differs depending on the clay component content. Engobe No. 2 is more viscous, with a flow rate of 15 seconds, which allows the slip to spread evenly over the surface of the ceramic sample. Engobes No. 1 and 3 have satisfactory flowability (11-13 s). Engobes with a lower content of clay component No. 4 and 5 have a very low flow rate of 9 s, which leads to surface exposure when applying the slips to the ceramic sample. These engobes are also prone to delamination, which causes their heterogeneity and can lead to defects such as microcracks, chips, etc.

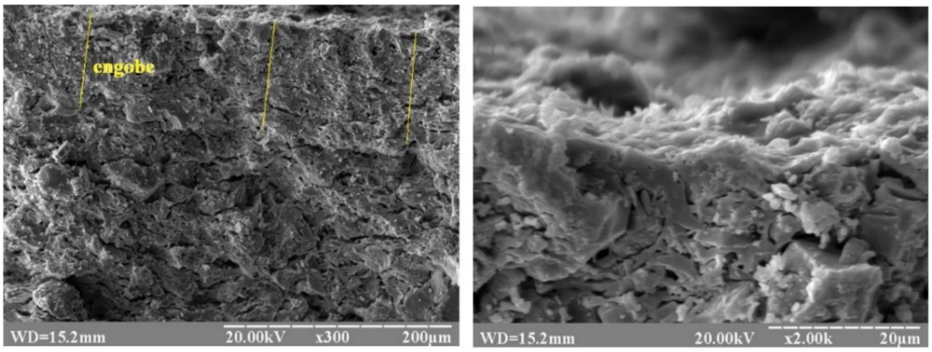
The shrinkage of engobes is no less important when they are matched to the ceramic mass [19; 20]. Thus, the closest in terms of total shrinkage to ceramic shards are engobes No. 1 and 2 (9.5 and 10.1%, respectively). Other engobes have a lower shrinkage (7.5-9.0%), which means that during drying and firing, shrinkage processes in the engobe and ceramic mass will occur with different intensities, which can contribute to the occurrence of microstresses and coating delamination.

The lowest values of water absorption after firing are characteristic of engobes No. 1 and 2 (2.0-3.5%). The main fluxing effect is performed by the glass cullet with a low content of quartz sand. However, for composition No. 1, a grid of cracks was observed on the surface, which arose due to a significant difference in the temperature coefficients of linear expansion of the ceramic mass and the engobe.

The thermal expansion values significantly higher than the ceramic mass ($58 \cdot 10^{-6}$ ºC$^{-1}$) are characteristic of engobes No. 1 and 4 ((98-92)$\cdot 10^{-6}$ ºC$^{-1}$). This is due to the high sodium calcium content of aluminosilicate glass cullet in the composition of the engobes, which are the source of the glass phase with a high expansion capacity. The compositions of engobes No. 2 and 3 differ in thermal expansion compared to the weight by no more than 10%, which is acceptable for ceramic coatings.

Thus, according to a set of indicators, the best in relation to the ceramic mass was engobe No. 2, which was applied to a ceramic sample of factory bricks and obtained the following indicators: water absorption of the front surface – 3.2%, frost resistance – 65 cycles of freezing and thawing, thermal conductivity coefficient 0.24 W$\cdot$(m$\cdot$K)$^{-1}$. Thus, the presence of a dense engobe layer (Fig. 3) made it possible to reduce water absorption of the outer layer from 14.8 to 3.2%, which increased the frost resistance index by more than 4 times.

A dense layer of engobe, 120-130 microns thick, is clearly visible on the porous coarse-grained tile. This layer has a protective effect and prevents moisture from penetrating into the ceramic brick. This prolongs the service life of the products in terms of frost resistance up to 65 freeze-thaw cycles. In addition, due to the presence of an additional engobe layer, albeit of a small thickness, the thermal insulation properties of the brick increase slightly.



a b

Fig. 3. **Microstructure of a ceramic brick sample with engobe No. 2 after firing at 950 ºC:**
a – in section, b – dense engobed surface

## Conclusions

Building brick samples with an open porosity of 29.1% and a thermal conductivity coefficient of 0.28 $W\cdot(m\cdot K)^{-1}$ were obtained by introducing a carbon-containing combustible additive into the ceramic mass. It allows them to be used in energy-efficient construction, but the porous structure limits the use of these products in conditions of temperature extremes.

Protective engobe coatings make it possible to reduce the water absorption rate of the outer layer of the brick from 14.8% to 3.2%, which leads to 3-4 times increase in frost resistance.

Engobe is recommended for building products with a firing temperature of 950 ºC, which contains, by wt.%: fire-resistant clay – 65; quartz sand – 10, broken glass – 25. Engobe in the form of a slip with fluidity of 15 seconds is applied to the surface of a ceramic brick, dried and fired together with the product. After firing, the ceramic coating is well conformed with the ceramic base according to indicator shrinking processes and thermal expansion.

## Author contributions

Conceptualization, conducting researches, writing the original text, Kh. O.; study of the microstructure of the samples, I. V.; conducting research, editing, S. N.; methodology, data visualization F. G. All authors have read and agreed to the published version of the manuscript.

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