Application of Magnesite Waste in Manufacturing High-strength Ceramics

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Abstract. Factories of the Republic of Tatarstan produce about 3 million tons of industrial waste per year. The solution of problem of utilization and processing of industrial waste is to reuse them in the production of ceramics. Currently, however, not all the waste is investigated and can be used repeatedly in industry. The article provides an analysis of magnesite waste, reveals its qualitative and quantitative composition. The effect of the additives from this waste is studied on the following types of clay: fusible clay from Alekseevsky field, refractory clay of Novoorsky field, clay of Salmanovsky field with high carbonate content. In the study we used the following methods: X-ray phase analysis (diffractometer XRD-7000S (Shimadzu, Japan), diffractometer D2 Phaser (Bruker, Germany)), electron microscopy (EVO-50XVP microscope), measurement of basic physical and mechanical properties (press SGP-500 CIM 4 SKB, Stroypribor, Russia, and others). Tests were conducted under identical conditions with the addition of pure magnesium oxide; however, the positive results were not found. Modification of Salmanovsky and Novoorsky clay fields with magensite waste also did not lead to the improvement of the characteristics of the samples. On the contrary, in the ceramic mass compositions based on clay of Alekseevsky field we established the feasibility of using magnesite waste in the range from 2 to 5 % by weight at a burning temperature of 1150 °C for the production of high-strength ceramics.

Keywords: magnesite, magnesium oxide, ceramics, clay, modification, low melting eutectics
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Introduction

According to the concept of long-term development of the Russian Federation for the period until 2020 (Decree of the Government of the Russian Federation of 17.11.2008 N 1662-r) one of the main areas of environmental security is environment of the production. Every year more and more priority is given to reduction of energy and materials through the introduction of best available technologies. On the other hand, it is a fairly complex and lengthy process.

As is known, the Republic of Tatarstan produces about 3 million tons of industrial waste per year. However, the present system of waste management in the republic is based mainly at their burial in landfills and storage in special facilities, which negatively affects the quality of life of the population (Concept of waste treatment in Tatarstan Republic, 2011). The best solution may be to re-use industrial waste in the factories. This will not only resolve the environmental issues that arise in the production process, but also save natural resources.

The problem of disposing industrial waste is enlightened in a considerable amount of works by Russian and foreign scientists (Bender, 2004; Rumi et al., 2015; Khomenko et al., 2014; Salakhov et al., 2014; Sukharnikova et al., 2016; Muller et al., 2009).

The authors considered the task by the use of magnesite waste as an additive for modifying clays, provided by the plant named after A.M. Gorkiy, for the purpose of the waste products implementation in the production cycle of manufacturing ceramics.

Experimental part

Annually on ship-building plant named after A.M. Gorkiy over 1 thousand tons of waste magnesite is produced, that is a serious problem for its disposal.

This waste is produced from grinding and dry regeneration of molds for the production of castings made of metal alloys; periclase (magnesite powder) is used as a molding composition for making molds from Satkinksy field deposits of PPLF brand with a mass fraction of magnesium oxide of not less than 89 %. The chemical composition of magnesite waste is as follows:

- mass proportion of magnesium oxide is not less than 88 %;
- mass proportion of calcium oxide is not more than 4.5 %;
- mass proportion of silica is not more than 5.0 %;
- other impurities (iron oxides and so on) are not more than 2.5 %.

Industrial waste consists of fine-crystalline grains, 25 % of which pass through the mesh No. 0063 (Specifications number TLT19 on magnesite waste (periclase powder of brands PPLF-89, PPLF-91)).

In the scanning electron microscope “EUO-50” the authors investigated the composition and structure of the waste. The structure of the waste is heterogeneous (Fig. 1-2), fragments are identified of different particle size and morphology. Loose and unstable formations, which are considered to be multicomponent, join together to form conglomerates. The particle size is of about two microns. In some places, formed structures are correlated with fractal (Fig. 2).

According to X-ray spectra elemental composition of magnesite waste varies at different plots. Some fragments differ by a substantial content of sodium metal alkali (Fig. 3). On the other fragments there is a high content of calcium (Fig. 4).

On the white areas of SEM images a complex elemental composition is identified (Fig. 5-6).

For the modification of waste different types of clay were selected: fusible Alekseevsky, refractory Novoorsky, and Salmanovsky clay with a high content of carbonates.

The clay was subjected to pre-dispersing in a dry form before passing through a sieve with a 0.5 mm cell. Then the clay was modified with waste magnesite with mass fractions of 2 %, 5 %, 10 % of the total weight; raw mixes were composed, which were averaged thoroughly in a dry form. After mixing with water operating humidity of initial mass was 10 %. Raw material was prepared by compression (15 MPa) molding.

Waste composition, %	0	2	5	10
Density, g/cm^3	2,2	2,3	2,3	2,2
Water adsorption, %	1,3	1,0	1,5	1,8
Line heat setting, %	5,3	5,7	5,3	5,3
Strength, MPa	139	155	148	103

Table 1. Characteristics of the samples from Alekseevsky clay modified by magnesite waste at firing temperature = 1150 °C.

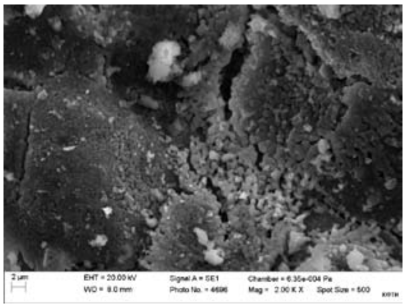

Fig. 1. SEM image of magnesite waste.

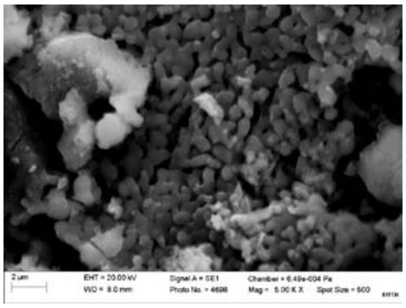

Fig. 2. SEM image of magnesite waste.

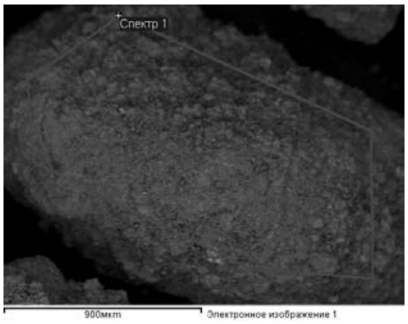

Fig. 3. SEM image of magnesite waste. The elemental composition of the X-ray part of the spectrum, designated as "Spectrum 1": O – 58, Na – 5, Mg – 29, Si – 6, Ca-1, of Fe – 1 %.

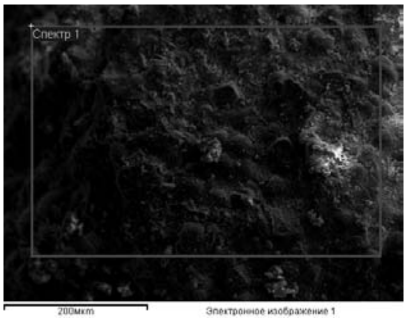

Fig. 4. SEM image of magnesite waste. The elemental composition of the X-ray part of the spectrum, designated as "Spectrum 1": O – 60, Mg – 27, Si – 8, Ca – 5, Fe – 1 %.

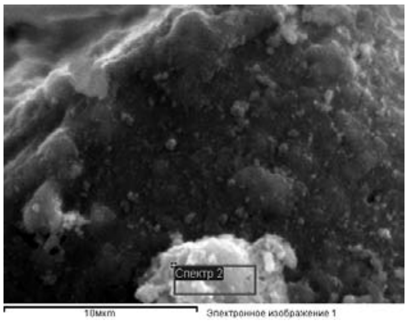

Fig. 5. SEM image of magnesite waste. The elemental composition of the X – ray part of the spectrum, designated as "Spectrum 2": O – 63, Na – 2, Mg – 23, Si – 11, Ca – 1 %.

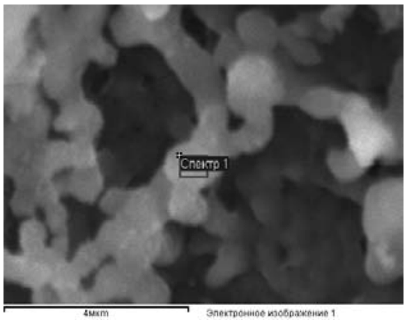

Fig. 6. SEM image of magnesite waste. The elemental composition of the X – ray part of the spectrum, designated as "Spectrum 1": O – 64, Mg – 21, Si – 11, Ca – 3 %.

Molded samples were kept under natural conditions for 1 day, and then firing was performed at 1050 °C-1150 °C in increment of 50 °C in a muffle furnace LOIP LF-7/13, the firing time – 4 h.

To compare the results for each sample "back-up" was prepared, which instead of the magnesite waste contained the same amount of pure magnesium oxide.

Polymineral fusible clay from Alekseevsky field during firing up to 1000 °C does not form new mineral phases (Fig. 7), with the exception of hematite, which share is less than 1 %.

Determination of the main characteristics of synthesized at 1050 °C materials showed that the addition of magnesite waste contributes to negligible change of physical and mechanical properties in comparison with samples of pure clay.

Samples of clay from Alekseevsky field supplemented with 5.2 % magnesite waste, calcined at 1150 °C in the press test showed high strength properties (Table 1). These results can be explained by the presence in the waste composition of a significant proportion of alkali metal oxides that contribute to the formation of low-melting eutectic, creating an environment for active cooperation.

These results are consistent with the nature of destruction. Figure 8 shows that degradation has occurred as a result of crack growth parallel to the axis of the sample, as evidenced by the shape of fragments and a minor amount of dispersed particles formed (Ariskina et al., 2015).

Tests of samples for compressive strength were conducted on press SGP-500 MGK 4 SKV Stroypribor.

Also X-ray diffraction analysis (XRD) was conducted for patterns with a 5 % magnesite waste from the plant named after A.M. Gorkiy, calcined at temperatures of 1100 °C and 1150 °C (Table 2). As a result of XRD, we can

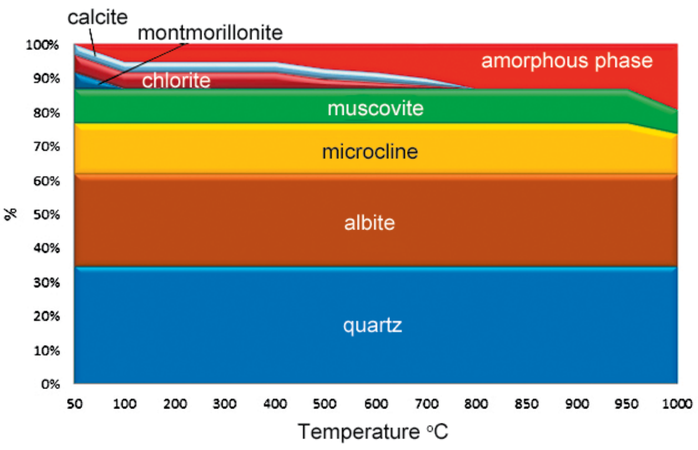

Fig. 7. Changes in the mineral composition of Alekseevsky clay due to a rise in temperature from 50 to 1000 °C.

conclude that with increasing temperature in the ceramic samples restructuring occurred. Only stable mineral phases were formed, increase in the share of amorphous phase was identified.

Fig. 9 shows the structure of the calcined sample with 5 % magnesite waste. Noncommunicating pores of about 15 microns are observed. Grains are in close contact with each other, grain boundaries are "glued" as a result of liquid phase sintering to form a monolithic structure.

Analogous studies on the basis of Novoorsky refractory clay and Salmanovsky clay with a high carbonate content resulted in a deterioration of strength properties. Methods and test conditions were identical.

Burning temperature, °C	Quartz	Albite	Hematite	Periclase	Enstatite	Amorphous phase
1100	44	10	4	3	1	38
1150	43	10	4	2	0	41

Table. 2. Mineralogical composition of the samples with 5 % magnesite waste.

Variation diagram of the phase composition of Salmanovsky clay in the firing process is shown in Fig. 10.

Salmanovsky clay contains 40 % of calcite, which dissociates with increasing temperature to form calcium oxide. In the firing process, calcium silicate is synthesized. Magnesium oxide, which included in the magnesite waste, does not have a 'partner' in the interaction that contributes to poor strength characteristics.

Variation diagram of the phase composition of Novoorsky clay in the firing process is shown in Fig. 11.

In the firing process mullite is synthesized, which requires a silicon oxide. Thus, magnesium oxide does not have a 'partner' to interact. This leads to a decrease in strength characteristics.

Parallel to studies carried out, properties were identified of the fired samples supplemented with pure magnesium oxide. It was established that the clay composition with magnesium oxide leads to a significant deterioration of performance: when its content is increased to 10 %, a sharp decline in strength by 4 times occurs and the water absorption factor increased by 2.5 times. It can be assumed that most of the magnesium oxide remains unreacted, as evidenced by the white blotches all over the sample volume. Positive results were not found.

Fig. 8. The destruction nature of the sample with 5 % magnesite waste. Firing temperature = 1150 ° C. Compressive strength – 148 MP.

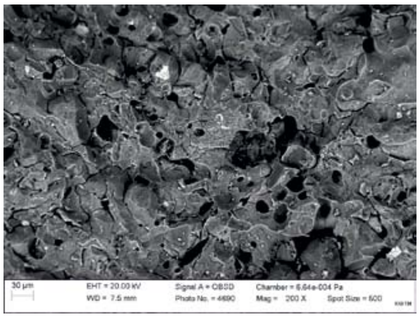

Fig. 9. SEM image of the sample with 5 % magnesite waste. T = 1150 °C.

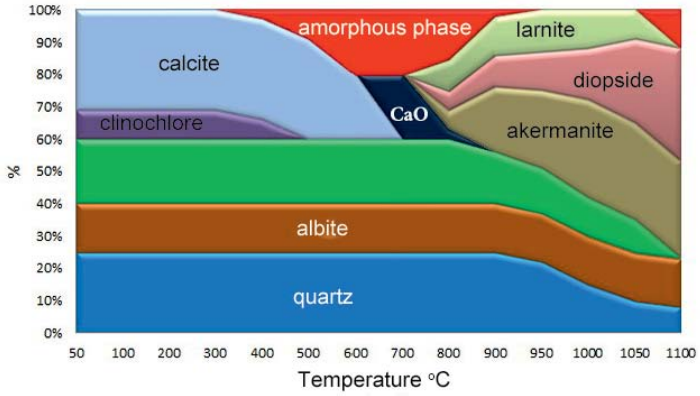

Fig. 10. Changes in the mineral composition of Salmanovsky clay due to a rise in temperature from 50 to 1100 °C.

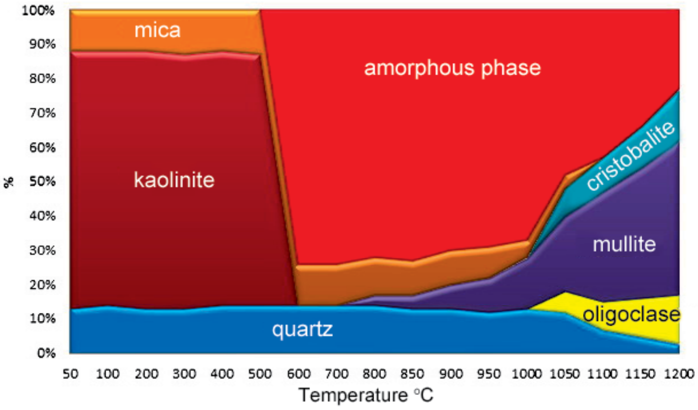

Fig. 11. Changes in the mineral composition of Novoorsky clay due to a rise in temperature from 50 to 1100 °C.

Conclusions

During these studies we established the feasibility of using waste additives of magnesia in the range of 2-5 % by weight of ceramic material compositions from Alekseevsky clay for manufacturing high-strength ceramics, in combination with fusible clay materials ensures completeness and uniformity of sintering at 1150 ° C.

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