The development of a modified composition of ceramic mass for the production of bricks

V F Torosyan, E S Torosyan, V A Yakutova and V K Antyufeev
Yurga Institute of Technology, Tomsk Polytechnic University, 26, Leningradskaya st., Yurga, 652055, Russia

E-mail: torosjaneno@mail.ru

Abstract. The need to improve the technical level of production of construction materials, their product range, to improve product quality and reduce its cost requires the expansion of the raw material base, the use of resource and energy saving technology and design solutions. To implement all these it is necessary to conduct a more detailed study of the properties of ceramic materials and to investigate the behavior-modifying components of their formulations. This paper presents the development of the composition of ceramic mass for the production of bricks, a modified silicon-waste production of ferrosilicon.

1. Introduction
Foreign and Russian authors have significant experience in manufacturing multi-purpose construction materials with the use of micro-silica. The chemical composition of the latter, peculiarities of its structure, composition and amount can influence colloid-chemical, structural and mechanical processes, a character and kinetics of hydration of different materials in conditions of thermal impact [1, 2].

Characteristics of ceramic materials are determined by properties of raw materials and a technology of their manufacturing. Clay materials are the basic ones for ceramic brick manufacturing. This issue viewed both as fundamental and applied one have attracted attention of many researchers (A I Avgustinik, A S Berkman, F D Ovcharenko, M I Rogovoy et al).

Sweden scientists have developed a method of manufacturing of high-strength cellular ceramic materials from clay and gas-treated dust, dust produced by electrofilters, utilized in manufacturing of ferrosilicium and silicochromium. A charge with this chemical composition can be used for manufacturing of construction elements, whereas a raw product is reasonably to be covered into foil keeping gas-reducers [3]. There are papers where in the authors apply micro-silica as a binding component in masses for manufacturing of ash-ceramic products [4]. A combination of micro-silica with titanium dioxide when sintering causes formation of a heat-resistant material due to the reaction of amorphous silica with titanium dioxide \( \text{TiO}_2 + \text{SiO}_2 = \text{TiSiO}_3 \) [5].

Refractoriness of micro-silica determines its application in manufacturing of fire-proof and heat-resistant materials which are highly resistant to thermal shock and do not shrink much [6, 7].

A mineralizing effect of micro-silica while crystal and amorphous phases are formed in heat-resistant and fire-proof materials makes it possible to control a special chemical composition in products of their synthesis [8].

Microsilica as a modifying component in fired ceramics has an effect on physical and chemical processes of structural-and-phase construction. A lot of scientists carried out fundamental and applied
research into issues of phase transformations of clay raw materials used in manufacturing of ceramics (A I Avgustinik, A S Berkman, F D Ovcharenko, M I Rogovoy, L N Tazki et al). The description of the main characteristics of these processes is given below.

While finish drying at temperature of 80-130 °C physical and mechanical bound water vaporizes and a product is uniformly warmed up. During the first phase of firing, source minerals are destroyed because of dehydration, dissociation and amorphization of components.

Within the temperature limit of 200…600 °C organic admixtures and combustible additives are burnt out at temperature 500…600 °C physical and chemical bound water escapes from clay and other minerals; therefore, clay loses its plasticity, mass is reduced, a crystal lattice of the mineral is damaged, mechanical strength fails and products shrink. Dehydration of kaolin proceeds with formation of metakaolinite \( \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} \rightarrow \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + 2\text{H}_2\text{O} \).

After shrinkage, the clay becomes more concentrated because low-melt compounds fill the pores among high-heat particles and due to further compaction – firing.

Within the temperature limit of 500…700 °C, minerals containing ferrum, sulphides and sulphates are dissociated and the remaining 700…900 °C – those containing carbonates. Burning out of organic admixtures and dissociation of minerals, emitting volatile gases (\( \text{CO}_2, \text{SO}_2 \) et al), are to be finished before a crock is fired in order to prevent its cracking and swelling. At temperature of 700 °C and above, alcalis in the clay interact with other components constituting a melt, where (as A. I. Avgustinik stated) hard particles are attracted, minerals dissolve and new thermo-dynamically stable crystal phases form. Further heating of the material above 700 °C results in decomposition of metakaolinite into free oxides \( \text{Al}_2\text{O}_3, 2\text{SiO}_2 \), sillimanite formation \( \text{Al}_2\text{O}_3 \cdot \text{SiO}_2 \) and \( \text{SiO}_2 \), and at temperature of 950…1150 °C and above – mullite \( (3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2) \). Mullite is the most stable compound, strengthening products, impacting them and making them thermally resistant.

Clays and loams are often sensitive to drying, after firing they have low compression and flexural and frost resistance. All these disadvantages make it impossible to use the materials without improving additives. Moreover, as many researchers emphasized [3-5], it is rather difficult to find an additive which can eliminate complex technological problems. Therefore, charge materials necessitate correction by introducing several additives or mineralizers into ceramic masses, consequently the price of finished products rises and materials consumption of production increases. In addition, the growing number of components that ceramic mass consists of causes further difficulties, which can have a negative effect on the quality of baked products.

This paper is aimed at the analysis of micro-silica behavior viewed as a modifying component in fired ceramic materials based on low-grade clays. The use of the latter for manufacturing ceramic bricks can improve their physical, mechanical and operational characteristics.

2. An experimental part
Stiff clay of Bolotnoye deposit and micro-silica produced by the public limited company ‘Yurga ferroallloys’ were applied for research. Loam of Bolotnoye deposit is a low-melt clay raw material with high concentration of \( \text{Fe}_2\text{O}_3 = 5.23 \% \) and a semi-acid loam \( (\text{Al}_2\text{O}_3 \text{calc} = 14.22 \%) \). Its technological characteristics are listed in table 1.

| Loam | Fraction 0.5 mm, % | Fraction maximum 0.5 | Chemical composition |
|------|-------------------|---------------------|---------------------|
| Bolotnoye deposit, explored by Rosgeolnerudrazvedka in 1969 | Smaller than 0.005 mm – 17.3…20.6%; 0.005...0.05 mm – 61.1…63.8 %; 0.05…1 mm – 15.5…21.6 % | SiO\(_2\) – 61.8…64.2; Al\(_2\)O\(_3\) – 12.0…14.66; Fe\(_2\)O\(_3\) – 5.23…5.84; CaO – 3.09…5.21; MgO – 1.31…2.24; P\(_2\)O\(_5\) – 17.27…20.5; others – 6.91…9.57 |
Microsilica is condensed silicate dust, which is a sideline product of crystal silicon or silicon alloys manufacturing. Some silicon monoxide SiO becomes gaseous and, being exposed to oxidation and condensation, it forms an extraordinary small product shaped like ball-shaped particles containing a lot of amorphous silica. Microsilica as opposed to conventional high-silica raw materials, for example, quartz sand, is a light fine-dispersed material containing small spheres of amorphous silica with 0.01 to 0.7 micron particles.

Despite differences in chemical composition, colour and carbon concentration, fine dust particles, being the wastes of silicon and ferrosilicium manufacturing, have some common characteristics:

- They are condensates of silicon vapors (silicon monoxide).
- They mainly consist of globules with the average size of 0.1…0.2 microns (100 times smaller than cement particles).
- They are amorphous.
- Concentration of SiO$_2$ is high (84…98 %).
- They are difficult to store and transport.

The dust colour varies from light-grey to wholly black and depends generally on carbon concentration and less on ferrum. The density is 2200…2300 kg/m$^3$. This coefficient can exceed the above-mentioned value in production of other silicon containing alloys, for example, ferrochrome silicon or calcium-silicon.

Melting temperature of various silicon containing alloys is different. In production of crystal silicon the temperature is higher than in melting of ferrosilicium with silicon concentration of 40…50%. Hence, the chemical and granulometric composition is different. It is significant that particles of the stopped dust are not separate micro-globules but conglomerates of finest particles of 0.01…2 microns in diameter.

Exceptionally high dispersibility of micro-silica and its structure are the reasons of its low apparent density (150…250 kg/m$^3$). These characteristics cause the difficulties in its application and transportation. Table 2 provides the information on the chemical composition and physical-mechanical characteristics of micro-silica, produced by the public limited company “Yurga ferroalloys”.

**Table 2.** Physical and mechanical characteristics of micro-silica, produced by the public limited company ‘Yurga ferroalloys’.

| Raw material | Chemical composition | Concentration in mass % | Density, kg/m$^3$ | Apparent density, kg/m$^3$ | Porosity of material layer, % | Specific surface, s m$^2$/g | Natural moisture, mass % |
|--------------|----------------------|--------------------------|------------------|-----------------------------|-------------------------------|-----------------------------|--------------------------|
| Micro-silica | SiO$_2$              | 94.2                     |                  |                             |                               |                             |                          |
|              | Al$_2$O$_3$          | 0.47                     |                  |                             |                               |                             |                          |
|              | Fe$_2$O$_3$          | 0.6                      |                  |                             |                               |                             |                          |
|              | CaO                  | 0.36                     | 2180             | 150                         | 300                           | 82                          | 79                       |
|              | MgO                  | 0.3                      |                  |                             |                               |                             |                          |
|              | K$_2$O               | 0.69                     |                  |                             |                               |                             |                          |
|              | Δm                   | 3.43                     |                  |                             |                               |                             |                          |

Experimental chemical compositions of ceramic mass were prepared in laboratory conditions. Raw material preparation included the following phases of treatment:

- drying of raw materials, grinding in a dry-grinding mill, sieving;
- samples in the form of a cube of 30x30x30 were made by the method of soft forming;
- when the soft forming mixture was moistened up to the necessary forming moisture, then soured during 24 hours till adsorbed hydrate films formed completely;
samples were preliminary dried in natural conditions during 7 days at ambient temperature of 23…25 °С;
- baked in a muffle furnace at temperature of 1000-1050°C for an hour.

Low-melt loams with high concentration of Fe₂O₃ = 5.23 % are used as a raw material at Yurga brick factory for brick manufacturing. These loams are acid or semi-acid ones (Al₂O₃calc = 14.22 %). Their technological characteristics are similar to those of Bolotnoye deposit. A raw-brick formed at Yurga brick factory was taken for comparison with finished products divided into cubes of 30×30×30 and burnt with experimental samples in the muffle furnace at temperature of 1000-1050 °C for an hour. Sulfate soap was used to increase cohesion between particles of masses. All samples were subjected to physical and mechanical tests. Table 3 shows the chemical composition of ceramics based on a low-melt clay raw material, containing micro-silica.

Table 3. Ceramics chemical composition.

| Components      | Amount, mass % |
|-----------------|----------------|
|                 | 1  | 2  | 3  | 4  | 5  | 6  |
| Clay            | 89 | 87 | 84 | 80 | 75 | 70 |
| Micro-silica    | 10 | 12 | 15 | 19 | 24 | 29 |
| Sulfate soap    | 1  | 1  | 1  | 1  | 1  | 1  |

3. Results discussion

Water absorption is one of negative characteristics of construction materials. It reduces the strength of bricks, they become more heat-conducting, and a mould can develop on their surface. A moistened brick is subjected to destruction in winter because of constantly repeated cycle freezing – defrosting. Micro-silica in ceramic masses promotes water extraction when heating and an amorphized component of micro-silica crystallizes at high temperatures. In general, water absorption of bricks is reduced and their strength increases. Table 4 provides the information on physical and mechanical properties of samples made on the basis of materials, listed in Table 3.

Table 4. Physical-mechanical properties of samples.

| Property                     | Chemical composition |
|------------------------------|----------------------|
|                              | 1  | 2  | 3  | 4  | 5  | 6  |
| Compression strength, MPa    | 33.5 | 32.7 | 30.2 | 28.3 | 23.5 | 18.7 |
| Mean density, kg/m³          | 1710 | 1660 | 1620 | 1580 | 1510 | 1480 |
| Frost resistance, cycles     | min60 | min 60 | min 60 | min 40 | min 40 | min 35 |
| Water adsorption, mass %     | 18.5 | 19.2 | 20.0 | 22.4 | 26.5 | 28.3 |
| Temperature of burning, °C   | 1050 | 1050 | 1050 | 1050 | 1050 | 1050 |

4. Conclusions

1. Micro-silica in ceramic masses is 10…15 %; water adsorption is 18.5…20.0%; 19…29% of water adsorption changes from 22.4% to 28.3%.
2. The number of frost resistance cycles of samples is increased, starting with 35 cycles under conditions of 29% of micro-silica up to more than 60 cycles under conditions of 10% of micro-silica.
3. Fracture strength of samples improves when samples are compressed up to 33.5 MPa.
4. Volumetric coloring of bricks containing micro-silica as a modifying component can be used in manufacturing of high quality facing ceramic bricks. Micro-silica has an advantage in this
process because there is no need to grind it as opposed to additives utilized in traditional methods of brick coloring.

References
[1] Kaprielov S S, Sheinfeld A V, Gazizulin V M and Voronov Yu N 1992 J. Steel 5 pp 83–85
[2] Milovanova R G, Semyonova V E and Chupina A I Building mortar Certificate of authorship № 637359 C04B13/06
[3] Mironyuk I F, Chuyko A A, Khoma M I, Kisly P S, Khaber N V and Ostafiychuk V K Ceramic material Certificate of authorship № 962262 C04B35/14 35/80
[4] Fredrik Wilhelm Anton, Dr. Lidingo Method for producing fired expanded clay Products 19.03.71 (Sweden) DE2213231 C3 C04 B21/04 Short
[5] Pak N V, Artyomov L N and Makarov L M 1990 J. Energy construction 3 p 38
[6] Kurz F W and Rudmark H 1978 Method of manufacturing porous ceramic products by reacting flue gas dust and filter dust with clays of the like, such as expanded clay Patent 4071369
[7] Shpirko N V and Chumak L I 1989 Heat-insulating materials on the basis of pearlite with the use of industrial wastes Proceedings of higher educational institutions Building and architecture (Tambov: Tambov State Technical University) pp. 62–66
[8] Onasenko Yu A and Peschanskaya V V 2013 J. Technol. Auditand Product. Reserv. Ⅱ 25–28
[9] Lokhova N A, Makarova I A and Patramanskaya S V 2002 Fired materials on the basis of micro-silica (Bratsk State Technical University) p 163
[10] Torosyan V F and Torosyan E S 2014 J. App. Mechanics and Materials 682 480–484