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Prospects and features of microwave sintering in the technology of building ceramics

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Abstract. The results of firing samples from natural aluminum silicates in a microwave oven are considered. The possibility of sintering aluminum silicate raw materials, traditional for the production of building ceramics, is established. The phase compositions of the calcined materials are determined. The influence on the sintering process of the mineral structure of clay raw materials, the composition of the diatomite and the addition of NaCl salt was established. Samples from the mass with the addition of NaCl show high quality and increased strength of the samples. Samples from masses based on brick clay with NaCl salt, annealed in the microwave electromagnetic field, did not show a large increase in strength. For all mass compositions with NaCl, a decrease in the energy of the endothermic process was revealed in the temperature range 400–600 °C. In all mass compositions from clays with NaCl and diatomite with salt and marl, the presence of an aluminum silicate phase with a size of less than 30 nanometers was found. Masses with such a phase structure showed increased strength of the samples.

Keywords: microwave roasting, clay, diatomite, NaCl, nanophase.

1 Introduction

In connection with the expansion of the range of construction objects in recent years, a need has arisen for building materials with special, often extreme properties. At the same time, the technology for obtaining materials must meet modern environmental, energy and resource-saving requirements. For the technology of firing material, such as ceramics, there is a need to search for alternative solutions, the most promising of which is the synthesis of the material in a high-frequency electromagnetic field (microwave).

In ceramic technology, sintering is the main stage of heat treatment, during which the final structure of the material is formed. The ability to obtain materials with extreme properties is associated with the formation of a nanoscale structure of the material. In ceramics, this is the level of passage of the main processes between phases during sintering, which is realized in a composition with nanoscale dispersion of components, for example, in the preparation of mass from a slip. However, in the process of traditional sintering technology, with prolonged heating and aging of samples at a final firing temperature, the recrystallization processes in the composition prevent the formation of a nanoscale material structure.

The increased interest in nanostructured ceramic materials is associated with extraordinary mechanical and physical properties that begin to appear when the size of the structural elements becomes less than the critical value defined for each composition [1–5].

Studies were carried out in the direction of increasing the strength and fracture toughness of ceramics by reducing the size of the grain and defective structure of the material [6–9]. The dependence of the material properties on the structure and dispersion of the introduced fillers, the conditions for the formation of the structure of the material is studied [10–13]. All studies were conducted on compositions with a limited number of components. Clay-based compositions used in the technology of building ceramics are multicomponent mixtures of various chemical and mineralogical compositions. This imposes its features on the passage of all phase formation processes to the final stage of formation of the material structure.



Recent studies have shown that sintering with a high heating rate in an electric field allows the production of microcrystalline material with special properties, such as unusually high density and strength of the material [14, 15]. Especially important are material properties such as high elasticity and ductility [16], which allow traditionally brittle ceramics not to break when a crack appears.

Researches in the field of high-speed sintering in the electromagnetic field showed the special mechanism of course of high-temperature processes and receiving as a result of the unusual properties of material caused by its microcrystalline structure. The process of heating ceramics in a microwave electromagnetic field was investigated on various compositions. When processing an aluminum silicate ceramic composition in a microwave field, its rapid heating in a short time was observed, during which a homogeneous composition and good diffusion of [17] components were obtained.

Upon activation of the raw clay compositions in the microwave field, it was found that in the fine fraction of clay suspensions, structures begin to form which, with subsequent convective heating, contribute to the formation of a composition including nanosized formations [18, 19].

In work [20], in the process of studying the microwave synthesis of nanoporous material, differences were found in the crystal structures obtained under the influence of the microwave field and in the process of convective heating.

Sintering of clay compositions upon heating begins after dehydration and decomposition of the initial components of the colloidal component of the charge and proceeds in the interphase, intergranular region. An analysis of microwave heating of dielectrics, including components of clay minerals, shows the beginning of the process in the interfacial region, which, due to diffusion, covers the entire volume of the material [21].

Sintering of clay compositions, from which mainly all building ceramics are obtained, is complicated by their multicomponent composition. To obtain high-strength building ceramics and refractories, refractory clays, kaolins and hydromica, consisting of clay minerals, are used. Therefore, the study of the features of the technology for firing clay compositions in a microwave electromagnetic field is of great importance for the development of the fundamental principles of the technology for producing ceramics in general and creates prospects for the development of technology for producing new types of building ceramics.

The purpose of the experimental work was to study the sintering features of compositions traditional for building ceramics in the microwave electromagnetic field.

2 Materials and methods

For sintering in the microwave field, natural aluminum silicates of the composition in mass % were selected: refractory (60 % kaolinite, 5 % mica) and bentonite clays (60 % montmorillonite, 5 % mica) with a content of basic oxides SiO_2 (53.6–56.4) % and Al_2O_3 (20.0–27.9) %; low-melting hydromica brick clay of chemical composition (wt. %): SiO_2 , 68.1; $\text{Al}_2\text{O}_3+\text{TiO}_2$, 12.6; Fe_2O_3 , 6.4; $\text{CaO}+\text{MgO}$, 3.3; $\text{Na}_2\text{O}+\text{K}_2\text{O}$, 3.2; diatomaceous earth (98.7% SiO_2). NaCl salt (extra following, Russian Federation State Standard GOST 51574-20000) was chosen as the fluxing additive because of its low melting point and the ability to form low-melting mixtures with [22] silicates. As a carbonate additive – marl (wt. %) (SiO_2 , 33.2; Al_2O_3 , 11.4; $\text{CaCO}_3 + \text{MgCO}_3$, 26.1; Fe_2O_3 , 3.6; $\text{Na}_2\text{O} + \text{K}_2\text{O}$, 3.6).

Samples were prepared using plastic technology. Milled in a dry state and sieved through a No. 1 sieve, the clay was moistened with distilled water. The NaCl salt was dissolved in water before wetting the composition.

Sintering of the samples was carried out in a microwave oven of the brand Samsung M 1711 NR (2.45 GHz, 800 W), equipped inside with a muffle from mullite-siliceous plates, in which the samples were installed. Temperature in the muffle was controlled by the thermocouple with the covering protected from radiation. A thermocouple wire connection is installed next to the samples.

Heat treatment was carried out to 1000 °C with exposure at a maximum temperature of 5 minutes. X-ray phase analysis of the calcined samples was carried out on a Shimadzu XRD 6000 diffractometer in CuK_α radiation (PDF 4+ base, POWDERCELL 2.4 full-profile analysis program), thermal analysis on a synchronous TG-DTA / DK analyzer (QMS 403 C quadrupole mass spectrometer).

3 Results

A study of the sintering process of the samples showed a different reaction of the compositions to heating in a microwave oven. Samples from bentonite clay and diatomite were sintered without destruction, brick clays had small cracks. Refractory clay samples collapsed. Diatomite samples had a loose structure and low strength. The appearance of cracks and fracture of the samples is observed in the temperature range 400–600 °C.

The structures were investigated by synchronous thermal analysis. Fragments of the experimental curves in the temperature range 300–750 °C, in which the destruction of the samples is observed and in which the phase transformations begin, are shown in Figure 1.

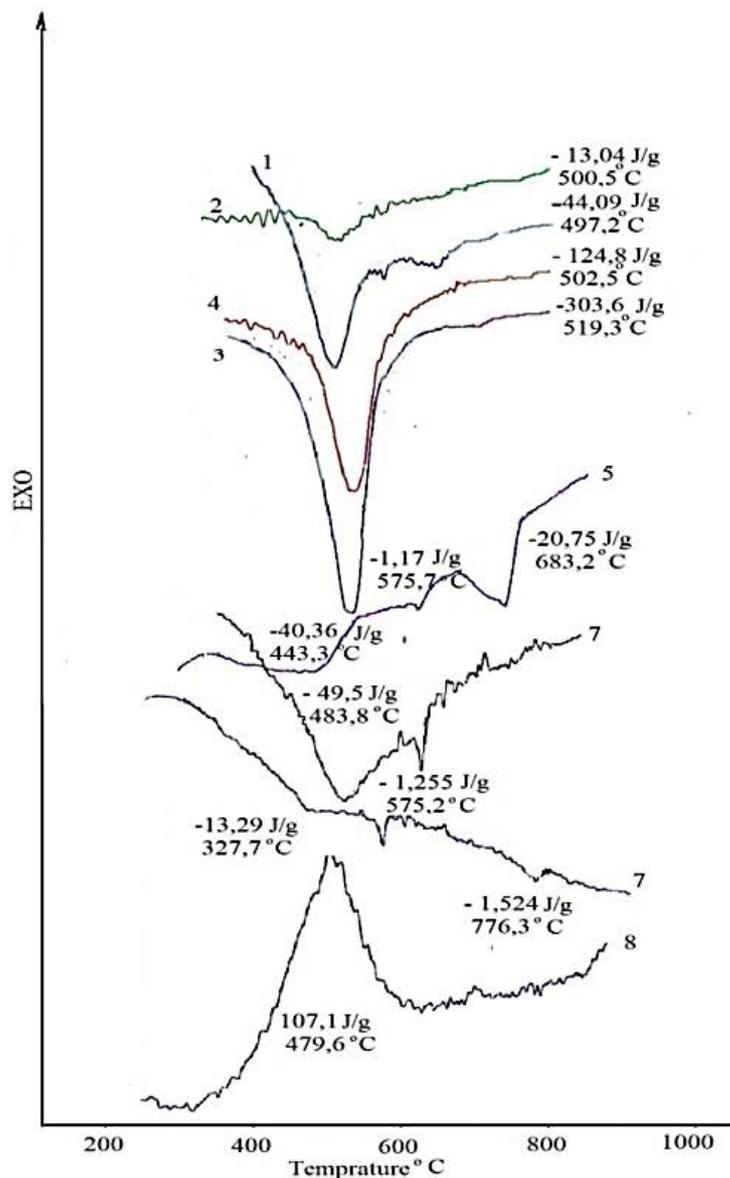


Figure 1. Thermograms (DTA): 1 – bentonite clay, 2 – bentonite clay and NaCl, 3 – refractory clay, 4 – refractory clay and NaCl, 5 – clay and NaCl, 6 – diatomite with NaCl and marl, 7 diatomites with NaCl, 8 – diatomite.

It is seen that when salt is added, the energy component of the endothermic process in the temperature range 400–600 °C for all introduced products is significantly reduced. For diatomite with NaCl, instead of the exothermic process of the transition of amorphous silicon dioxide to the crystalline phase at 479.6 °C, an endothermic process is visible at 327.7 °C and weak at 776.3 °C. These two endothermic effects are significantly enhanced when marl is added to the mass. The magnitude of the endothermic effect in terms of parameters is close to the endothermic effect of clay.

The results of X-ray phase analysis of the fired ceramic masses are shown in Table 1.

Table 1. Phase composition of the samples after firing.

Composition	Weight percent					Crystallite size, nm
	SiO ₂	Al ₂ SiO ₅	Al ₂ O ₃	γ-Fe ₂ O ₃	FSP	
Bentonite	63.6	28.	-	8.1	-	96.88 69.32 >100
Bentonite + NaCl	1.6	38.5	-	-	60	>100 22.4 30-35
Refractory	46.0	-	54	-	-	54.8 9.44
Refractory + NaCl	63.4	17.1	19.5	-	traces	139 29.6 >200 34
Diatomite + NaCl	56.2	16.9	-	-	26.9	>200 >200 >200
Diatomite + NaCl + marl	36.7	8.7	-	0.8	5.4	92 24.9 >200 16.9
Gley and NaCl	45.3	27.1	-	3.4	24.8	>200 29.7 >200 28.8

FSP Sodium Aluminum Silicate

The results of X-ray phase analysis of samples with NaCl showed the presence in the compositions of the nanoscale phase of aluminum silicate with sizes up to 30 nm, sodium aluminum silicate of various compositions and an amorphous phase. The presence of a modifier (low-melting NaCl salt) in the aluminum silicate composition promotes the formation of high-temperature phases with lower energy costs. This contributes to the sintering of the samples without destruction.

All samples from the compositions modified with NaCl were burnt without cracks and showed a significant increase in strength (Figure 2).

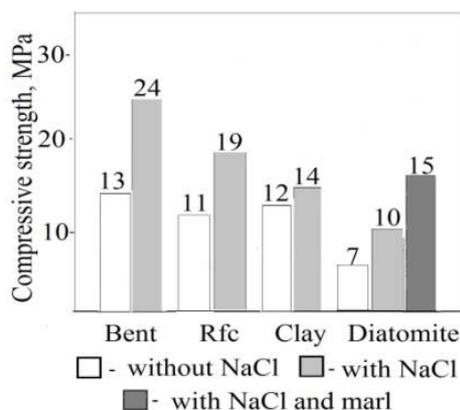


Figure2. Durability on compression of the samples burned at 1000°C from: bentonite clay – Bent, refractory clay - Rfc, clay - Clay, Diatomite.

The greatest increase in strength was shown by samples based on bentonite clay with NaCl, diatomite with NaCl and marl. Brick clay based compositions have shown close strength levels, probably due to the multicomponent composition of clay.

4 Discussion

The performed studies showed that:

- it is possible to sinter in the microwave field compositions based on traditional aluminum silicate raw materials used to obtain building ceramic products;
- sintering of samples from natural aluminum silicate raw materials in the microwave field allows to obtain high material strength in a shorter time compared to convective heating;
- the technology of sintering of material in the field of the microwave oven depends on mineral structure of raw materials of which it is made;
- it is possible to obtain samples without defects during firing in the microwave field, if a fusible mineralizer (in our case, NaCl salt) is introduced into the mixture from natural aluminum silicate raw materials;
- as a part of all studied compositions calcined in the microwave field, nanophases of aluminum silicate, the main component of the phase composition of building ceramics, were found.

The results obtained in the work on sintering in the microwave field of multicomponent natural aluminum silicates coincided with the previously described conclusions on sintering in the electromagnetic field of low-component mixtures. Increasing the strength of the material is associated with the formation of a special structure of the material, the presence of nanoscale phases.

A study of the sintering process of natural aluminum silicates in a microwave electromagnetic field is necessary to develop an energy-efficient rational technology for the production of ceramic material. Obtaining new characteristics of materials based on aluminum silicates using energy-saving, environmentally friendly technology opens up prospects for producing ceramics with special properties and expanding the range of products for building ceramics.

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